

**The effect of seawater salinity on sea spray aerosol production**

J. Zinke <sup>1,2</sup>, E. D. Nilsson <sup>1,2</sup>, P. Zieger <sup>1,2</sup>, and M. E. Salter <sup>1,2</sup>

[1] Department of Environmental Science, Stockholm University, Stockholm, Sweden

[2] Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden

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## Introduction

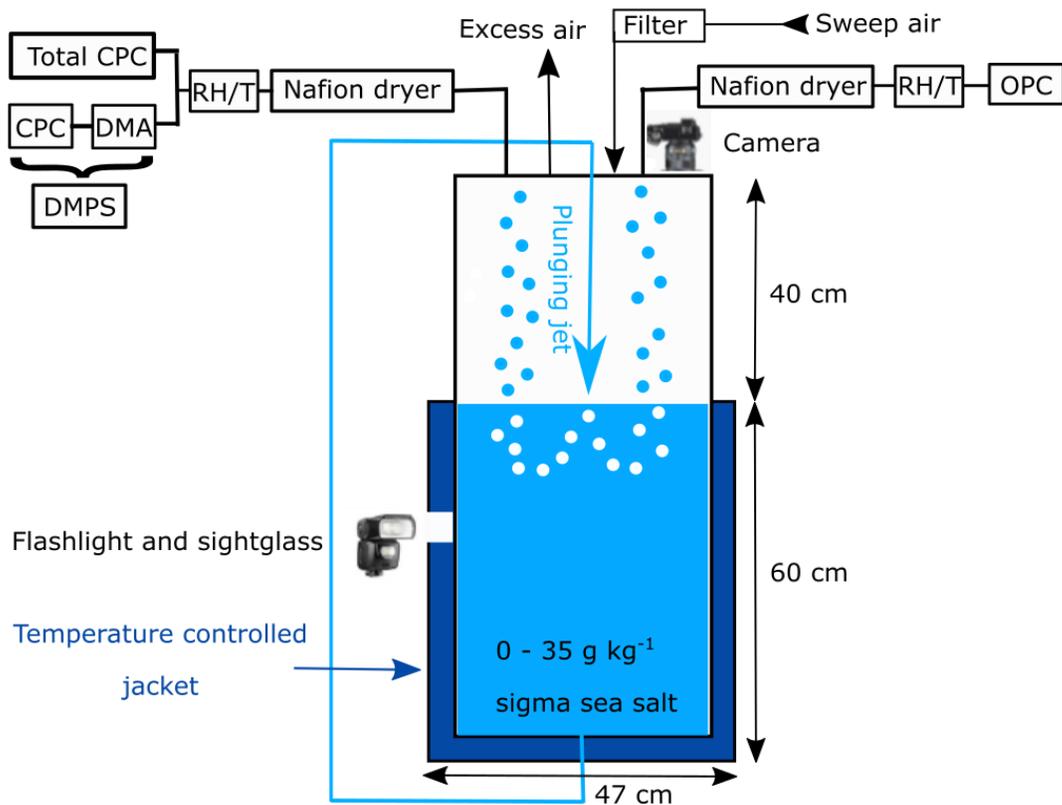
The supplementary information contains a description of the sea spray generation chamber that was used to conduct the experiments (Section S1) as well as a schematic of the chamber (Figure S1). Furthermore, we have added a description of the surface bubble spectrum measurements (Section S2) including a photograph of the water surface (Figure S2). Additional figures were included to depict the estimated particle losses in the sampling set up (Figure S3), the location of the size distribution modes at different salinities (Figure S4), a comparison of the measured total number by the CPC and the integrated number (Figure S5) and the effect of temperature at different salinities on aerosol number, surface, volume and effective radius (Figure S6). All figures result from the same dataset that was used in the main manuscript. Moreover, a table was added to provide an overview on previous studies on the effect of salinity (Table S1), as well as an overview table on the experiments conducted in this study (Table S2).

### S1 Description of the Sea Spray Chamber

A temperature-controlled sea spray generation chamber fabricated from stainless steel was generating sea spray aerosols using a plunging jet. The jet, exiting a stainless steel nozzle with an inner diameter of 4.3 mm that was situated 30 cm above the air–water interface, was used to entrain air into the water. The plunging jet was generated by circulating the water from the bottom of the tank through the nozzle using silicone tubing and a peristaltic pump (620S, Watson–Marlow, Sweden) at a flow rate of 1.73 litres per minute. The inside of the tank was coated in polytetrafluoroethylene (PTFE) below the waterline, and was rinsed thoroughly with reagent grade ethanol and deionized water. Both seawater salinity and temperature were measured continuously using a conductivity sensor (model number 4120, Aanderaa, Norway) located halfway between the tank base and the air–water interface. Furthermore, concentrations of dissolved oxygen concentration were measured with an oxygen optode (model number 4175, Aanderaa, Norway). Relative humidity and temperature were measured using a Vaisala model HMT333 probe situated in the headspace of the sea spray chamber. Dry zero-sweep air entered the tank at 10 L min<sup>-1</sup> after passing through an ultrafilter (Type H cartridge, MSA) and an activated carbon filter (Ultrafilter, AG-AK). A mass flow controller (Brooks, 5851S) was used to maintain and quantify the airflow rate. The particle-laden air was sampled through ports in the lid of the sea spray chamber and transferred to all aerosol instrumentation under laminar flow. Before entering the aerosol instrumentation, the particle-laden air was passed through nafion dryers (MD-700-48F/MD-700-36F, Perma Pure, USA) in order to reduce the relative humidity to well below 30%. The sea spray chamber was operated under slight positive pressure by maintaining the sweep air flow several litres per minute greater than the sampling rate to prevent contamination by room air and the excess air was released through a valve on the lid of the system. Figure S1 is a schematic of the set-up used.

## S2 Surface bubble spectra measurements

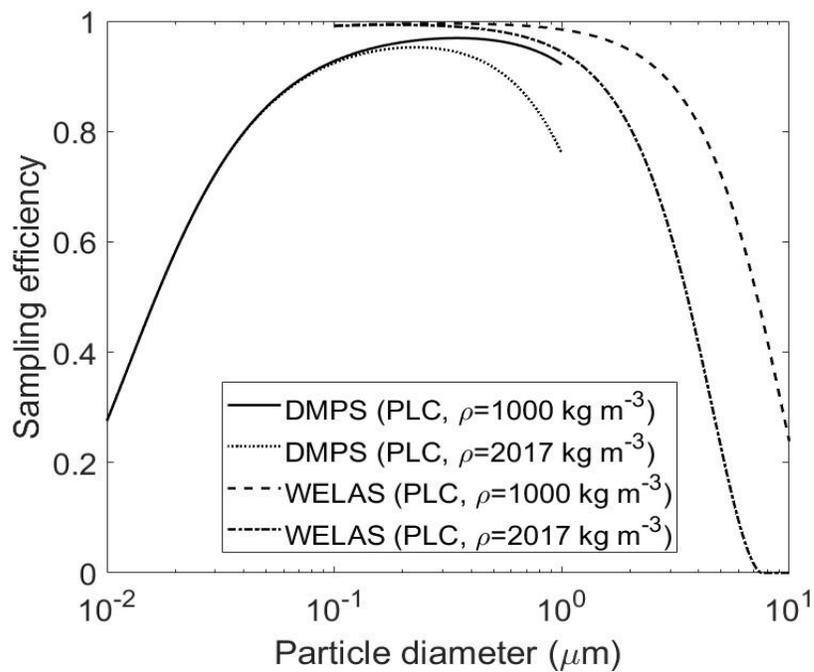
The bubble size distribution at the water surface was determined by photographing the bubbles using a Pentax K-7 Digital Single Lens Reflex camera (DSLR) equipped with a SMC Pentax-DFA Macro 100 mm F/2.8 lens. The camera was located 40 cm above the water surface and approximately 10 cm from the centerline of the plunging jet. The aperture was closed to F/11 to increase the focal depth, and the lens was automatically focused on the water surface. The bubbles were illuminated against the dark background using a Pentax AF-540FGZ flashlight that was positioned in front of a submerged viewing window in the tank wall. Photos were taken every 60 s using the PK-Tether software (Tether Tools Inc.) to ensure that the same bubbles were not counted more than once. Circles were manually fit to each bubble in an imaging software (Inkscape editor) and their radii were read from the respective SVG-files. The mean number and standard deviation of bubbles at each salinity was estimated by averaging across 20 pictures at salinities  $\leq 4 \text{ g kg}^{-1}$ , where the number of bubbles was low, and across five pictures at higher salinities, where the bubble density was much higher. The captured images had a resolution of 3104x4672 pixels corresponding to a monitored water surface area of 61x92 mm. Based on the pixel size and resolution of the camera sensor, the minimum discernable bubble film radius was estimated to be 0.02 mm.



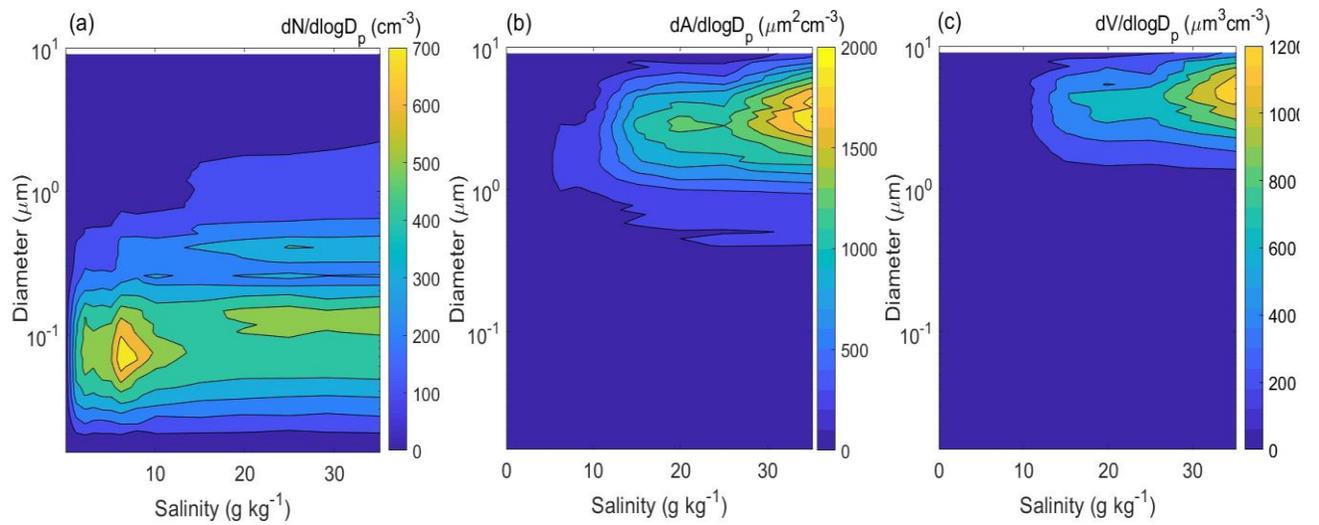
**Figure S1.** Schematic of the sea spray simulation chamber used for the experiments.



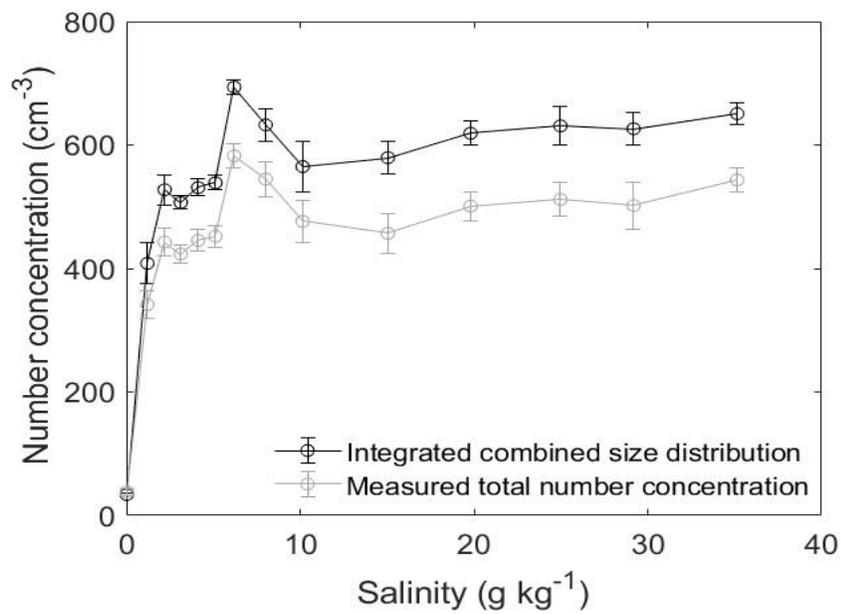
**Figure S2.** Water surface photographed with a wide angle lens with picture frame used in this study for reference. The picture was taken at  $S=35\text{gkg}^{-1}$  and  $T=20^\circ\text{C}$ .



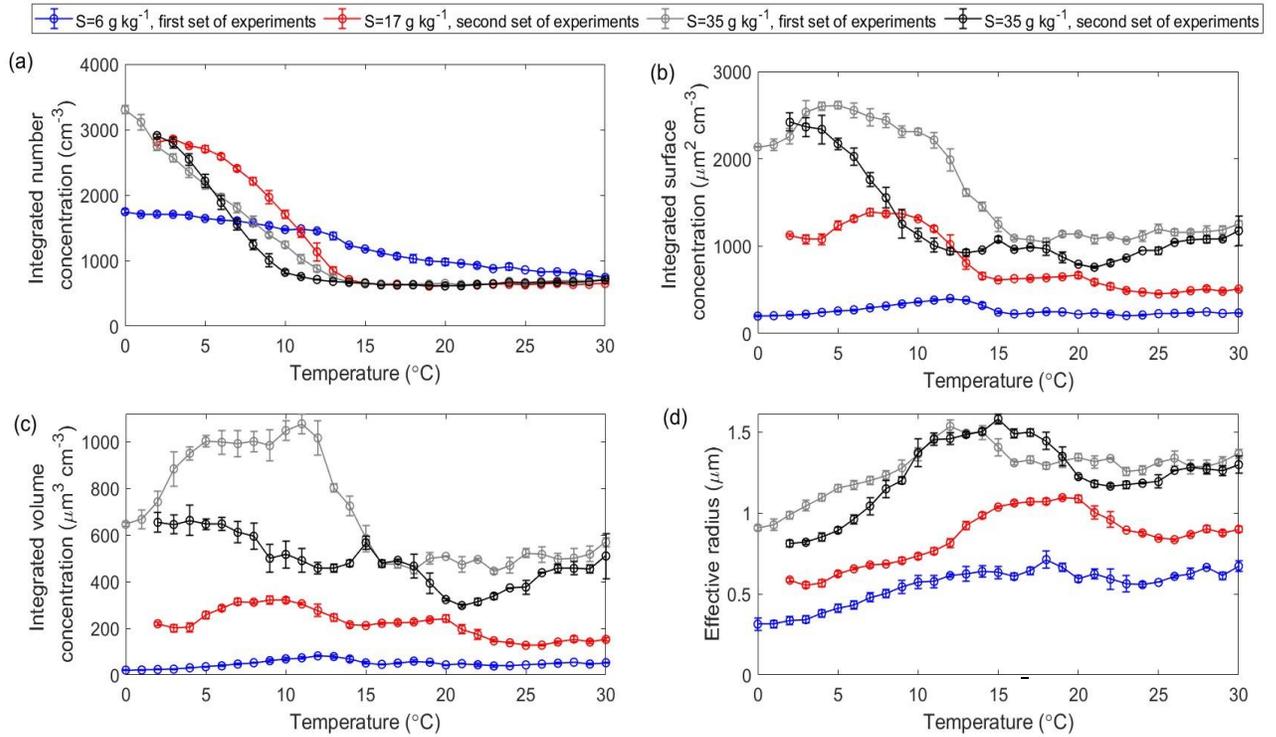
**Figure S3.** Estimated sampling efficiencies for the DMPS and OPSS (WELAS). Efficiencies were calculated with the Particle Loss Calculator (PLC) software (von der Weiden et al., 2009) based upon the densities of seawater ( $1000\text{ kg m}^{-3}$ ), and dry sea salt particles ( $2017\text{ kg m}^{-3}$ ).



**Figure S4.** The location of the modes at different salinities for (a) the number size distribution, (b) surface size distribution and (c) volume size distribution.



**Figure S5.** Comparison of particle concentrations measured by the total CPC and integrated over the combined size distribution.



**Figure S6.** Integrated concentrations of aerosol (a) number, (b) surface, (c) volume and (d) effective radius as a function of temperature at salinity 35 g kg<sup>-1</sup>, 17 g kg<sup>-1</sup> and 6 g kg<sup>-1</sup>.

Study	Simulation system	Sea water composition	Salinity range
Mårtensson et al. (2003)	Frit	Tropic Marin aquarium sea salt	0, 9.2, 33 g kg <sup>-1</sup>
Tyree et al. (2007)	Frit	Analytical grade salt mixture	1, 10, 20, 33, 70 g kg <sup>-1</sup>
Zábori et al. (2012)	Plunging jet	NaCl	0-35 g kg <sup>-1</sup> , intervalls of 3 g kg <sup>-1</sup>
Park et al. (2014)	Frit	Sigma Aldrich sea salt	2, 5, 15, 32 g kg <sup>-1</sup>
May et al. (2016)	Plunging jet	NaCl	0.05, 0.15, 0.5, 10, 35 g kg <sup>-1</sup>

**Table S1.** Overview of previous studies on the effect of salinity on sea spray production.

Type of experiment	Salinity	Temperature	Duration
First set of experiments			
Temperature ramps	35, 6 g kg <sup>-1</sup>	30-0 °C	46.5, 41.5 h
Salinity experiments	35, 30, 25, 20, 15, 10, 8, 6, 5, 4, 3, 2, 1, 0 g kg <sup>-1</sup>	20 °C	On average 2.5-3 h or over night
Second set of experiments			
Temperature ramps	35, 17 g kg <sup>-1</sup>	30-2 °C	36.5, 43 h
Salinity experiments	35, 17, 10, 9, 8, 7, 6, 5.5, 4.5 g kg <sup>-1</sup>	20 °C	On average 2.5 h or over night

**Table S2.** Overview of the salinity experiments and temperature ramps conducted during this study.