Emission fluxes of coarse-mode sea spray aerosols measured in the SOARS wind/wave tunnel facility

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1. Introduction

Coarse mode marine aerosol (1) accounts for a large fraction of the atmospheric aerosol burden, (2) represents a reaction medium for important chemical processes, (3) dominates the transport of elements like sodium and chlorine from the oceans to the continents, and (4) can strongly influence cloud and precipitation processes. Despite these important roles in the Earth system, the emission flux of coarse mode sea spray aerosols (SSA) is poorly constrained, with estimates from laboratory and field studies ranging across several orders of magnitude.

In an ongoing project, we are investigating the emission of SSA in the Scripps Ocean-Atmosphere Research Simulator (SOARS), a combined wind tunnel and wave channel capable of replicating a wide range of surface ocean conditions.

Aerosol production in SOARS occurs under wind speeds up to an equivalent U_{10} of 21 m s^{-1} and whitecap coverage of 8%.

2. Sea Spray Generation

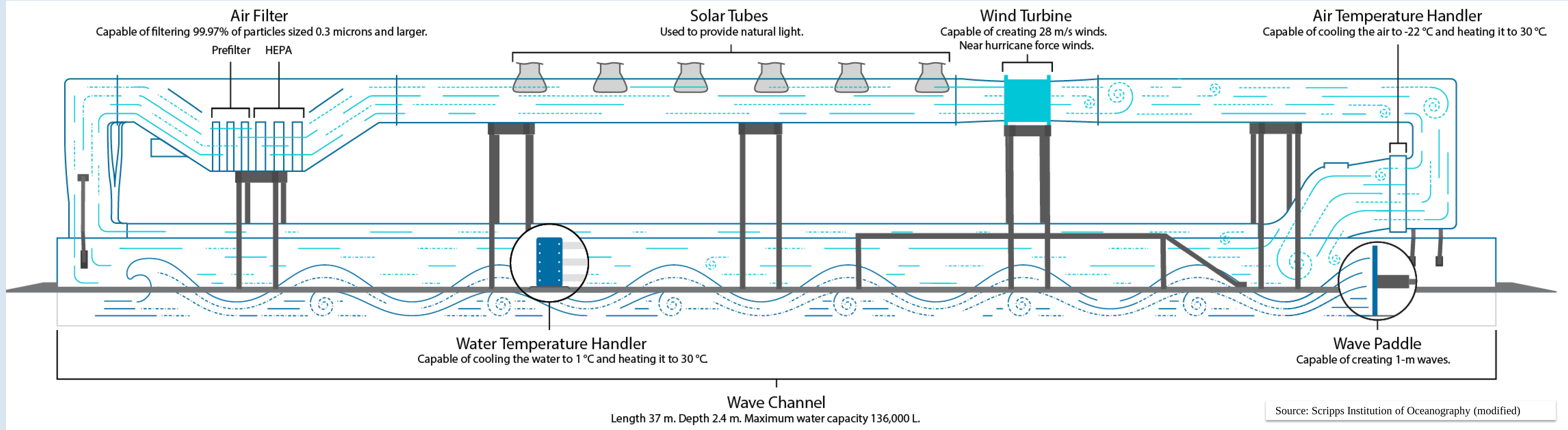

Figure 1: Schematic diagram of the Scripps Ocean-Atmosphere Research Simulator (SOARS)

From May 2022 to the present, we have been making measurements of sea spray aerosol size distributions in the SOARS wind/wave tunnel at Scripps. We used an SMPS (TSI 3938), an APS (TSI 3321), and an OPS (TSI 3330). Only examples of the results from the OPS will be discussed here. For the other results, **see the eLightning A41T-05 by Leibensperger et al. on Thursday at 8:42!**

3. Data Analysis

For the sea spray production experiments, we selected wind speed and wave patterns that were intended to produce a constant rate of sea spray production over periods of up to several hours. Before starting wind and waves, the aerosol concentration in the headspace was reduced to near zero by circulating the air through a set of HEPA and activated charcoal filters. The wind and waves were allowed to run until the aerosol concentrations reached a steady state in the head space. After an appropriate sampling period, the waves were turned off and the particle concentrations were allowed to decay while the wind was kept on. A typical run is shown in Fig. 2

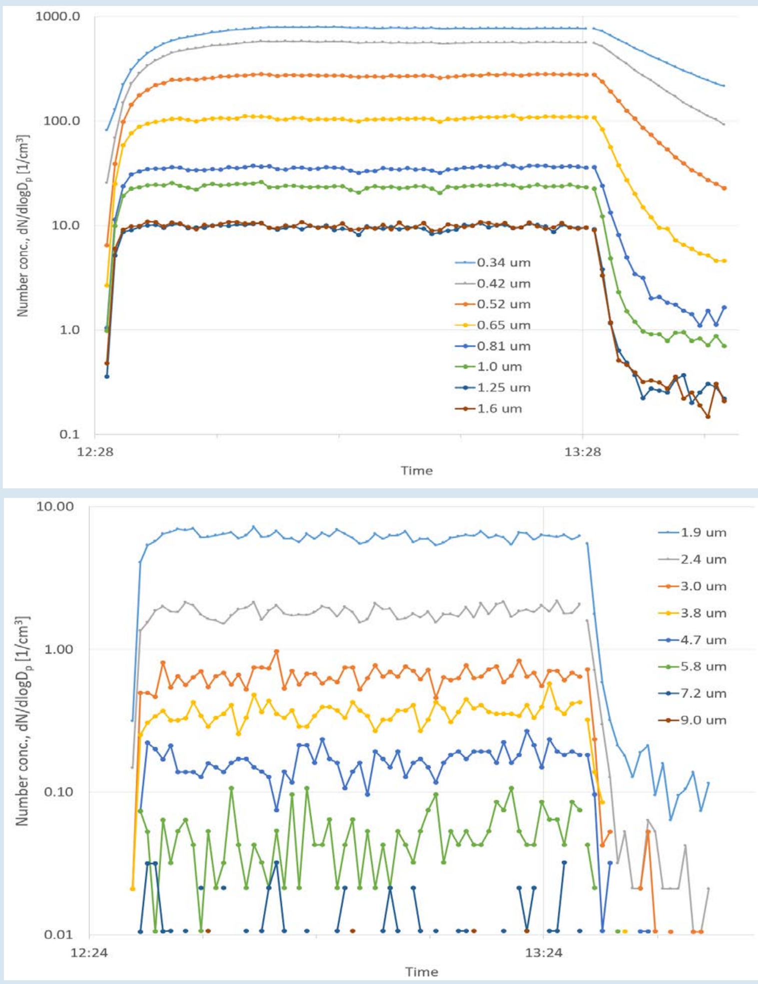

Figure 2: Time series of particle concentrations

How can production rates be obtained?

Assuming SOARS to be a well stirred reactor, we can model the concentration C_i in size-bin i using a constant production rate, P_i, and a first-order loss rate, L_i, with a loss rate constant, k_i.

$$\mathbf{dC_i/dt = P_i - L_i = P_i - k_i{*}C_i}$$

To obtain P_i, we can use the following approaches

1) By fitting the entire production run to $dC_i/dt = P_i - k_i{*}C_i$, where we leave both P_i and k_i as free fitting parameters
2) By fitting the entire production run to $dC_i/dt = P_i - k_i{*}C_i$, but prescribing k_i, obtained from the loss rate after the waves stop, and leaving only P_i as free parameter
3) In steady state, $dC_i/dt = 0$, and thus $P_i = k_i{*}C_{i,SS}$, where $C_{i,SS}$ is the steady state concentration, and k_i is obtained by fitting the decay at the end of the run, as in (2).
4) At the beginning of the run, C_i is very small, and thus the first-order loss can be neglected, and $P_i \cong dC_i/dt$

To obtain the actual flux in dimensions of $length^{-2}$ $length^{-3}$ $time^{-1}$, e.g., particles cm^{-3} m^{-2} sec^{-1}, we need to divide by the whitecap surface area.

We get the size distribution of the produced particles simply recognizing that, if there were no loss ($L_i = 0$), $C_i(t) = P_i{*}t$, and thus $C_i \sim P_i$, i.e., the normalized size distributions of concentration and production rate are identical.

4. Results

Method 1 [fit with free P and k]: Fundamentally, this should be the best method since it includes all information from the run, but above ~2 µm, there are too few points to constrain the fit.

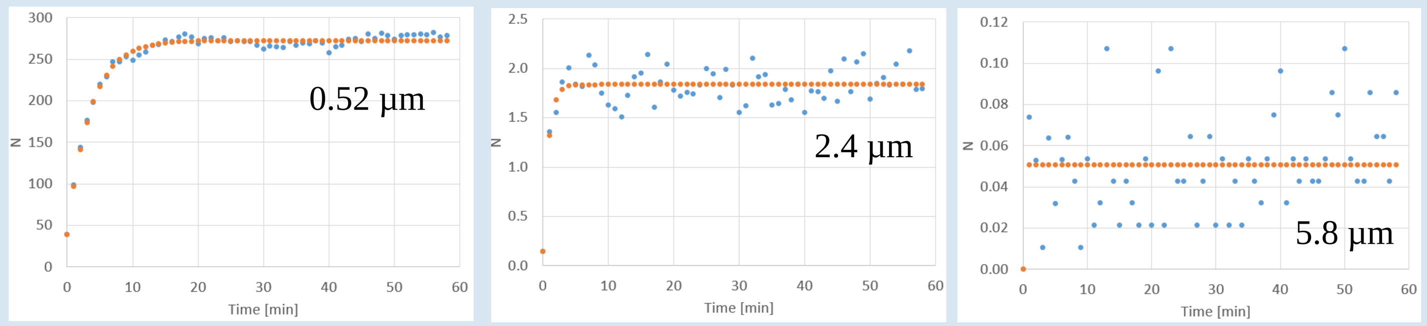

Figure 3: Fits with Method 1 to three different size bins

Method 2 [steady state and decay constant]: The decay constants obtained after shutting off the waves are probably underestimates, since turbulence is reduced, thus this probably yields underestimates.

Method 3 [fit to initial rate of increase]: Suitable only for the smallest sizes, and even there, there are very few points to constrain the fit.

Method 4 [fit with free P and prescribed k from decay at end of run]: Same problem as with (2): the decay constants obtained after shutting off the waves are probably underestimates, since turbulence is reduced.

Table 1: Production rates from different fit methods

Diameter	Steady state concentration	Loss rate constant	Half-life	Production Rates from: Fit with free P and K	Production Rates from: From steady state and k	Production Rates from: From initial production	"Best Values"
µm	cm^{-1}	min^{-1}	min	cm^{-3} min^{-1}			
0.34	776	0.18	3.9	138	137	89.1	138.2
0.42	565	0.20	3.4	115	115	79.4	115.7
0.52	273	0.29	2.4	78.4	78.5	52.6	78.4
0.65	106	0.50	1.4	52.9	52.8	33.6	52.9
0.81	35.4	0.55	1.3	19.5	19.4	11.3	19.5
1.01	23.7	0.72	1.0	17.2	17.2	9.2	17.2
1.25	9.6	1.04	0.7	8.6	10.0	4.15	10.0
1.6	10.0	1.01	0.7	10.1	10.1	4.30	10.1
1.9	6.24	1.12	0.6	6.3	6.98	3.76	6.98
2.4	1.84	1.20	0.6	2.2	2.21	1.12	2.21
3.0	0.66	1.42	0.5	0.6	0.94	0.47	0.94
3.8	0.37	1.4	0.5		0.51	0.23	0.51
4.7	0.17	1.4	0.5		0.24	0.07	0.24
5.8	0.051	1.4	0.5		0.072	0.074	0.072
7.2	0.008	1.4	0.5		0.011	0.011	0.011
9.02	0.001	1.4	0.5		0.001		0.001

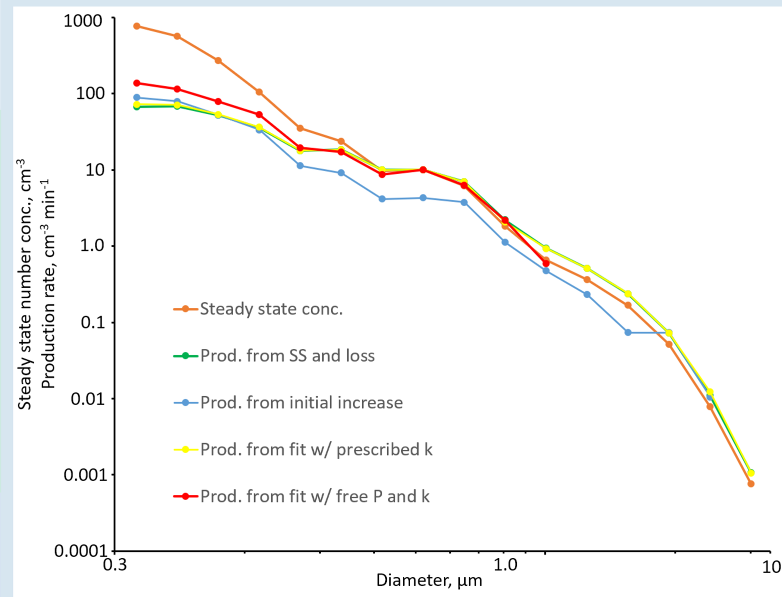

Figure 4: Steady state concentration and production rates from different methods

- Concentrations and production rates range over ~6 orders of magnitude
- Half-lives in the tunnel vary from several minutes to <1 min
- Different analysis methods yield reasonable agreement for smaller particles, but diverge for larger ones
- Poor counting statistics and low time resolution limit the ability to assess production for the larger particles

5. Summary and Conclusions

- **The SOARS facility is suitable for generation of sea spray aerosol reflecting a wide variety of environmental conditions**
- **Different approaches to analyzing the resulting data have been explored and evaluated**
- **Fitting a differential equation to obtain the size-dependent sea spray particle production rate provides the most robust data, but is poorly constrained at particle diameters greater than about 2 µm**
- **For larger particles, only relatively rough production rate estimates can be obtained using steady state concentrations and loss rate constants**

Publication

- **This poster is published in the ESS Open Archive and can be cited as**
- **DOI: 10.22541/essoar.170158291.12831376/v2**

Funding

- Max Planck Society of Germany
- National Science Foundation through the NSF Center for Aerosol Impacts on Chemistry of the Environment (CAICE), CHE-1801971.

Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the NSF.