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Comment on acp-2021-845

Anonymous Referee #1

Referee comment on "High number concentrations of transparent exopolymer particles in ambient aerosol particles and cloud water – a case study at the tropical Atlantic Ocean" by Manuela van Pinxteren et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-845-RC1>, 2021

Transparent exopolymer particles (TEP) have been shown as highly prevalent in the sea surface microlayer (SML) with a potentially significant effect on air-sea release of marine aerosols. They are also highly important in sedimentation processes and carbon cycling in the sea. This study presents TEP number concentrations $> 4.5 \mu\text{m}$ in ambient atmospheric samples from the tropical Atlantic Ocean during the MarParCloud campaign as well as in generated atmospheric particles using a plunging waterfall tank. The publication presents a robust data set on atmospheric TEP measurements that are rare to date and concluded interesting new findings. Authors speculate that the high enrichment of TEP in supermicron particles and in cloud water result from a combination of enrichment during bubble-bursting transfer from the ocean and in-situ atmospheric formation. They also propose that similar (biotic and abiotic) formation mechanism reported for TEP formation in the (sea)water might take place in the atmosphere as well, as the required conditions (e.g. high concentrations of dissolved TEP precursors such as polysaccharides, presence of bacteria in the cloud water) were given. I believe that this manuscript is suitable for ACP (and notably this Special Issue). The publication sets new research trends and points to the necessity of further investigations on the assessment of the importance of the biotic versus the abiotic TEP formation pathways in the atmosphere. It seems to be important to continue such measurements in other remote oceanic locations, since marine gel particles, their in-cloud formation and connection to bacteria in the atmosphere could be highly relevant for a better understanding of marine cloud properties. Especially important, in my opinion, is the determination of TEP concentrations in waters with high productivity, i.e. the Baltic Sea, especially since high TEP concentrations are usually associated with phytoplankton blooms, with the special importance of diatoms.

The introduction is written very clearly and allows to fully understand the mechanisms of TEP formation, their properties, distribution to the atmosphere and the role they play in the environment. This chapter describes the current state of knowledge on TEP subject.

Please indicate some examples of particles or highly dense matter, that support downward carbon fluxes and those, that will result lead to rise of TEP to the surface and to form or stabilize the SML (Lines 104-108).

Line 101- there is no indentation in the paragraph

Material and methods

Lines 2015-216; 223 and 235- what acid was used? Could the use of the acid influence further analysis? How were filters/bottles etc., treated after using acid? - there is no precise description or reference to the literature in which it was previously described. The remaining methodological description does not raise my reservations.

Results and Discussion

The discussion is logical and brings a lot of interesting information. Statistical analysis of the results also does not raise any reservations. Below are some comments, questions and suggestions.

Lines 330-333 - It seems to me that it is exaggerated to say that the majority of the TEP particles are activated to cloud droplets when a cloud forms, only on the basis that striking similarity for TEP concentrations in the aerosol particles and the cloud water was found Especially since the samples from the clouds were collected only in the amount of 3 ... - Please explain where this statement came from.

Line 343- I propose to divide Fig. 3 and here leave only part "a", because in the text there is description only of that figure. Part b I suggests inserting after line 433- when the authors describe the EF coefficients.

Line 365- remove "polymer gels"

Lines 391-392- Remove "and are discussed in more detail in Engel et al. (2020)". There is a reference to this literature at the end of the paragraph, which is enough.

Lines 415-418- Remove "Ocean water, atmospheric particles, and cloud water are different marine compartments". It doesn't sound logical. I propose to start the sentence

with: "To compare seawater and atmospheric TEP concentrations in terms of..."

Lines 479-484- I think that an important aspect of the influence of wind speed on the generation of marine aerosols has been overlooked in this section and should be noted. However, there is a mention of this on lines 99-101 (Introduction). Wind speed has a direct impact on the concentration of sea salt (both sodium and chloride) in the atmosphere above the sea/ocean. The effectiveness of marine drops generating and dispersing of large sea salt nuclei from the surface of breaking waves increases with square of the wind speed and, in the case of whitecaps occurrence (wind speed above 10 m s^{-1}), changes with wind speed cubed (Nair et al, 2005; O'Dowd and Hoffmann, 2005). Thus the increase in wind speed is directly related to the increase in sea salt concentrations in aerosols. Perhaps the same process applies to TEP, hence its higher concentrations in the ambient atmosphere than from the plunging waterfall tank were noted. Lewandowska and Falkowska (2013) determined that the amount of sea salt transferred into the near-water layer of the atmosphere increases exponentially already with wind speed over $3 \text{ m}\cdot\text{s}^{-1}$. The limit value over sea was the same as suggested in literature (Nair et al, 2005; Meira et al, 2007).

Lines 538-539- Remove the sentence "The pH in the cloud water analysed here was between 6.3 and 6.6." and in Line 541 I propose to change the sentence like this:- "At cloud water pH-values was between 6.3 and 6.6, and marine gels could split into smaller units (Chin et al., 1998), that are below the minimum detectable particle size of $4.5 \mu\text{m}$."

Line 541- In my opinion it should be: "... the different factors such as pH, ion density ..." or "...the variables pH, ion density..."

Line 547- I propose like this: "...fully explain the role of each of these effects but our investigations..."

In Chapter 3.3.2.2 (Biotic formation)- Authors indicated that TEP can be directly released as particulates from aquatic organisms involving phytoplankton and bacteria (Lines 551-552). However, cyanobacteria and microalgae, which are also present in the air, have been omitted. An entire chapter is devoted to bacteria. While there are no reports on cyanobacteria and microalgae in cloud water, there have been many publications on their presence in aerosols recently (e.g. Sharma et al., 2007; Genitsaris et al., 2011; Després et al., 2012; Sahu and Tangutur, 2014; Lewandowska et al., 2017; WiÅńniewska et al., 2019 and much more). It is also worth mentioning them in this publication, even if it were only a few sentences. Especially that in the introduction Authors mentioned that high TEP concentrations can be associated with phytoplankton (mainly diatom) blooms. Perhaps the considerations in Chapter 3.4 (Lines 626- 637) regarding bacteria could also apply to cyanobacteria and microalgae and their metabolic degradation products that occur in aerosols? Such a reflection for the future.

Conclusions are fine to me.

Caption of Figures are comprehensive and, in my opinion, correct. The same for **tables**.

Figures:

Fig. 1- Enlarge so that the scale in the drawings was visible

Fig. 2- TEP concentrations were below the limit of detection (LOD) only on 26th of September 2017 - as shown in the picture. And also: "...the three cloud water samples (blue-red squares)"- it looks black - red rather than blue - red squares.

Fig. 5- Please give superscripts on the oy axis [mL^{-1}]

Fig. 6- Remove the comma in front of the [%] in the oy axis. The % values for the range 5.5 to 7 μm should be given above the bars as for the other bars

Fig 7- Increase the descriptions for both axes in all figures

Literature- Align to margins and validate against journal guidelines