How surfactants affect cloud droplet activation?

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ABSTRACT

Atmospheric aerosols usually consist of inorganic salts and organic substances, where a significant part is *surfactants*, that is, amphiphilic molecules containing a hydrophilic and a hydrophobic part. Due to their amphiphilic character, surfactants preferentially adsorb at the surface of liquids lowering the surface tension of the interfaces, thus affecting processes, such as droplet coalescence, development of precipitation and (ultimately) cloud lifetime. We created a numerical model of cloud droplet formation with a presence of surfactant and salt. The model is based on the Lagrangian particle-based microphysics scheme Super-Droplet Method (SDM) [1]. By means of the SDM, we provided an evidence that surfactants influence cloud formation.

Super-droplet method

The super-droplet method (SDM) was created to perform an accurate simulation of large aggregates of droplets [1]. Each super-droplet represents a multiple number of droplets with the same attributes and position, and the multiplicity is denoted by the positive integer $\xi_i(t)$, which can be different in each super-droplet and time-dependent (due to coalescence process). In other words, a super-droplet is a kind of coarse-grained view of droplets both in real space and attribute space.

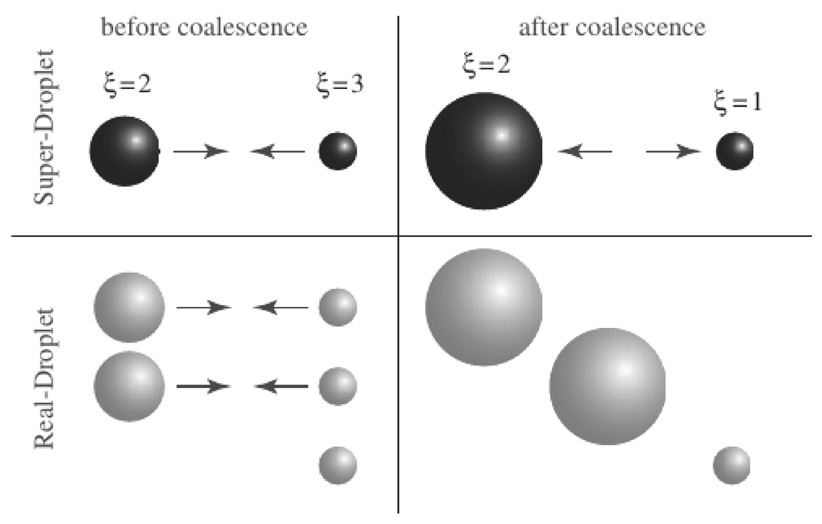


Figure 1. Schematic view of the coalescence of super-droplets. Two super-droplets with multiplicity 2 and 3 undergo coalescence (upper left panel). This represents the coalescence of two droplet pairs (lower panels). As a result, the super-droplet with multiplicity 2 becomes larger and the multiplicity of the other super-droplet decreases 3 → 1 (upper right panel).

Influence of surfactants

We consider an aqueous droplet with an (outer) radius r that can be divided into a surface monolayer of thickness δ and an interior (bulk) with radius $r - \delta$ and apply the following relation between droplet surface tension σ, as a function of bulk composition, and surface composition $\boldsymbol{x}^s = (x_1^s, x_2^s, \ldots)$ [2]:

$$\sigma(\boldsymbol{x}^b, T) = \frac{\sum_i \sigma_i v_i x_i^s}{\sum_i v_i x_i^s}, \qquad (1)$$

where $\boldsymbol{x}^b = (x_1^b, x_2^b, \ldots)$ are the bulk mole fractions $n_i^b / \Sigma_j n_j^b$ of each species i in the droplet, x_i^s are the corresponding surface mole fractions, and v_i and σ_i are the molecular volumes and surface tensions of each pure component i. n_i^s, n_i^b, and $n_i^t = n_i^s + n_i^b$ are the number of particles for ith component, for the surface, bulk, and the whole droplet, respectively. The total amounts of molecules in the monolayer n_i^s are evaluated from the volume of the monolayer, $V^s = 4\pi[r^3 - (r - \delta)^3]/3$, where the thickness δ of the surface monolayer is given from the equation:

$$\delta = \left(\frac{6}{\pi} \sum_i v_i x_i^s\right)^{1/3}.$$

Considering three mole fractions: water (subscript "1"), salt (subscript "2"), and surfactant (subscript "3"), equation (1) reduces to:

$$\sigma(x_1^b, x_2^b, x_3^b) = \frac{\sigma_1 v_1 x_1^s + \sigma_2 v_2 x_2^s + \sigma_3 v_3 x_3^s}{v_1 x_1^s + v_2 x_2^s + v_3 x_3^s}.$$

Surfactant partitioning:

$$n_1^s = \frac{V^s - n_3^s v_3}{v_1 + n_2^t v_2 / n_1^t}, \quad n_2^s = n_1^s \frac{n_2^t}{n_1^t}, \quad n_3^s = min(n_3^t, V^s / v_3),$$

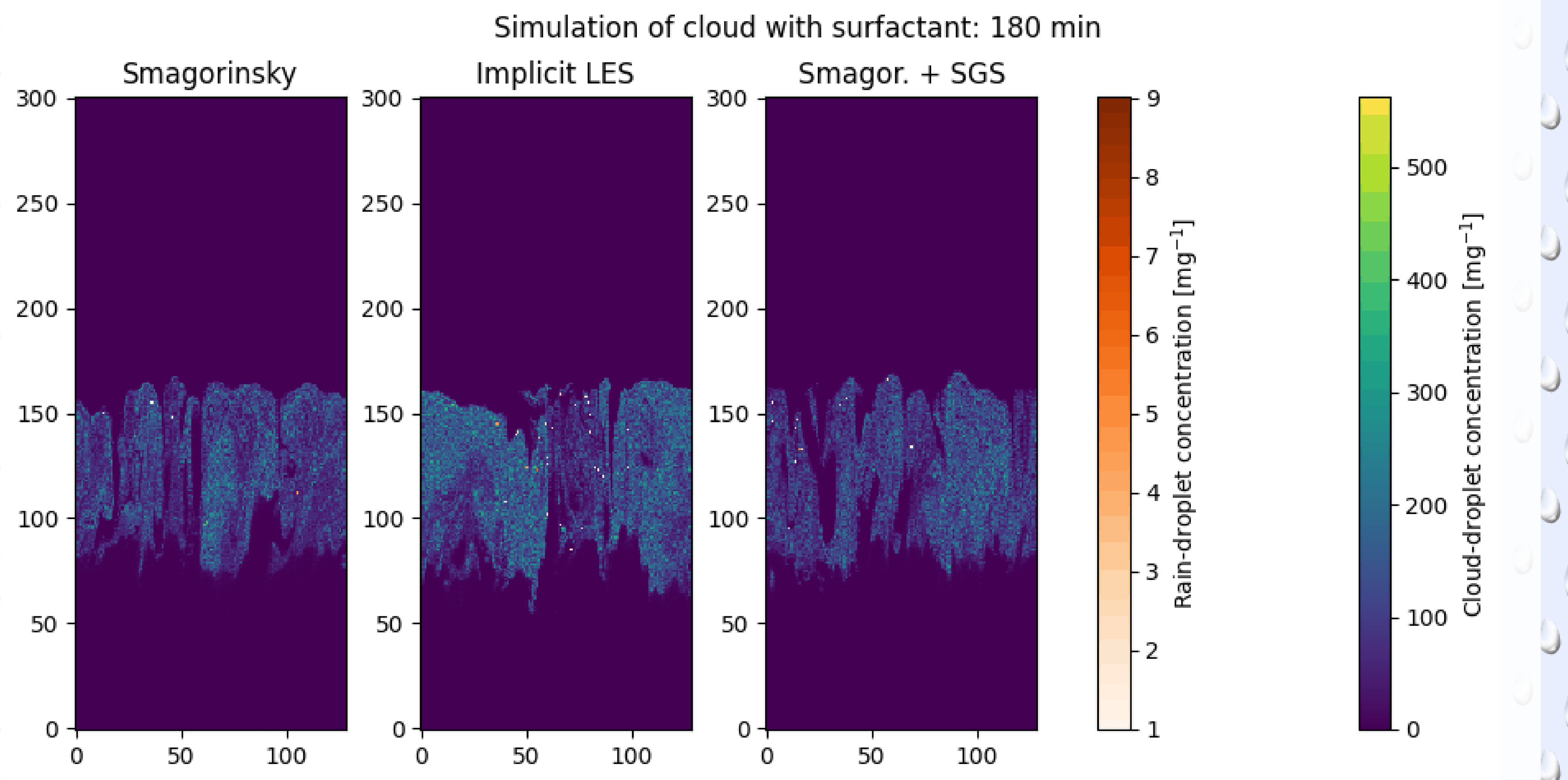
where

$$n_1^t = \frac{4\pi}{3} \cdot \frac{(r^3 - r_d^3)}{v_1}, \quad n_2^t = (1 - c_{32}) \cdot \frac{4\pi}{3} \cdot \frac{r_d^3}{v_2}, \quad n_3^t = c_{32} \cdot \frac{4\pi}{3} \cdot \frac{r_d^3}{v_3},$$

$c_{32} = n_3^t v_3 / (n_2^t v_2 + n_3^t v_3)$ is volume fraction of surfactant in atmospheric surfactant + salt particles, and r_d is dry radius of a superdroplet.

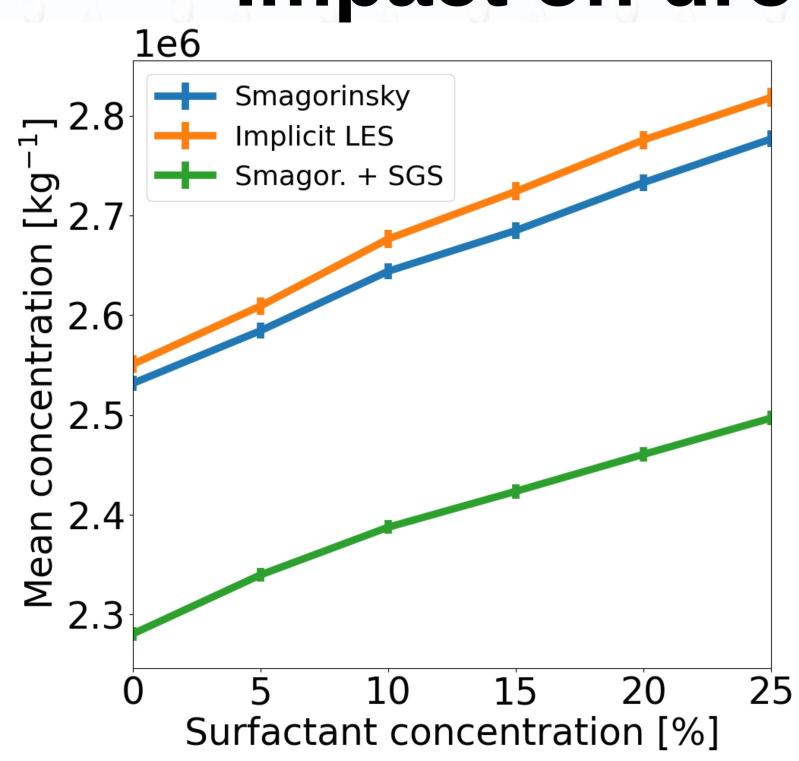
Simulations

The calculations for the SDM took place by proper modifications of the libmpdata++, libcloudph++, and UWLCM libraries [3].



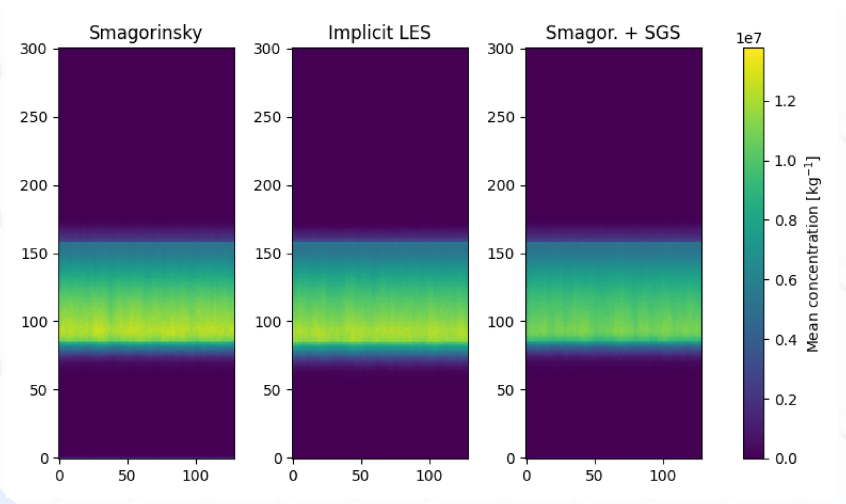
Cloud-droplet (purple–yellow scale) and rain-droplet (white–red scale) concentration for the snapshot of the SDM simulation with 25% of SDS surfactant (mass of surfactant in the droplet divided by the collective mass of surfactant and salt), for three different modelling approaches.

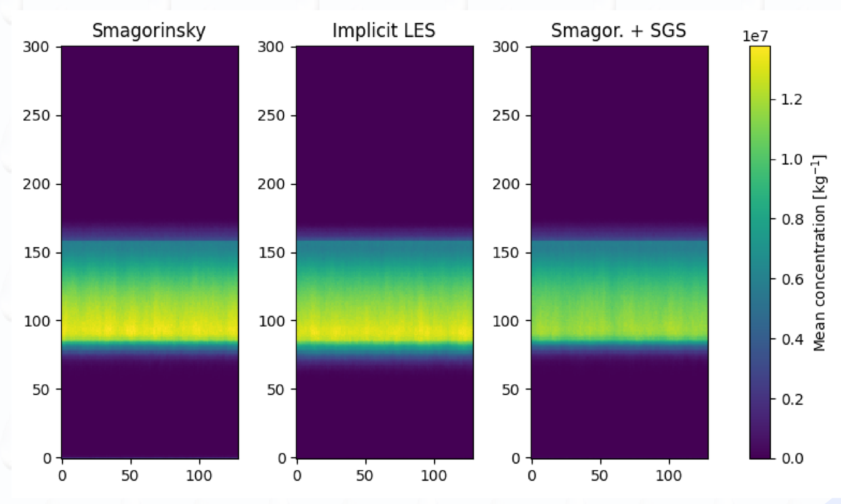
Impact on droplet activation



Left: Number of activated droplets per kilogram of dry air vs. surfactant concentration (mass of surfactant in the droplet divided by the collective mass of surfactant and salt), averaged over 100 6-hour simulations, for three different modelling approaches.

Bottom: Maps of the number of activated droplets per kilogram of dry air for the whole simulation grid, averaged over 100 simulations for the same sets of the model parameters and over 6-hour time of each simulation, for three different modelling approaches. On the left: reference model (without surfactant). On the right: model with 25% of SDS surfactant.





CONCLUSIONS

- Surfactants facilitate cloud-formation process.
- Concentration and size of activated droplets increase with growing surfactant concentration.
- The effect is independent of a modelling approach used.

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