

## ***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.***

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

1. Abstract: Lines 2-9 I think this information can be abbreviated Line 15: : : and CSP? Complete the sentence. \*\* We agree and will edit here. The sentence 'The accumulation of gel particles in the SML and their potential implications for gas exchange and emission of primary organic aerosols have generated considerable research interest in recent years' will be deleted. The description on CSP will be added as well as the

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information on PSD in the bulk water.

2. Introduction: I think it is too long and repetitive. Maybe the intro could be abbreviated and reorganized as follows: (1) introduce the SML and its properties. (2) introduce gels, their PSD, their biochemical relevance, and their accumulation and role in the SML (3) role of wind speed in SML formation and in gel dynamics, particularly in PSD \*\* We agree and will edit here according to the referee's suggestion.

Page 3 line 6-8 These sentences are repeating the same information. \*\* The repeating information will be deleted.

Page 3 line 9: Start new line, since you talk about something different \*\* Will be done.

3. M&M: Page 7 lines 7-9, how long did it take from sampling to start the experiment? \*\*It took 41 days from sampling to start the experiment. The time of collection and starting experiment will be added: 'Effects of different wind speeds on the size distribution of organic gel particles in the SML were studied during the Aeolotron experiment from 3rd to 28th November 2014. In total 22000 L of North Atlantic seawater were collected on the 22. 09. 2014 by the research vessel FS Poseidon, including ~14000 L of high salinity water 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.'

Page 9 lines 22-23 I do not think the calculation of TEP-C is necessary in this study that does not focus on carbon fluxes \*\*We agree that this part can be removed.

Page 10 line 17 include space between 'distribution' and 'after' \*\* Will be adopted

4. Results: Page 11, lines 3-10 include average changes in TEP in the SML \*\*We will add the average changes in TEP in the SML.

Figure 2. Is this the average of the different wind speed conditions? Clarify. Include SD bars. I would use the same symbols for the same parameters; e.g., if columns are for total area (as they are in SML and bulk CSP), then use also columns for TEP total area. Anyway, I do not think it is necessary to show the TEP-C; as your paper is not

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focused on these measurements. Include panel letters ABCD. \*\*The average of the different wind speed condition will be clarified in the revised version, and the SD bars will be added. The style of Figure 2 can be revised according to the suggestion, using the same symbols for the same parameters. TEP-C will be deleted.

Figure 3 and 4 say if this is SML or bulk water. \*\* Figure 3 and 4 refer to the SML. This information will be added in the revised manuscript and figure captions.

Figure 6. Day 22 panel: Use the same color and symbol code as in Figure 5 and in the rest of panels. \*\*Will be adopted.

Page 11 lines 11-16. Here include average changes in CSP in bulk water \*\*The information on average changes in CSP in bulk water will be added: 'Similar to TEP in the SML, CSP abundance and total area in the SML declined gradually between day 1 and 12. Here, CSP abundance in the SML decreased from  $186.7 \pm 84.3 \times 10^6 \text{ L}^{-1}$  on day 1 to  $29.5 \pm 16.4 \times 10^6 \text{ L}^{-1}$  on day 12. CSP total area in the SML dropped from an initial  $20.5 \pm 2.7 \times 10^2 \text{ mm}^2 \text{ L}^{-1}$  to  $15.6 \pm 0.7 \times 10^2 \text{ mm}^2 \text{ L}^{-1}$  on day 12. CSP concentration in the bulk water started with  $12.9 \pm 10.7 \times 10^6 \text{ L}^{-1}$  in abundance and  $0.5 \pm 0.04 \times 10^2 \text{ mm}^2 \text{ L}^{-1}$  in total area respectively, and increased to the first peak on day 9 for abundance and on day 5 for total area, and then declined.'

Page 11 lines 14-15 in bulk or in SML? \*\* We will add the information 'in SML'.

Section 3.3 Authors do not say whether they are describing PSD's in the SML or in bulk water at any moment. Assuming that this is only SML, some wording about changes in PSD in the bulk water could help understand these differences and to infer gel dynamics in the whole system through time \*\*We agree that the assignment to the SML or to the bulk water is missing in the current version. We will thoroughly revise the text to give this information. Information of PSD's in bulk water will be added: 'Size distribution of gel particles (dp: 2-16 $\mu\text{m}$ ) in the bulk water also followed the power law relationship of Eq. (2) (mean of  $r^2=0.99$ ), varying between -3.48 and -1.94 (mean value: -2.56, SD: 0.49) for TEP and between -3.43 and -2.01 (mean value: -2.50, SD: 0.42) for CSP.

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For the slope of the size distribution in the bulk water, no significant difference was observed between high and low wind speeds. The PSD of both TEP and CSP in the bulk water and SML were shallower after adding a seed culture of *E. huxleyi* on day 20 and a biogenic SML from a previous experiment on day 21 (Fig. 8) ( $p < 0.05$ , two sample-Kolmogorov-Smirnov test), i.e., the average slope of CSP in size of 2-16 $\mu\text{m}$  in the bulk was -2.84 before and -2.15 after addition of the *E. huxleyi* culture.

Page 12 lines 16-12 include some wording about enrichment factors in the high wind speed treatments \*\*The information will be added.

Page 12 lines 23-24 and page 13: Where, in the SML or in bulk? This differentiation should be clearly stated across the whole MS. \*\*It is in the SML. This differentiation will be clarified across the whole MS.

Page 13 lines 1-19 Maybe include the different slope values in a Table, as in the Figure it is hard to see if the difference is in slope or in the intercept. \*\* We agree and will add the table in the supplementary material.

4. Discussion: Page 14 lines 8-12 I don't think this sentence is necessary since you are not discussing any results. \*\* We agree and will delete it.

Page 14 line 24 remain enriched \*\* Will be adopted.

Page 15 line 4-page 16 line 2. This paragraph is very long and it is not clear how it is connected to the results obtained, which I think should be more carefully introduced in the discussion: For instance, do you refer to your measurements in the SML, in bulk water, or in both? And, according to Kepkay 1994, shear is a dominant mechanism for particle aggregation; so how do you link this with the trend towards smaller gel particles at high wind speeds? \*\* We will modify the paragraph to improve its understanding: 'Aggregation processes are primarily driven by encounter rates between particles that depend on particles concentration and turbulent shear (Mari and Robert, 2008; Ellis et al., 2004). It has been suggested that TEP volume concentration increases con-

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tinuously under the low turbulence intensity by promoting the formation of TEP, but that TEP volume concentration and the fraction of large TEP are reduced at stronger shear (Mari and Robert, 2008). Thus, the effect of wind shear on gel aggregation is double-edged, and large aggregates may be broken apart when the turbulence intensity increases. In this study, the disruptive effect of the wind shear becomes more important when the wind speeds increases, therefore, larger gel became smaller at high wind speed. Reference: Ellis, K.M., Bowers, D.G., Jones, S.E.: A study of the temporal variability in particle size in a high energy regime. *Coast. Shelf Sci.* 61, 311–315, 2004. Mari, X., Robert, M.: Metal induced variations of TEP sticking properties in the southwestern lagoon of New Caledonia, *Marine Chemistry*, 110, 98–108, 2008.

Page 16 lines 18-23. I do not see why. Average PSD are similar for TEP and CSP, and even lower for CSP at high wind speeds (page 13 lines 4-11). Or you said that because the change in PSD between high and low wind speeds was higher for TEP? Please clarify; and please refer to the results. To support this conclusion, maybe authors could look at the change in PSD of TEP and CSP through time; so check if these gel particles had been actually aggregated in the SML or not. \*\* According to the results, the average slopes showed about 41.2% changes at speed > 8 ms<sup>-1</sup> for TEP in the SML, but only 23.8% for CSP in the SML. The change in PSD between high and low wind speeds was thus higher for TEP than CSP in the SML. In addition, after adding the *E. huxleyi* seed culture, no influence of wind speed on size distribution of CSP was detected. These results indicated that the influence of wind speed on PSD of gel particles in the SML may be more pronounced for TEP than for CSP, and that CSP are less prone to aggregation than TEP during the low wind speed.

Section 4.3. I think it would be nice to comment about the changes in EF's through time. They apparently decrease until the phytoplankton culture is added (Table 2), even though you say that "a strong accumulation occurred in the SML (e.g. abstract line 13). How do you explain these decreases at low wind speeds? I would appreciate some comments about your day 15; any explanation to this exceptional behavior? \*\*We

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think your suggestion is valuable. We will add the information: 'Pronounced changes through time in gel size slope and EF's were observed after the addition of *E. huxleyi* seed culture. At that time, shallower slopes for PSD of CSP and TEP revealed a higher abundance of larger gel particles relative to smaller ones for both SML and bulk water. Gel particles produced by autotrophs may be more surface active and more prone to aggregation (Zhou et al., 1998). The larger particle combined with the ballast effect of *E. huxleyi* are more easily to sink out of the SML. This, to a certain extent, may explain that a decrease in the EF's of CSP and TEP after the addition of *E. huxleyi* seed. The observed changes after addition of the *E. huxleyi* seed culture indicates that variations of gel particles in the SML may also depend on the source of gels and gel precursors. Zhou, J., Mopper, K., and Passow, U.: The role of surface-active carbohydrates in the formation of transparent exopolymer particles by bubble adsorption of seawater, *Limnol Oceanogr*, 43, 1860-1871, 1998.

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