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Laboratory study of the heterogeneous ice nucleation on black-carbon-containing aerosol

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Supplementary Material

1. Activated fraction

A representative activated fraction of 100 nm and 800 nm Monarch900 (Cabot) is plotted vs. the supersaturation over ice (Fig. S1). The 1% AF was selected as the threshold for activation onset.

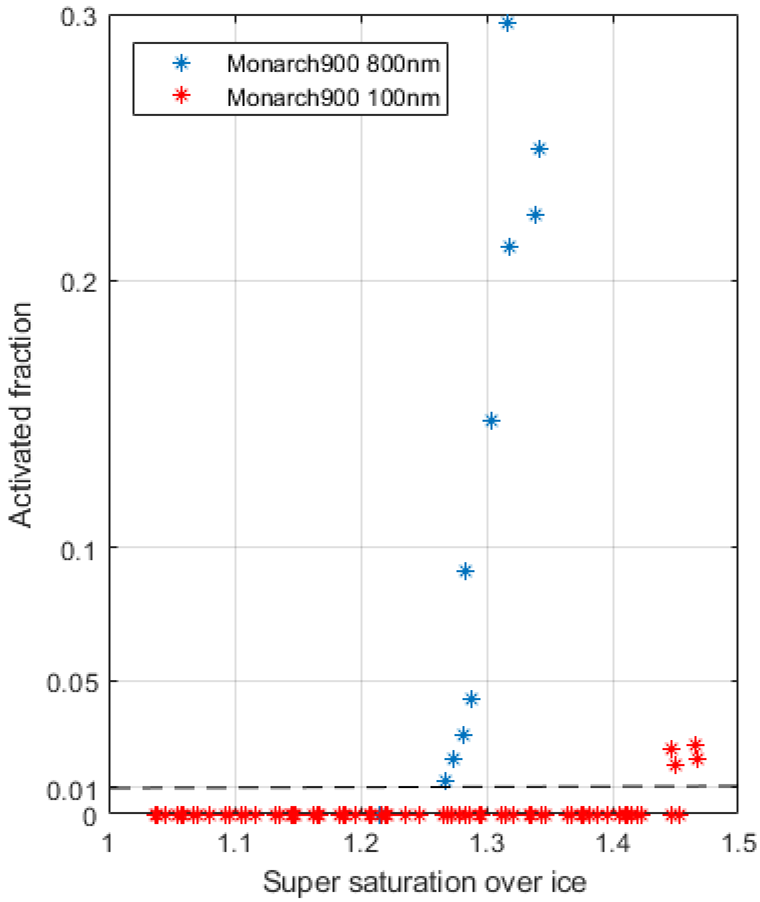

Figure S1. Activation threshold for Monarch 900 (100 nm and 800 nm).

2. Effective surface area

For the calculation of effective surface area we assumed a spherical volume of the agglomerate and a stacked structure of spherule layers as illustrated in Fig.

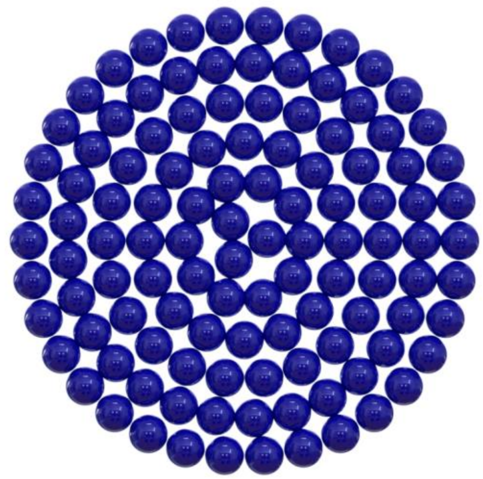

Figure S2. Illustration of the cross section of the simplified geometric structure we use to calculate the effective surface area.

For a spherule radius derived from SEM measurements, we calculated the single spherule volume. Next, we calculated the number of spherules in each of the stacked layers of the spherical 800 nm agglomerate. For each layer we calculated the total effective surface area by summing all the individual surface areas of all the spherules in the layer.

Table S1: An example of effective surface area calculations

	Ds [nm]	V_{sphere} [nm^3]	S_{area} [nm^2]	$N_{spherule}$ #
single spherules	70	1.80E+05	1.50E+04	1
outer shell of agglomerate	800	2.70E+08	2.00E+06	131
2nd inner shell of agglomerate	660	1.50E+08	1.40E+06	89
3rd inner shell of agglomerate	520	7.4E+07	8.5E+05	55
4th inner shell of agglomerate	380	2.9E+07	4.5E+05	29
5th inner shell of agglomerate	240	7.2E+06	1.8E+05	12
6th inner shell of agglomerate	100	5.2E+05	3.1E+04	2

In the example shown in Table S1, we can see that once the surface area of the inner layers is taken into account, the calculated effective surface area of the agglomerate can grow up to a factor of 3 (Eq. S1) in comparison to the outer shell alone, for spherule sizes of tens of nanometers.

$$\frac{\sum_1^6 \sum_1^N S_{ij}}{\sum_1^N S_i} \leq 3 \qquad \text{Eq. S1}$$

For a particle of 100 nm in diameter the effective surface area will be on the order of ~10^4 m^{-2} , while for a particle diameter of 800 nm the effective surface area is ~10^6 m^{-2}, therefore assuming the same activated fraction of 0.01 (defined as the onset of IN), the active site density for smaller diameters will need to be at least 2 orders of magnitude higher. Therefore, the isoline will pass closer to the homogeneous freezing line at higher super saturation.

3. Coatings

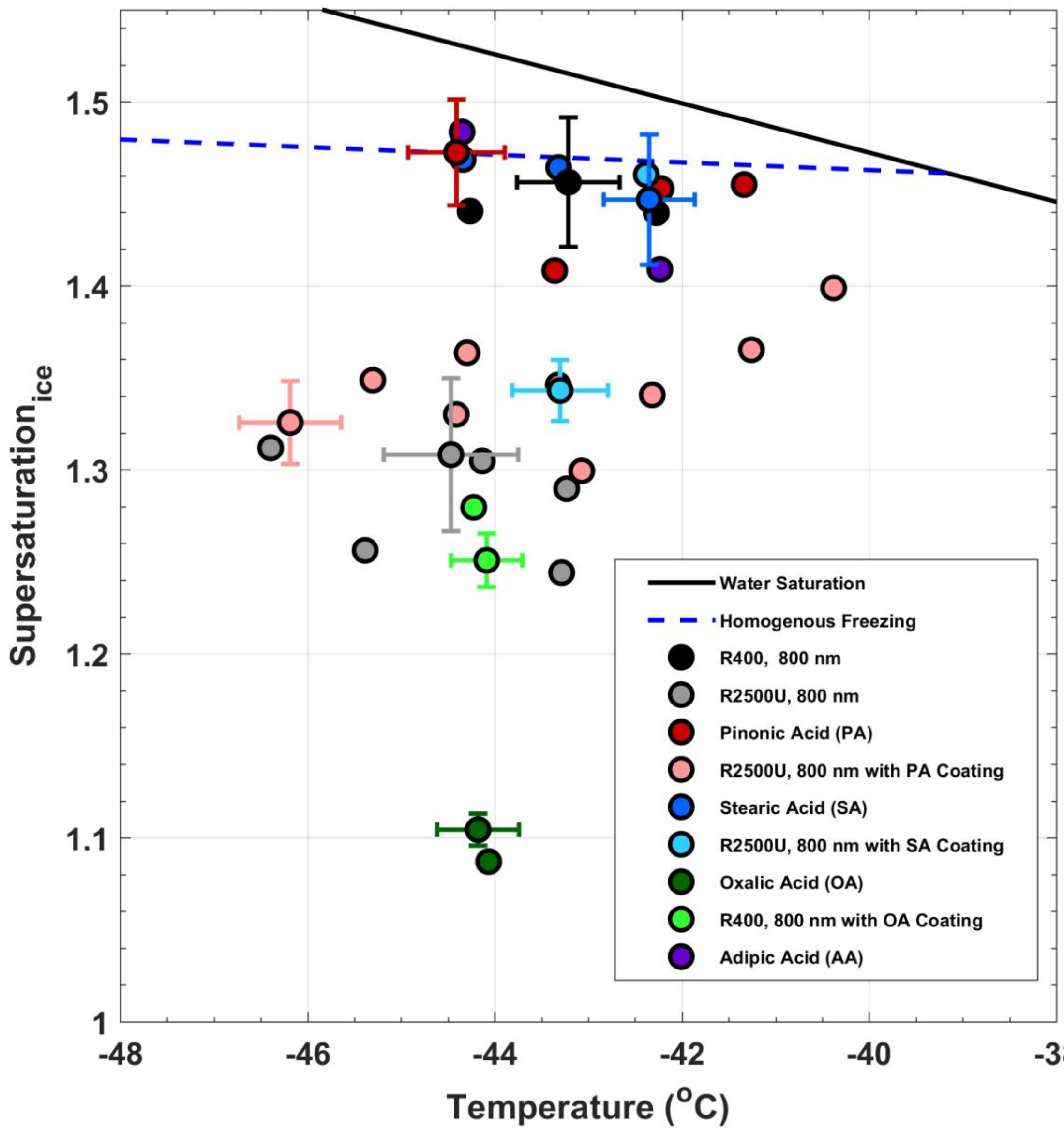

Figure S3. Modification of ice nucleation onset on BC particles by organic coating, summary plot showing homogeneous and heterogeneous ice nucleation onset of pure organic compounds, pure BC, and coated BC.