

Read me for data associated with figures in “Temperature Dependence of Sea Spray Aerosol Production” by Forestieri et al., to be submitted to Geophysical Research Letters

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Data are available as tab delimited *.txt files and as *.xlsx

General Description of Column Names within figure data files: DataType_ExperimentName

Description of Figure data files:

F01-data.txt

- Figure 1
- Contains normalized concentration or flux data as a function of temperature for new measurements from the miniMART and MART, and for similar from literature measurements or parameterizations.
- Temperatures (T or Temp) are in Celsius
- Fluxes are normalized to 20 degrees Celsius
- Studies:
 - o miniMART_NaCl_10nm = measurements from the miniMART using a 0.5 M sodium chloride solution. Concentrations are from the entire measured size distribution, starting at 10 nm. Only two points are reported, which are the results of a linear fit to the observations.
 - o miniMART_NaCl_200nm = same as miniMART_NaCl_10nm, but for particles with mobility diameters > 200 nm
 - o Z12a_SW = measurements from (Zábori *et al.*, 2012b) for Arctic ocean seawater
 - o Z12b_NaCl = measurements from (Zábori *et al.*, 2012a) for sodium chloride water
 - o S14_Dp10nm = measurements from (Salter *et al.*, 2014) for particles > 10 nm
 - o S14_Dp300nm = same as S14_Dp10nm, but for particles > 300 nm mobility diameter
 - o G14 = from (Grythe *et al.*, 2014)
 - o J11 = parameterization from (Jaeglé *et al.*, 2011)
 - o O14 = parameterization from (Ovadnevaite *et al.*, 2014)

F02-data.txt

- Figure 2
- Contains particle number concentration versus temperature for various water types and generation methods
- Temperatures (T or Temp) are in Celsius
- Particle concentrations (Np) are in #/cm³

- Studies:
 - o MART_FARS1/2 = experiments using the MART with filtered and autoclaved seawater
 - o MM_NaCl_Dp10nm = experiments using the miniMART with 0.5 M NaCl, for all particles measured with the SEMS
 - o MM_NaCl_Dp200nm = same as MM_NaCl_Dp10nm but only for particles with mobility diameters > 200 nm
 - o MM_RS1/2 = experiments using the miniMART with reef salt
 - o MM_NASW1 = experiment #1 using the miniMART with non-autoclaved sea water

F03a-data.txt

- Figure 3a
- Contains particle number-weighted size distributions measured at different Temperatures (T or Temp) for particles generated using the miniMART. Measurements were made using the scanning electrical mobility sizer with an 85-Kr source
- Diameters (Dpm) = mobility diameters in nm
- Particle concentrations (dNdlogDp) are in #/cm³
- Nomenclature:
 - o dNdlogDp_XXC = measurement at XX degrees C, where XX = 14, 13, 9, 7, 2, 0 and -1 degrees C. Note that -1 degrees C is listed as “neg1”.

F03b-data.txt

- Figure 3b
- Contains the average number-weighted size distributions measured at different Temperatures (T or Temp) for particles generated using the miniMART and the bimodal lognormal fit to the observed distribution. Measurements were made using the scanning electrical mobility sizer with an 85-Kr source
- Diameters (Dpm) = mobility diameters in nm
- Particle concentrations (dNdlogDp) are in #/cm³
- Nomenclature:
 - o dNdlogDp_obs = observations
 - o dNdlogDp_Mode1 = first mode of bimodal fit
 - o dNdlogDp_Mode2 = second mode of bimodal fit
 - o dNdlogDp_SumModes = sum of the modes from the bimodal fit

F03c-data.txt

- Figure 3c
- Contains particle number-weighted size distributions measured at different Temperatures (T or Temp) for particles generated using the MART. Measurements were made using the scanning mobility particle sizer with an 85-Kr source
- Diameters (Dpm) = mobility diameters in nm
- Particle concentrations (dNdlogDp) are in #/cm³

- Nomenclature:
 - o dN_{dlogDp_XXC} = measurement at XX degrees C, where XX = 25, 15, 10, 8, 4, or 1 degree C.

F03d-data.txt

- Figure 3d
- Contains the average number-weighted size distributions measured at different Temperatures (T or Temp) for particles generated using the MART and the bimodal lognormal fit to the observed distribution. Measurements were made using the scanning mobility particle sizer with an 85-Kr source
- Diameters (Dpm) = mobility diameters in nm
- Particle concentrations (dN_{dlogDp}) are in $\#/cm^3$
- Nomenclature:
 - o dN_{dlogDp_obs} = observations
 - o dN_{dlogDp_Mode1} = first mode of bimodal fit
 - o dN_{dlogDp_Mode2} = second mode of bimodal fit
 - o $dN_{dlogDp_SumModes}$ = sum of the modes from the bimodal fit

F04a-data.txt

- Figure 4a
- Contains time-series of particle concentrations ($\#/cm^3$) for different water types, starting from when plunging first began. Measurements are integrated number concentrations from the Scanning Electrical Mobility Sizer, operating with a 85-Kr source, with the exception of NaCl. For NaCl, the measurements are from a condensation particle counter.
- Water types:
 - o NaCl = 0.5 M sodium chloride solution
 - o RS1/RS2 = 0.5 M synthetic reef salt solutions #1 and #2
 - o NASW1-4 = non-autoclaved sea water samples from four different experiments
- Time is in hours
- Particle concentrations are in $\#/cm^3$

F04b-data.txt

- Figure 4b
- Contains mobility number-weighted size distributions measured for different water types, averaged over the entire sampling period, measured using a Scanning Electrical Mobility Sizer, operating with a 85-Kr source. The size distributions have not been corrected for transmission losses, but have been corrected for multiple charging of particles using the built-in SEMS algorithm
- Diameter (Dpm) = the mobility diameter in nm
- Size distributions (dN_{dlogDp}) = the concentration in each size bin, in $\#/cm^3$
- Water types:
 - o NaCl = 0.5 M sodium chloride solution
 - o RS1/RS2 = 0.5 M synthetic reef salt solutions #1 and #2

- NASW1-4 = non-autoclaved sea water samples from four different experiments

fS02-data.txt

- Figure S2
- Amplitudes of individual modes of the observed size distributions obtained by fitting to two lognormal modes for MART reef salt experiment (40 minute averages) and the miniMART NaCl experiment (10 minute averages).
- Temperatures (T or Temp) in degrees C
- Amplitudes in $\#/cm^3$
- Terminology:
 - T_X = temperature of X (= MART or miniMART)
 - Conc_X_Y = amplitude of X (= MART or miniMART) and Y (mode = SmallMode or LargeMode)
 - SmallMode = the small mode amplitude
 - LargeMode = the large mode amplitude

fS03-data.txt

- SSA particle number concentrations averaged over 10 minutes as a function of water temperature for four different miniMART untreated seawater experiments
- Temperature (T): degrees C
- Concentration (Np): $\#/cm^3$
- Time (Time): M/DD/YYYY
- Terminology:
 - NASW1-4 = the four different water samples, all obtained from Bodega Bay, CA

fS04-data.txt

- Figure S4
- Average size distributions from this study and from the literature for measurements made from various water types and using various generation methods, at multiple Temperatures (T or Temp)
- Terminology:
 - Dpm = mobility diameter in nm
 - dNdlogDp = number-weighted concentration ($dN/d\log Dp$), in particles/ cm^3
 - This Study (miniMART)
 - Dpm_MM, dNdlogDp_MM_14degC, dNdlogDp_MM_0degC
 - This Study (MART)
 - Dpm_MART, dNdlogDp_MART_25degC, dNdlogDp_MART_1degC
 - Martensson (2003)
 - Dpm_M03_25degC, dNdlogDp_M03_25degC, Dpm_M03_5degC, dNdlogDp_M03_5degC
 - Sellegri (2006)

- Dpm_S06_23degC, dNdlogDp_S06_23degC, Dpm_S06_4degC, dNdlogDp_S06_4degC
- Salter (2014)
 - Dpm_S14_30degC, dNdlogDp_S14_30degC, Dpm_S14_neg1degC, dNdlogDp_S14_negdegC
- Zabori (2012)
 - Dpm_Z12_15degC, dNdlogDp_Z12_15degC, Dpm_Z12_0degC, dNdlogDp_Z12_0degC
- Schwier (2017)
 - Dpm_S17, dNdlogDp_S17

fS05-data.txt

- Figure S5
- Relationship between SSA particle number concentrations and temperature for particles generated using the MART (i.e. intermittent plunging sheet; grey squares) or a continuous plunging jet that was built into the MART (orange circles). Also, experiment-averaged size distributions for the sheet and plunging jet experiments.
- Terminology:
 - Data Set 1: Number vs temperature
 - T_MART = temperature, degrees C
 - Np_Sheet = number concentration when the intermittent plunging sheet was used, #/cm³
 - Np_Jet = number concentration when the continuous plunging jet was used, #/cm³
 - Data Set 2: size distributions
 - Dp_MART = mobility diameter, nm
 - dNdlogDp_Sheet = number-weighted size distribution when the intermittent plunging sheet was used, in #/cm³
 - dNdlogDp_Jet = number-weighted size distribution when the continuous plunging jet was used, in #/cm³

Description of Raw Data files:

The measured water temperature and aerosol size distributions for each experiment (MART or miniMART and water-type) are provided as *.csv files. General naming convention for files: GenerationMethod_WaterType_rawdata.csv

- Water Types:
 - NaCl = 0.5 M sodium chloride aqueous solution
 - RS = non-filtered, non-autoclaved reef salts
 - FARS = filtered and autoclaved reef salts
 - NASW = non-autoclaved, filtered seawater from Bodega Bay
- File Contents:

- Column 1 = water temperature, degrees C
- Column 2 = Date/time associated with temperature measurement (MM/DD/YYYY HH:MM:SS)
- Column 3 = Intentionally blank
- Row 1, Column 5-125 = particle mobility diameter, in nm
- Column 4, starting at row 3 = Start time of each size distribution measurement (MM/DD/YYYY HH:MM:SS)
- Column 5-125, starting at row 3 = measured particle concentrations in each size bin, i.e. $dN/d\log D_p$, in $\#/cm^3$.

Methods Overview:

Measurements of sea spray particle size distributions were made for particles produced from either a marine aerosol reference tank (MART) (Stokes *et al.*, 2013) or mini-MART (Stokes *et al.*, 2016). General details are provided below.

The MART is a 210 L enclosed acrylic tank filled with ~ 120 L of water and generates particles using an intermittent plunging sheet, which was pumped on a 4-second-on 10-second-off duty cycle. Particles generated from the MART were sampled at a height of either 3 cm or 20 cm above the water surface through a 0.95 cm stainless steel tube. The miniMART encompasses a 17.5 L acrylic tank. The miniMART is filled with ~ 7.5 L of water, leaving ~ 10 L of head space from which particles are sampled. The particles generated from the miniMART were sampled through a 0.95 cm stainless steel tube located 2 cm above the water surface.

The flowrate of air through the MART was 6.5 LPM, with the air generated by a zero-air generator (Sabio Instruments, Model 1001). The flowrate of air through the miniMART was 0.85 LPM, with the clean air generated by passing compressed air through an activated carbon filter, a desiccant denuder, and finally a particle filter. The airflow rates into the MART and miniMART were controlled by a mass flow controller. The flow into the MART and miniMART exceeded slightly that required by the downstream instrumentation, by 0.9 LPM for the MART and 0.2 LPM for the miniMART.

The water and air temperatures for the MART experiments were controlled by placing the MART in a temperature-controlled room and by concurrent direct contact cooling of the water using a chiller coil inside of the tank. The temperature was monitored by a temperature sensor (Thermo Scientific AC150). The water and air temperatures for the miniMART experiments were controlled by placing the miniMART in a cold room and by contact cooling via two cold plates attached to the outer walls of the tank. The temperature of the cold plates was controlled with a chiller. The air and water temperatures for the miniMART experiments were monitored with two thermocouples: one in the water and the other located just above the water surface. In both MART and miniMART experiments, the water and air temperatures were set to be approximately equal.

Text S2. Continuous plunging jet experiments

The continuous plunging jet measurements were made concurrently with the plunging sheet (i.e. traditional MART operation) measurements by switching between generation methods while holding the T_w constant and using the identical source water within the MART. Jet and sheet comparison experiments were conducted such that T_w was constant while switching from sheet (traditional MART operation) and jet. That is, at each T_w , particle size distributions and

concentrations were first measured using the plunging sheet, the sheet flow was stopped, and then the plunging jet was started. The vertical water jet was produced from a 4 mm diameter stainless steel tube and fittings with the jet outlet located 20 cm above the water surface. The jet was run continuously at a flowrate of 2.15 L min^{-1} in a setup and operation following Salter *et al.* (2014).

Text S3. Source water preparation

The NaCl solution had a NaCl concentration of 0.5 M and was produced by dissolving NaCl (99% purity; Fisher Scientific) into milliQ water (18.0 M Ω and total organic carbon < 3 ppb). The reef salt (RS) experiments used an aqueous solution produced from dissolution of common reef salts (Brightwell Aquatics Inc.) in milliQ water. The reef salt was composed of the inorganic components of sea salt, including 1,290 ppm magnesium, 413 ppm calcium, 399 ppm potassium, 8 ppm strontium, in addition to NaCl. The mass concentration of dissolved salt in the reef salt solution was equivalent to the NaCl solution. The MART used autoclaved reef salt water that was filtered with 0.2 μm mesh.

For non-autoclaved seawater experiments, seawater was acquired from the Bodega Bay Marine lab. The seawater used for the miniMART experiments was collected and filtered by the Bodega Bay Marine Lab (Bodega, CA) on either 30 August 2017 or 12 September 2017 and stored in pre-cleaned plastic tubs. The seawater was sand filtered with a diameter of $\sim 30 \mu\text{m}$. The approximate chlorophyll-a concentration and salinity at the time of collection are listed in Table S2. The first experiment used water collected on 30 August 2017, with the experiment started that same day (Table S1). NASW experiments 2 through 4 all used water that was collected on 12 September 2017, but where the experiment was started at different times after collection (Table S1). One (NASW2) was started on the day the water was collected. The other two were started 15 days (NASW3) and 20 days (NASW4) after collection, and the water was stored at 4 $^{\circ}\text{C}$ after collection until use.

Text S4. Particle sizing and concentration measurements

Particle size distributions were monitored during the MART experiments using a scanning mobility particle sizer (SMPS; TSI Inc.) and aerodynamic particle sizer (APS; TSI, Inc.). For miniMART experiments, a scanning electrical mobility sizer (SEMS) was used. The air stream exiting the MART or miniMART was dried to $\text{RH} < 20\%$ prior to the size or concentration measurements by passing the air through a desiccant denuder. The SMPS is a combination of a differential mobility analyzer (DMA) and a condensation particle counter (CPC). Similarly, the SEMS is combination of a DMA and a mixing condensation particle counter (MCPC). In addition, particle number concentrations in the dried air stream were monitored with a CPC (TSI Inc.) at 5 s time resolution for both the miniMART and MART experiments (shown in Figures S6-S8).

The SMPS characterized mobility diameters ($d_{p,m}$) over a size range ($d_{p,m}$) of $10 \text{ nm} < d_{p,m} < 700 \text{ nm}$ and the APS measured the aerodynamic particle size ($d_{p,a}$) over the range $0.7 \mu\text{m} < d_{p,a} < 20 \mu\text{m}$. The SEMS characterized mobility diameters over the range $10 \text{ nm} < d_{p,m} < 1.8 \mu\text{m}$. The SEMS can scan to larger diameters than the SMPS due to differences in the design of the DMA column. The scan times of both the SEMS and SMPS were 5 minutes. The SMPS and APS distributions were merged at $d_{p,m} 700 \text{ nm}$ by converting the APS-measured aerodynamic diameters to mobility diameters assuming spherical particles and a density (ρ) of 1.8 g cm^{-3} . Here, the SEMS was modified to use a ^{85}Kr source, rather than the standard ^{210}Po source, to

bring the particles to an equilibrium charge distribution before they enter the DMA. The SMPS also used a ^{85}Kr source.

References

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