Supplementary Information: Enhancement of Surfactants in Nanoparticles Produced by an Electrospray Aerosol Generator

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This supplementary information document describes a way to estimate the weight fraction of SDS in the SDS/NaCl particles based the measured *GFs*. In view of multiple assumptions, the formulas listed here should be treated as ***approximate***. However, they are expected to be more accurate compared to the Zdanovskii−Stokes−Robinson (ZSR) model (Stokes and Robinson 1966; Svenningsson et al. 2006; Varutbangkul et al. 2006) described in the main text.

Following NaCl deliquescence and further hygroscopic growth, a mixed SDS/NaCl nanoparticle can be viewed as an aqueous droplet containing dissolved NaCl coated with an SDS film. Even though SDS does not take up water by itself to a measurable extent (Harmon et al. 2010), it is expected to lower the surface tension of the droplet and affect the *GF* indirectly through the Kelvin effect. Furthermore, as SDS has non-zero solubility in solution with NaCl, the presence of dissolved SDS in the aqueous solution changes the water activity. We can correct for the Kelvin effect using the following equation:

$$\frac{RH}{100} = a_{aq}(x) \cdot \exp\left(\frac{4M_w \sigma_{aq}(x)}{RT\rho_w d_{m,wet}(RH)}\right) \tag{S1}$$

In Equation (S1), x is the weight percent of NaCl in the aqueous phase (mass of NaCl relative to the mass of the solution expressed in percent), M_w is the molecular weight of water, and ρ_w is the

density of water. We take the dependence of the activity of water in the aqueous phase (a_{aq}) on x from (Tang et al. 1997) ($C_0 = 1$; $C_1 = -6.366 \times 10^{-3}$; $C_2 = 8.624 \times 10^{-5}$; $C_3 = -1.158 \times 10^{-5}$; $C_4 = 1.518 \times 10^{-7}$).

$$a_{aq}(x) = \sum_{i=0}^{4} C_i x^i \tag{S2}$$

We assume that the amount of SDS dissolved in the aqueous phase is small relative to that of NaCl and, therefore, the presence of SDS does not significantly change a_{aq}. The x-dependent values of the surface tension, $\sigma_{aq}(x)$, for pure NaCl solution is taken from (Pruppacher and Klett 1997).

$$\sigma_{aq}(x) = \left(0.072 + \frac{0.029x}{100 - x} \right) \frac{N}{m} \tag{S3}$$

The x-dependence of the surface tension of an SDS-coated solution of NaCl is not known, and a value 0.040 N m^{-1} is adopted, which is appropriate for an SDS-coated water surface (Iyota and Krastev 2009).

Before the measured GF can be converted into ω_{NaCl} and ω_{SDS}, we have to make additional assumptions about the shape of the initial particle before it was exposed to water vapor. The most straightforward case to consider is that of a spherical NaCl core surrounded by a uniform shell of SDS ("spherical model"). The calculations are done iteratively. A certain guessed value of ω_{NaCl} is assumed, and the corresponding ε_{NaCl} value is calculated

$$\varepsilon_{NaCl} = \frac{\omega_{NaCl} \cdot \rho_{SDS}}{\rho_{NaCl} - \omega_{NaCl} \cdot \left(\rho_{NaCl} - \rho_{SDS} \right)} \tag{S4}$$

using the bulk densities of $\rho_{NaCl} = 2163$ kg m^{-3} and $\rho_{SDS} = 1010$ kg m^{-3}. The diameter of the NaCl core in the initial dry particle is then

$$d_{core,dry} = d_{m,dry} \cdot \left(\varepsilon_{NaCl} \right)^{1/3} \tag{S5}$$

After the particle interacts with water vapor, the NaCl core dissolves and increases in size by the GF of the NaCl core:

$$GF_{core} = \frac{d_{core,wet}}{d_{core,dry}} = \left(\frac{100 \cdot \rho_{NaCl}}{x \cdot \rho_{aq}(x)} \right)^{1/3} \tag{S6}$$

The density of the NaCl solution $\rho_{aq}(x)$ is taken from (Tang et al. 1997) ($A_0 = 997.1$; $A_1 = 7.41$; $C_2 = -3.741 \times 10^{-2}$; $A_3 = 2.525 \times 10^{-3}$; $A_4 = -2.060 \times 10^{-5}$).

$$\rho_{aq}(x) = \left(\sum_{i=0}^{4} A_i x^i\right) \frac{kg}{m^3} \tag{S7}$$

Once again, we are making an assumption that SDS does not significantly affect the density of the NaCl solution.

For a given assumed value of ω_{NaCl}, the above system of equations has a unique solution for x which can be found numerically. This makes it possible to predict $GF_{calculated}$,

$$GF_{calculated} = \left(1 + \varepsilon_{NaCl}\left(GF_{core}^3 - 1\right)\right)^{1/3} \tag{S8}$$

and compare it to the experimentally measured GF value. The trial ω_{NaCl} is adjusted until the experimental GF is matched to within 0.001, with the conversion typically achieved after 3-4 iterations. The resulting values of ω_{NaCl} are shown in Figures 3b and 4b of the main text.

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