Referee 1:

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 µm 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 present 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 a 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.

We thank the Reviewer for the evaluation and the constructive comments. Replies to the specific Referee's comments are provided below in red and new parts included in the manuscript are marked in **green**. Line numbers refer to the revised (clean) version.

1. **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).

As the reviewer suggested, we included some more references that show downward carbon fluxes and the rise of TEP to the surface forming the SML. We included the study of (Logan et al., 1995) who studied the "rapid formation and sedimentation of large aggregates is predictable from coagulation rates (half-lives) of transparent exopolymer particles (TEP)" as well as the review paper of (Mari et al., 2017)" Transparent exopolymer particles: Effects on carbon cycling in the ocean" which covers all aspects of

TEP rising and sinking. In addition, we included two papers studying TEP at the SML (Wurl and Holmes, 2008;Wurl et al., 2011).

2. Line 101- there is no indentation in the paragraph

   This was corrected.

Material and methods

3. 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.

   The revised version described the cleaning procedure in more detail. It is a standard procedure for DOC and TEP analysis and after cleaning with HCl, the equipment is rinsed with ultrapure water as recommended in Engel et al. (2009). This cleaning procedure removes contaminations very efficiently and the usage of acid does not influence further analysis. We added a reference and it reads now:

   Line 227-230: "All equipment that was in contact with the cloud water samples (Teflon®strands, sampling bottles, filters) had been cleaned with 10% HCl and rinsed with ultrapure water (resistivity=18.2MΩ cm) before each application as recommended in (Engel, 2009)".

   And:

   Line 201-203: "The PC filters had been cleaned with 10% HCl and rinsed ultrapure water (resistivity = 18.2MΩ cm) water before application."

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.

4. 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.

We thank the reviewer for this comment. This was clearly a mistake and the term “the majority of the TEP are activated to cloud droplets” was not correct. Unfortunately, the cloud water sampling cases for TEP measurements were limited. For a more correct assignment, we compared the #TEP concentrations in cloud water to the #TEP concentrations in the ambient aerosol particles when sampling times coincided (20.09., 28.09., and 4.10. 2017). For these dates, the cloud water #TEP concentrations made up between 10 and 34% (average: 20%) of the #TEP ambient aerosol concentrations. In addition, we related the average cloud water #TEP concentrations to the average #TEP concentrations in the ambient aerosol, which showed that the cloud water #TEP concentrations made up 24% of the #TEP ambient aerosol concentrations. Regarding these numbers, we conclude that the #TEP concentrations in cloud water were about 20% of the #TEP concentrations in the ambient aerosol (with a good agreement regarding the matching dates and the average values) and added this information in the manuscript as follows:

Line 336-338: “Comparing the #TEP concentrations in cloud water to the ones in the ambient aerosol particles suggested that about 20% of the ambient TEP particles are activated to cloud droplets when a cloud forms.“

5. 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.

   We understand the point the reviewer raised and it makes sense from a logical point of view. However, as the Figure 3b is in a very similar format as Figure 3a they somehow belong together (although being discussed in different chapters). For the sake of clarity as well as for avoiding including too many single Figures, the authors would prefer to leave it as it is.

6. Line 365- remove “polymer gels”

   Done

7. 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.

   Done

8. 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…”.

We agree and adjusted the text as suggested.

9. Lines 479-484- I. 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).

   We thank the reviewer for this comment. The wind speed data are listed in the SI (Tab. S1) and present average data of a 24 h sampling interval. As shown in the manuscript, we observed a good correlation of sodium, magnesium and sea-salt calcium to the TEP concentrations that indicates some connection to bubble bursting transfer. However, there is no correlation between #TEP and wind speed. In addition, the correlation between sodium and wind speed is surprisingly weak (R^2 = 0.2). It may be that since wind speed data represented an average value of 24 hours, short but pronounced changes in the wind speed were not visible in the average wind speed value. We think this is a separate topic to investigate.

   We added to the manuscript: Line 499-501: "However, a correlation of TEP to wind speed was not found. It may be that since wind speed data represented an average value of 24 hours, short but pronounced changes in the wind speed were not visible in the average wind speed value."

10. II. 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 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 $m \cdot s^{-1}$. The limit value over sea was the same as suggested in literature (Nair et al, 2005; Meira et al, 2007).

    Regarding the tank samples, the waterfall is simulating the bubble bursting (and the wind speed). We cannot translate the waterfall intensity and the resulting sea spray formation to a certain wind speed for a comparison with the ambient conditions. However, sodium concentrations in the tank were higher than in the atmospheric concentration, which shows that there was a strong sea spray generation. At the same time, TEP concentrations were significantly lower in the tank (compared to ambient conditions), suggesting that bubble bursting is not the major driver for TEP on the aerosol particles, supporting the idea of a secondary formation in the atmosphere. We underlined this in the manuscript more strongly by adding the following text:

Line 450-455: “It should be noted that the lower enrichment in the tank resulted from the lower TEP number concentrations in the generated aerosol particles, as the particulate sodium concentrations in the tank aerosol were even higher than in the ambient particles (Tab. S3). This suggests that, although an artificial tank study cannot represent the ambient environment, the generation of sea spray aerosol was in progress; however, TEP transfer seemed to be not pronounced. “

To underline the effect of wind speed on the sea spray aerosol production and potential TEP transfer, we added the reference of Lewandowska and Falkowska (2013) and others in the introduction as follows:

Line 108-111: “Due to wind and breaking waves, sea spray aerosol particles are formed (de Leeuw et al., 2011;Lewandowska and Falkowska, 2013;Liss and Johnson, 2014) that could be a transfer mechanism for TEP from the ocean to the atmosphere.”

-1

11. 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 µm.”

We agree and rephrased these sentences. However, we wanted to differentiate between marine gels (mentioned by Chin et al. 1998) and the TEP analysed here. It now reads:

Line 578-581: “The measured cloud water pH-value of the samples analysed here was between 6.3 and 6.6, at which marine gels could split into smaller units (Chin et al., 1998). Hence, a part of the cloud water TEP might be below the minimum detectable particle size of 4.5 µm due to the slightly acidic conditions.”

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

We agree and changed to: Line 577: “…might be affected by the different factors such as pH, ion density, temperature and pressure in the atmosphere.”

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

We agree and changed to: Line 584-585: “from our data we cannot fully explain the role of each of these effects and such investigations warrant further studies”.

14. 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; WiÅ›niewska 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.

We agree that cyanobacteria and microalgae and their metabolic degradation products that occur in aerosols might contribute to atmospheric TEP processing. We thank the reviewer for his constructive input and took up this interesting thought, adding the following text:

Line 622-626: “Besides, although not measured here, microalgae and cyanobacteria, that are relevant for direct TEP formation in seawater, have been reported to occur in the atmosphere (e.g. Lewandowska et al., 2017;Sharma et al., 2007;Wiśniewska et al., 2019;Wiśniewska et al., 2022). It is worth studying, if these species and their metabolic degradation products contribute to atmospheric TEP processing.”

Line 709-713: Finally, while dust might be a dominant INP source in the here investigated tropical Atlantic region close to the Saharan desert, in other remote oceanic locations, marine gel particles, their in-cloud formation and connection to bacteria **and phytoplankton** in the atmosphere could be highly relevant for a better understanding of marine cloud properties.

Conclusions are fine to me.

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

Figures:

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

The resolution of the Figures appear partly poor due to the requested portrait mode. For publication, they will be provided in the best possible format and resolution in a separate document to ensure readability.

16. **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.

    Thanks for noting these issues. The dates were corrected and the colour description was corrected.

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

    Done

18. **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

    Done

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

    We increased the axis descriptions. As mentioned above, the resolution of the Figures appear partly poor due to the requested portrait mode. For publication, they will be provided in the best possible format and resolution in a separate document to ensure readability.

20. **Literature**- Align to margins and validate against journal guidelines

    Done

Citation: https://doi.org/10.5194/acp-2021-845-RC1

Cited Literature:

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