Supporting information for:

High number concentrations of transparent exopolymer particles (TEP) in ambient aerosol particles and cloud water – A case study at the tropical Atlantic Ocean

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Table S1: Sampling period of ambient aerosol particles (TSP) and meteorological parameters and inorganic ions sampled from PM_{10} during the sampling interval.

Sample ID	Start sampling (UTC)	Stop sampling (UTC)	V [m^{-3}]	wind speed [$m\ s^{-1}$]	wind speed [deg]	Temp [degC]	RH [%]	Magnesium [$\mu g\ m^{-3}$]	Calcium [$\mu g\ m^{-3}$]	ss Ca [$\mu g\ m^{-3}$]	non-ss Ca [$\mu g\ m^{-3}$]	Dust [$\mu g\ m^{-3}$]
TEP (Air)-2	19.09.2017 17:05	20.09.2017 16:05	11.8	2.4	159.3	27.4	86.6	0.35	1.12	0.15	0.97	38.2
TEP (Air)-3	20.09.2017 16:38	21.09.2017 14:20	11.3	4.6	79.3	27.4	87.3	no data	no data	no data	no data	30.0
TEP (Air)-4	21.09.2017 14:41	22.09.2017 15:30	12.7	8.4	32.5	27.0	82.3	0.35	0.44	0.16	0.27	14.5
TEP (Air)-5	22.09.2017 15:50	24.09.2017 14:55	24.1	6.8	52.9	26.4	75.1	0.25	0.21	0.12	0.09	4.1
TEP (Air)-6	24.09.2017 15:20	25.09.2017 16:27	12.9	4.2	58.4	26.1	75.3	0.23	0.67	0.11	0.56	2.2
TEP (Air)-7	25.09.2017 16:58	26.09.2017 15:13	11.3	3.6	40.8	26.5	82.2	0.31	no data	no data	no data	11.6
TEP (Air)-8	26.09.2017 15:36	27.09.2017 10:08	9.7	3.9	42.5	26.8	84.1	0.21	0.90	0.05	0.84	37.6
TEP (Air)-9	27.09.2017 10:42	28.09.2017 12:25	13.2	4.6	44.1	27.1	84.2	0.24	0.80	0.07	0.73	20.6
TEP (Air)-10	28.09.2017 12:48	29.09.2017 15:50	13.6	6.2	31.8	27.2	85.5	0.47	0.99	0.19	0.80	27.3
TEP (Air)-11	29.09.2017 16:20	30.09.2017 10:30	9.1	6.9	34.8	27.0	85.0	0.58	0.84	0.25	0.59	27.3
TEP (Air)-12	30.09.2017 11:00	01.10.2017 10:45	12.1	8.2	38.0	26.7	81.9	0.48	0.60	0.20	0.39	42.7
TEP (Air)-14	01.10.2017 11:03	02.10.2017 10:55	12.2	8.5	40.3	26.2	78.9	0.32	0.47	0.14	0.33	29.1

TEP (Air)-15	02.10.2017 11:19	03.10.2017 12:44	13.0	6.1	46.2	26.2	78.9	0.25	0.54	0.12	0.42	14.8
TEP (Air)-16	03.10.2017 13:10	04.10.2017 13:09	12.1	5.7	37.8	26.2	77.7	0.21	0.60	0.10	0.50	13.2
TEP (Air)-17	04.10.2017 14:34	05.10.2017 13:45	11.5	5.2	49.3	26.4	79.7	0.19	0.53	0.12	0.41	17.2
TEP (Air)-18	05.10.2017 14:13	06.10.2017 10:32	10.2	4.9	40.0	26.5	84.2	0.47	0.56	0.18	0.38	17.0
TEP (Air)-19	06.10.2017 11:00	07.10.2017 16:09	14.5	7.0	19.0	26.5	85.9	0.54	0.66	0.21	0.45	16.8
TEP (Air)-20	07.10.2017 16:31	08.10.2017 11:38	9.5	6.8	21.1	26.4	86.1	0.51	0.56	0.20	0.36	16.8
TEP (Air)-21	08.10.2017 12:00	09.10.2017 09:47	10.9	6.6	39.0	26.4	84.2	no data	no data	no data	no data	27.6
TEP (Air)-22	09.10.2017 10:13	10.10.2017 11:05	12.4	7.3	41.3	26.3	78.6	0.25	0.44	0.13	0.31	27.6

Table S2: TEP concentrations as well as inorganic ion concentrations and calculated enrichment factors of the ambient aerosol particles (TSP)

	Atmosphere							Ocean			EF
Sample ID	TEP [# m^{-3}]	TEP [μm^3 m^{-3}]	$TEP_{5\text{-}10\mu m}$ [# m^{-3}]	$TEP_{5\text{-}10\mu m}$ [μm^3 m^{-3}]	Na^+ [mg m^{-3}]	Na^+- PM_1 [mg m^{-3}] 1)	TEP/$Na_{aer.}$ [μm^3 mg^{-1}]	$TEP_{5\text{-}10\mu m}$ [μm^3 L^{-1}]2)	Na^+ [mg L^{-1}]3)	TEP/Na_{ocean} [μm^{-3} mg^{-1}]	$EF_{aer.\ ambient}$ 4)
TEP (Air)-2	1.5E+04	9.8E+06	1.2E+04	2.0E+06	0.0040	0.0039	5.14E+08	4.57E+08	1.0E+04	4.57E+04	1.12E+04
TEP (Air)-3	1.5E+04	1.7E+07	9.9E+03	2.2E+06	no data	no data	no data	4.57E+08	1.0E+04	4.57E+04	no data
TEP (Air)-4	6.0E+03	7.0E+06	4.2E+03	9.1E+05	0.0042	0.0041	2.22E+08	4.57E+08	1.0E+04	4.57E+04	4.85E+03
TEP (Air)-5	4.3E+03	3.2E+06	1.6E+03	5.7E+05	0.0031	0.0030	1.92E+08	4.57E+08	1.0E+04	4.57E+04	4.19E+03
TEP (Air)-6	5.0E+03	2.8E+06	3.5E+03	3.1E+05	0.0028	0.0027	1.13E+08	4.57E+08	1.0E+04	4.57E+04	2.46E+03
TEP (Air)-7	1.7E+04	1.8E+07	1.3E+04	2.2E+06	0.0033	0.0032	6.80E+08	4.57E+08	1.0E+04	4.57E+04	1.49E+04
TEP (Air)-8	< LOD	-	-	-	0.0014	0.0013	0.00E+00	4.57E+08	1.0E+04	4.57E+04	no data
TEP (Air)-9	6.9E+02	1.2E+06	4.3E+02	8.3E+04	0.0018	0.0017	4.94E+07	4.57E+08	1.0E+04	4.57E+04	1.08E+03
TEP (Air)-10	2.1E+04	1.9E+07	2.0E+04	2.9E+06	0.0050	0.0049	5.85E+08	4.57E+08	1.0E+04	4.57E+04	1.28E+04
TEP (Air)-11	2.7E+04	3.7E+07	1.8E+04	3.9E+06	0.0065	0.0064	6.09E+08	4.57E+08	1.0E+04	4.57E+04	1.33E+04
TEP (Air)-12	9.7E+03	6.4E+06	7.7E+03	1.3E+06	0.0053	0.0052	2.40E+08	4.57E+08	1.0E+04	4.57E+04	5.24E+03
TEP (Air)-14	8.9E+03	3.8E+06	7.3E+03	1.3E+06	0.0037	0.0036	3.58E+08	4.57E+08	1.0E+04	4.57E+04	7.82E+03
TEP (Air)-15	4.8E+03	3.3E+06	3.9E+03	5.9E+05	0.0033	0.0032	1.88E+08	4.57E+08	1.0E+04	4.57E+04	4.11E+03
TEP (Air)-16	9.6E+03	7.4E+06	7.6E+03	1.4E+06	0.0026	0.0025	5.52E+08	4.57E+08	1.0E+04	4.57E+04	1.21E+04
TEP (Air)-17	8.4E+03	6.9E+06	6.7E+03	1.2E+06	0.0030	0.0029	4.14E+08	4.57E+08	1.0E+04	4.57E+04	9.04E+03
TEP (Air)-18	1.6E+04	1.3E+07	1.2E+04	2.2E+06	0.0047	0.0046	4.89E+08	4.57E+08	1.0E+04	4.57E+04	1.07E+04
TEP (Air)-19	1.3E+04	8.8E+06	1.0E+04	1.8E+06	0.0055	0.0054	3.29E+08	4.57E+08	1.0E+04	4.57E+04	7.19E+03
TEP (Air)-20	*2.6E+04*	*2.4E+07*	*2.4E+04*	*4.7E+06*	0.0052	0.0051	9.13E+08	4.57E+08	1.0E+04	4.57E+04	2.00E+04
TEP (Air)-21	1.1E+04	8.4E+06	8.1E+03	1.4E+06	no data	no data	no data	4.57E+08	1.0E+04	4.57E+04	
TEP (Air)-22	3.9E+03	2.7E+06	3.1E+03	5.2E+05	0.0033	0.0032	1.62E+08	4.57E+08	1.0E+04	4.57E+04	3.53E+03
average	*1E+04*	*1E+07*	*9E+03*	*2E+06*	*0.004*	*0.004*	*4E+08*	*5E+08*	*1E+04*	*5E+04*	*8.5E+03*

[1)]Sodium mass concentrations for PM_1 show an average concentration of 0.0001 µg m^{-3} (Triesch et al. 2021) and are therefore below 3% of PM10 and are subtracted from PM_{10} sodium concentration.

[2)]TEP volume concentration were calculated from the number concentrations in a size range between 5 and 10 µm (Tab. S5) assuming spherical particles.

[3)]Average sodium concentration in seawater.

[4)] Error discussion of the $EF_{atm.}$: Even though the same TEP size ranges (diameters between 5 and 10 µm) were compared for oceanic and atmospheric measurements, the size distribution of TEP in the respective compartments might be affected by the different temperature, pressure, ion strength and pH in seawater and in the atmosphere. Such effects cannot be accounted for in the present study.
Applying sodium concentrations for the $EF_{aer.}$ and EF_{cloud} calculations also represents a source of error. TEP can form networks with inorganic cations and that might affect sodium concentrations in the seawater and in the atmosphere. However, it was reported that the TEP formation was essentially determined by the calcium concentration, while monovalent cations, such as sodium, seem not to be significantly involved in TEP formation (Meng and Liu, 2016). Hence, this error should be neglible.
Finally, the TEP number concentrations in the ocean surface water were obtained from the biologically productive Mauritanian Upwelling region in the year 2012, hence at another time and season. Compared to other oceanic regions, the values from the Mauritanian Upwelling region were at the higher end (Engel et al., 2020). The region around the CVAO is rather oligotrophic and Chlorophyll-a values during the MarParCloud campaign were relatively low with 0.1 up to 0.6 µg L^{-1} (van Pinxteren et al., 2020). As TEP is often connected to phytoplankton activity, the TEP concentration at the CVAO might be lower compared to more productive regions (Robinson et al., 2019). Lower TEP concentrations would result in higher $EF_{atm.}$ (following equation 1 in the main manuscript). Hence the here reported $EF_{atm.}$ represent a lower limit. Nevertheless, even though absolute numbers can vary, the strong differences between the $EF_{aer.\ ambient}$ and $EF_{aer.\ tank}$ (Tab. S3) are evident, as they result from the same type of seawater.

Table S3: TEP concentrations and sodium concentrations as well as enrichment factor for the tank-generated aerosol particles (TSP).

	Atmosphere								Ocean			EF
Sample ID	Start sampling (UTC)	Stop sampling (UTC)	V [m^{-3}]	TEP [# m^{-3}]	$TEP_{5\text{-}10\mu m}$ [$\mu m^3\ m^{-3}$][1)]	Na^+ [$\mu g\ m^{-3}$]	Na^+ [$mg\ m^{-3}$]	$TEP/Na_{aer.}$ [$\mu m^{-3}\ mg^{-1}$]	$TEP_{5\text{-}10\mu m}$ [$\mu m^3\ L^{-1}$][3)]	Na^+ [$mg\ L^{-1}$] [4)]	TEP/Na_{ocean} [$\mu m^3\ mg^{-1}$]	$EF_{aer.\ tank}$
Tank 1	03.10.2017 11:05	04.10.2017. 11:04	9.7	2.4E+03	4.4E+05	97.20	0.097	4.51E+06	4.57E+08	1.0E+04	5.E+04	98.6
Tank 2	04.10.2017. 11:14	05.10.2017 11:17	8.8	1.2E+03	2.1E+05	62.70	0.063	3.38E+06	4.57E+08	1.0E+04	5.E+04	73.8
Tank 3	05.10.2017 11:27	06.10.2017 12:03	9.8	5.5E+02	9.9E+04	90.05	0.090	1.10E+06	4.57E+08	1.0E+04	5.E+04	24.2
Tank 7	07.10.2017 12:00	08.10.2017. 09:05	9.4	3.9E+02	7.2E+04	51.93	0.052	1.38E+06	4.57E+08	1.0E+04	5.E+04	30.1
Tank 9	08.10.2017 09:15	09.10.2017 07:00	9.5	4.2E+02	7.7E+04	78.05	0.078	9.80E+05	4.57E+08	1.0E+04	5.E+04	21.4
average				*9.9E+02*	*1.8E+05*	*75.99*	*0.076*	*2.3E+06*	*4.6E+08*	*1.0E+04*	*4.6E+04*	*49.6*

[1)] For the tank-generated aerosol particles, solely the total number concentrations were available. TEP volume concentration were calculated from the number concentrations in a size range between 5 and 10 µm assuming spherical particles after the equation from Fig. S1: (y = 181.66x) assuming a similar size distribution of the tank-generated TEP to the ambient TEP.

[2)]Sodium was measured on TSP, however it was shown that particles greater than 10 µm radius are quickly removed in the atmosphere (Madry et al., 2011) and in sea spray model systems (Hoffman and Duce, 1976). Hence the sodium data on TSP is expected to be comparable to PM_{10} and therefore to the TEP volume concentrations between 5 and 10 µm.

[3)]TEP volume concentration were calculated from the number concentrations in a size range between 5 and 10 µm (Tab. S5) assuming spherical particles.

[4)]Average sodium concentration in seawater.

Table S4: TEP concentrations and sodium concentrations as well as enrichment factor for the cloud water samples.

			Atmosphere							Ocean			EF
Sample ID	Start sampling (UTC)	Stop sampling (UTC)	V [mL]	TEP [# L^{-1}]	TEP [# m^{-3}][1)]	$TEP_{5\text{-}10\mu m}$ [μm^3 m^{-3}]	Na^+ [mg L^{-1}]	Na^+ [µg m^{-3}][1)]	$TEP/Na_{aer.}$ [μm^{-3} mg^{-1}]	$TEP_{5\text{-}10\mu m}$ [μm^3 L^{-1}][3)]	Na^+ [mg L^{-1}] [4)]	TEP/Na_{ocean} [μm^{-3} mg^{-1}]	EF_{cloud}
WW2	20.09.2017 16:38	21.09.2017 14:20	200	4.0E+06	1.6E+03	7.2E+08[2)]	11.89	4.6	6.10E+07	4.57E+08	1.0E+04	45737	1.3E+03
WW5	28.09.2017 12:48	29.09.2017 15:50	150	9.1E+06	3.5E+03	1.6E+09[2)]	22.09	8.6	7.47E+07	4.57E+08	1.0E+04	45737	1.6E+03 [4)]
WW5 meas[3)]	*28.09.2017 12:48*	*29.09.2017 15:50*	*150*	*9.1E+06*	*3.5E+03*	*1.2E+03[3)]*	*22.09*	*8.6*	*5.51E+07*	*4.57E+08*	*1.0E+04*	*45737*	*1.2E+03[4)]*
WW8	03.10.2017 13:10	04.10.2017 13:09	200	8.2E+06	3.2E+03	1.5E+09[2)]	32.30	12.6	4.63E+07	4.57E+08	1.0E+04	45737	1.0E+03
average				*7E+06*	*3E+03*	*1E+09*	*22*	*9*	*6E+07*	*5E+08*	*1E+04*	*45737*	*1E+03*

[1)] based on a cloud liquid water content of 0.39 g m^{-3}

[2)] TEP volume concentration were calculated from the number concentrations in a size range between 5 and 10 µm assuming spherical particles after the equation from Fig. S1: (y = 181.66x) assuming a similar size distribution of the cloud water TEP to the ambient TEP

[3)] TEP volume concentrations were calculated from measured TEP number size distributions (shown in Fig. 4e).

[4)] EF_{cloud} derived from measured TEP volume concentration agreed well with EF_{cloud} from the calculated TEP volume concentration.

Table S5: Size-resolved TEP concentrations from ocean surface water (sampling depth: 10 m, average over three stations) from Engel et al., (2020). Numbers in **bold** represent values between 5 and 10 µm, corresponding to the TEP diameter on the aerosol particles and in the cloud water.

particle diameter [µm][1)]							
max	min	average	TEP[# mL^{-1}]	RSD [%][1)]	TEP [μm^3 mL^{-1}][2)]	TEP [# L^{-1}]	TEP [μm^3 L^{-1}]
1.3	*0.9*	0.6	9.04E+03	36	5.48E+03	9.04E+06	5.48E+06
1.8	*1.3*	1.7	2.42E+03	52	4.19E+03	2.42E+06	4.19E+06
2.5	*1.8*	4.8	2.08E+03	57	1.01E+04	2.08E+06	1.01E+07
3.5	*2.5*	13.9	1.86E+03	39	2.58E+04	1.86E+06	2.58E+07
5.0	**3.5**	**39.1**	**1.53E+03**	**34**	**5.99E+04**	**1.53E+06**	**5.99E+07**
7.1	**5.0**	**110**	**1.11E+03**	**33**	**1.22E+05**	**1.11E+06**	**1.22E+08**
10	**7**	**311**	**8.83E+02**	**37**	**2.75E+05**	**8.83E+05**	**2.75E+08**
14	*10*	*880*	*5.48E+02*	36	*4.82E+05*	5.48E+05	4.82E+08
20	*14*	*2492*	*3.38E+02*	62	*8.43E+05*	3.38E+05	8.43E+08
28	*20*	*7041*	*1.92E+02*	67	*1.35E+06*	1.92E+05	1.35E+09
40	*28*	*19915*	*6.10E+01*	75	*1.22E+06*	6.10E+04	1.22E+09
57	*40*	56364	3.40E+01	81	1.92E+06	3.40E+04	1.92E+09
80	*57*	159391	8.53E+00	118	1.36E+06	8.53E+03	1.36E+09
113	*80*	450908	3.34E+00	155	1.50E+06	3.34E+03	1.50E+09
160	*113*	1275410	3.40E+00	155	4.33E+06	3.40E+03	4.33E+09
226	*160*	3606696					
320	*226*	10201003					
453	*320*	28855845					
640	*453*	81617124					
10000	*640*	230792172					
10	**5**	**4.61E+02**	**3.53E+03**		**4.57E+05**	**3.53E+06**	**4.57E+08**

[1)] Relative standard deviation of three measurement stations, each probed in duplicate (n=6)

[2)] TEP volume concentration was calculated from the number concentration in a size range between 5 and 10 µm assuming spherical particles.

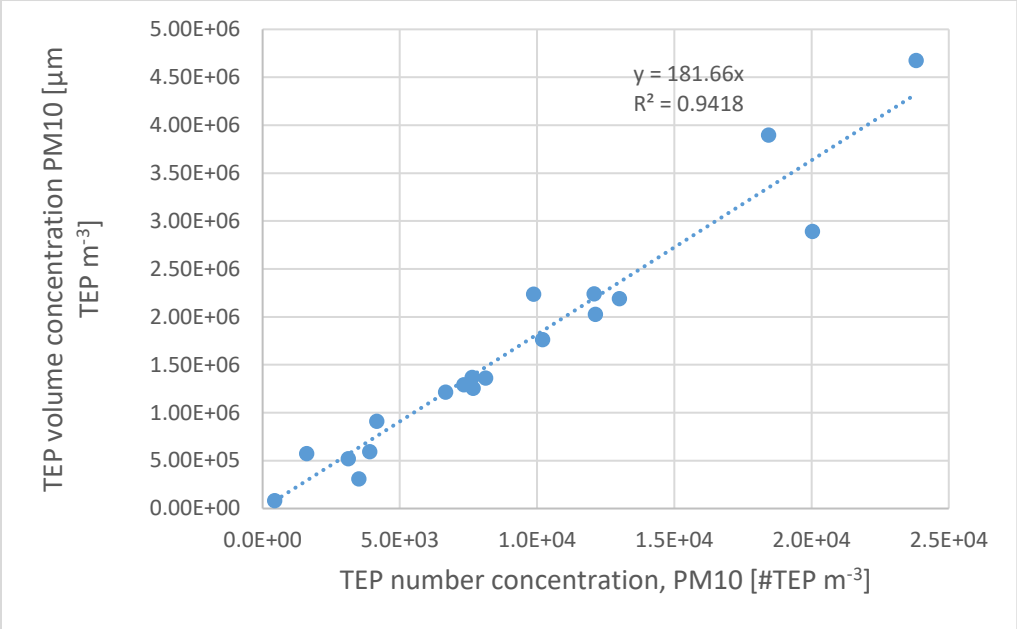

Figure S1: Correlation between TEP number concentration and TEP volume concentration achieved from the ambient aerosol samples (TEP diameter: 5-10 µm). The function was applied for the tank-generated aerosol particles and for the cloud water samples to calculate the volume concentration from the measured number concentration. A similar size distribution of the TP in the size range between 5 and 10 µm was shown for the cloud water and assumed for the tank -generated aerosol particles.

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