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Comment on acp-2021-845

Anonymous Referee #2

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-RC2, 2022

Comment on "High number concentrations of transparent exopolymer particles (TEP) in ambient aerosol particles and cloud water – A case study at the tropical Atlantic Ocean" by Manuela van Pinxteren et al.

The manuscripts deals with an interesting and quite new topic and presents some potentially important hypothesis and scientific questions (Are "secondary" TEPs more important than "primary" ones in the marine atmosphere? Is the marine aerosol TEP population connected in any way to the INP population?). The paper is well written and sufficiently clear. I recommend publications once the following (minor) issues are clarified.

L40. The TEP concentrations reported do not represent supermicron aerosols. They correspond to particles larger than 4.5 microns (as correctly stated above), which represents a subset of the supermicron TEP population. Please modify for major accuracy.

L42-43. I would suggest to add that the conversion was based on the observed cloud LWC.

L43-46. The TEP concentration in the tank headspace has no atmospheric relevance. It results from the experimental parameters chosen to operate the tank and can be modified

just by varying them (e.g. headspace flush flow, intensity of the plunging jets, and so on…). The only general and valuable information that can be extracted from sea-spray tanks regards the (size-dependent) relative chemical composition of the produced sea-spray particles. I am not against presenting the obtained TEP concentration in the tank, but it should not be reported in the abstract. More comments on this issue follow below.

L81. Instead of "contain" maybe "have" would be more correct?

L303-321. Please clarify where the seawater TEP concentration used to derive the EFs comes from. Reading further on, one understands that it derives from Engel et al. (2020). For major clarity, this information should be added here.

L330-333. Please, provide a more quantitative comparison between the TEP concentration in aerosol and cloud water. If the authors think that this is not possible, they should re-consider the following sentence: "suggesting that the majority of the TEP particles are activated to cloud droplets when a cloud forms". Judging by the plot in Figure 2 and comparing the cloud cases with the corresponding aerosol samples (which is a very raw approach), I would say that less than 30% of TEPs are activated into a cloud. This is in contrast with the above statement. If the authors have data to support their statement, I would invite them to discuss them quantitatively. By the way, the highlighted statement seems to be contradicted by the authors themselves in Lines 543-545.

L336-341. The concentration of TEP in the sea-spray aerosol generated by the tank is not an atmospheric relevant information; it depends only on the chamber design and settings. Therefore, there is no reason to compare the tank TEP concentration with that of atmospheric samples, either by performing a statistical test or not. The fact that the chamber produced lower concentrations of TEP with respect to what observed in ambient samples is irrelevant; the only informative data available from the tank are the EF data (because they are based on the relative chemical composition i.e., TEP/Na+), which the authors use correctly to infer about TEP sources later on in the text.

L347. "besides for the…". Please double check English.

L429-433. [Par. 3.2]. Which seawater TEP concentration was used to calculate the EF for the sea-spray tank samples? Where TEP directly measured in the same seawater used for the bubbling experiments? Or did the authors use the average ETNA values from Engel et al. (2020)? Please provide this information here and in the Experimental Section. This may be quite a critical point. Considering how variable biochemical parameters may be in seawater, assuming that the average TEP concentration measured by Engel et al. (2020) is representative of the water samples used for the bubbling experiments is quite risky. The comparison of TEP EFs between atmospheric and tank samples is the only solid base that supports the (very interesting) hypothesis of important "secondary" TEP formation processes in the atmosphere. If the EF calculated for the tank samples are biased by assuming a seawater TEP concentration that does not truly represent the real situation in

the tank, this base appears much less solid. In this case, I must invite the authors to add some caveat in the text, making it clear to the readers that the hypothesis, although very interesting, needs to be further demonstrated as doubts still persist at this stage given the uncertainties inherent to the EF calculations for tank samples.

L479-484. “the high abundance and enrichment of #TEP in the ambient aerosol particles compared to literature data (Kuznetsova et al., 2005) and compared to the concentration and enrichment of the #TEP from the plunging waterfall tank performed here, suggests that additional TEP sources in the ambient atmosphere exist from which TEPs are added to their primary transfer by bubble bursting from the oceans”. I stress again that the difference in concentration of TEP between atmospheric and tank samples is not supporting the existence of secondary TEP formation processes in the atmosphere. It is only the result of the tank properties. Only the difference in the TEP enrichment with respect to Na between tank and atmosphere supports this. Please, remove the references to “abundance” and “concentration” in the above sentence.

Par3.3.2. This paper focuses on atmospheric particles larger than 4.5 μm, which are characterized by fairly short atmospheric residence times. It may be worth discussing if the atmospheric lifetime of these particles is consistent with the timescale of TEP (biotic and abiotic) formation reactions. Data should be available at least for the seawater compartment if not for the atmosphere.