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Size-dependent particle activation properties in fog during the ParisFog 2012/13 field campaign

E. Hammer et al.

Correspondence to: M. Gysel (martin.gysel@psi.ch)

S 1 Potential method artefacts caused by in-fog processes

Potential artefacts in the lower estimate of the dry activation diameter (D_{act}^{lower}) and the corresponding upper estimate of the effective peak supersaturation (SS_{peak}^{upper}) arising from effects on in-fog processes on the particle size distribution are described here. The total, interstitial and fog droplet residual dry particle number size distributions are shown in Fig. 1 for fog event F9, along with the corresponding activation curve. Based on this representative example fog event, the potential effects of different types of in-fog processing on the inferred dry activation diameter are determined compared to the base case without in-fog.

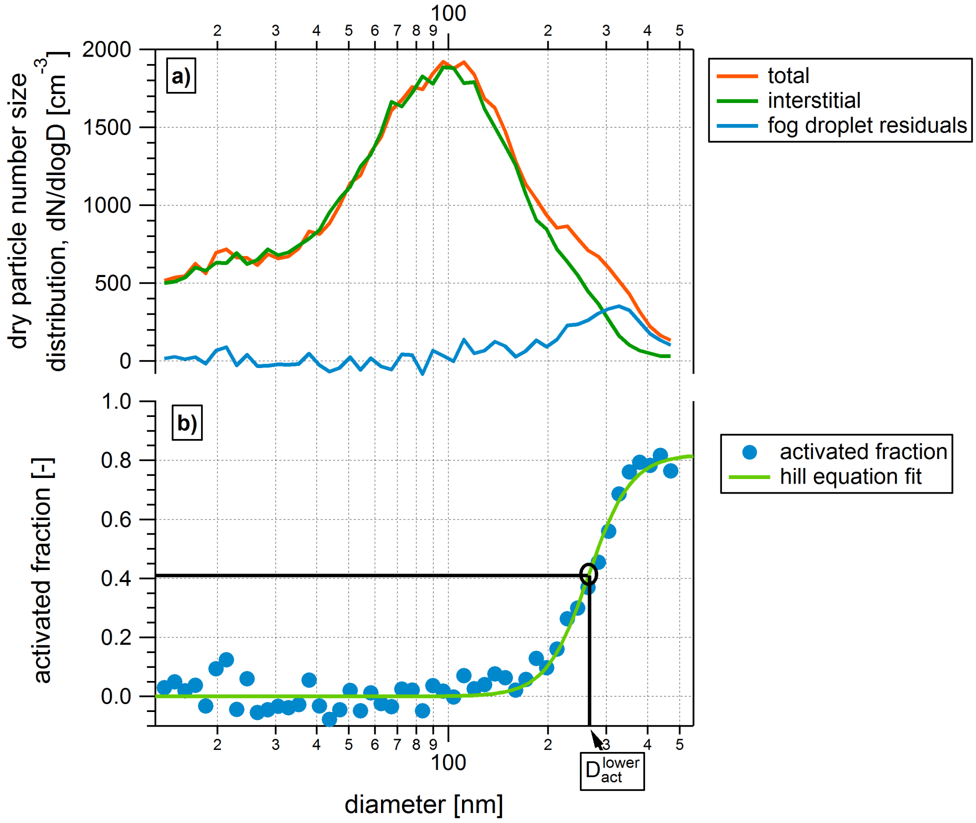

Figure 1: For the fog event F9 starting on 12 November 2012: **(a)** median total (red), interstitial (green), fog droplet residual (blue) dry particle number size distribution, and **(b)** calculated activated fraction (blue dots) with hill equation fit (green). In **(b)** the vertical black line indicates the inferred D_{act}^{lower}, which corresponds to the diameter at half-rise of the hill equation fit (marked by the black circle)

S 1.1 Entrainment

Entrainment is the process when subsaturated air is mixed into a fog. Here we assume that the total dry particle number size distributions in the fog and entrained air are equal. Thus the total size distribution will not be altered by the entrainment. We further assume that no activation of entrained particles occurs. The interstitial size distribution after entrainment then becomes the weighted mean of x times the original interstitial size distribution and $(1 - x)$ times the total size distribution. Consequently, the residual droplet size distribution after entrainment becomes x times the original droplet size distribution. This results a simple scaling of the activation curve by the factor x, as shown in Fig. 2 for $x = 0.2$. The $D_{\text{act}}^{\text{lower}}$ is not altered by this process, i.e. entrainment does not introduce an artefact.

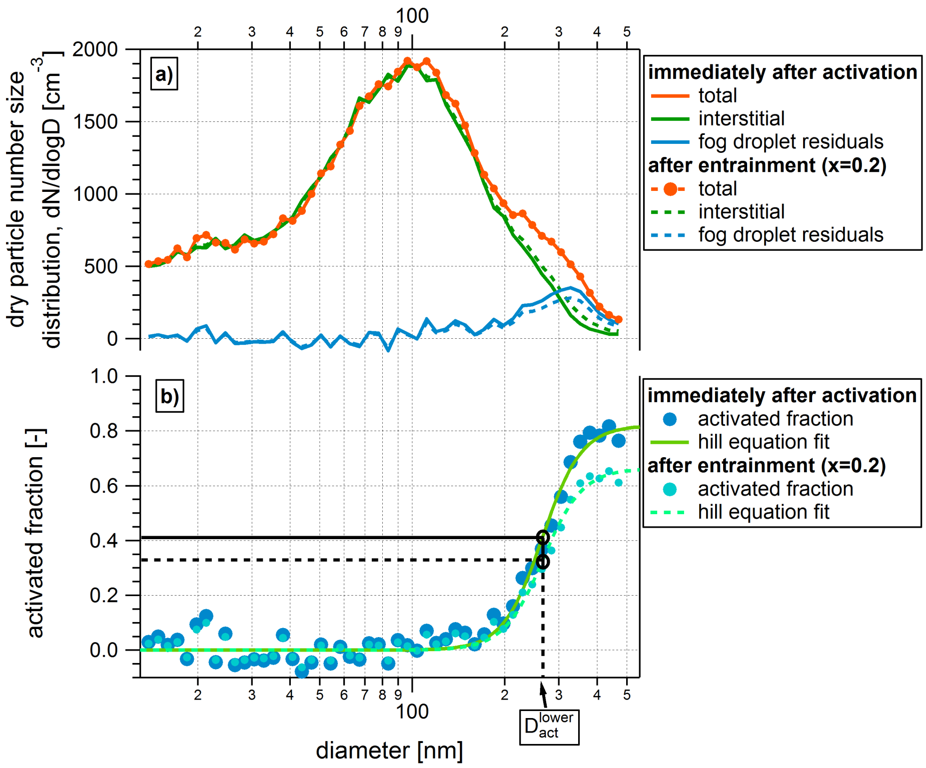

Figure 2: Same as Fig. 1 but additionally showing the size distributions resulting after entrainment.

S 1.2 Sedimentation of fog droplets

Activated fog droplets can be removed from the fog through sedimentation. We simulate this effect by scaling down the fog droplet residuals size distribution, while the interstitial aerosol remains unchanged. The processed total aerosol size distribution is obtained as the sum of the processed interstitial and fog droplet size distributions. Sedimentation of 50 % of the fog droplets increases the D_{act}^{lower} by $\sim 8\,\%$ in this example.

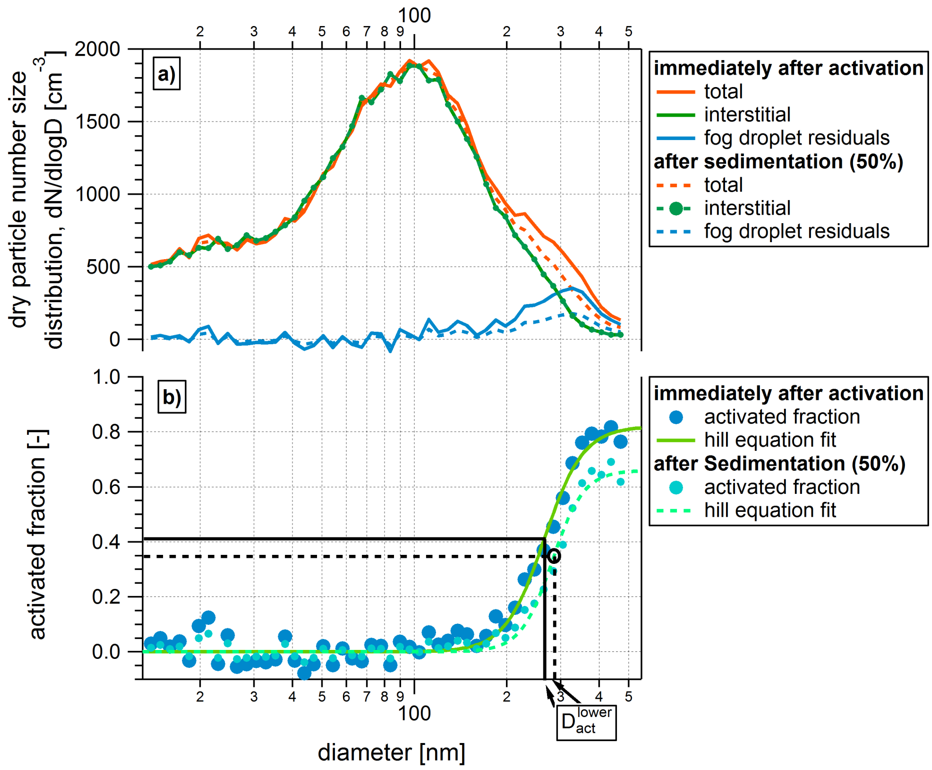

Figure 3: Same as Fig. 1 but additionally showing the size distributions resulting after fog droplet sedimentation.

S 1.3 In-cloud scavenging

The process when interstitial aerosol particles collide with fog droplets and are captured is called in-cloud scavenging. In-cloud scavenging reduces the number of interstitial particles, while the number of droplet residual particles remains unchanged. We simulate the effect of in-cloud scavenging by reducing the interstitial aerosol size distribution by constant factor of 0.8 (20 % scavenging) across all sizes, thereby ignoring the potential size dependence of the collision probability. The processed droplet residual size distribution is assumed to remain unchanged, as the addition of a small interstitial particle to a larger CCN has very little effect on the droplet residual size. The processed total aerosol size distribution is obtained as the sum of the processed interstitial and droplet residual size distributions. Figure 4 shows that 20 % scavenging reduces the measured D_{act}^{lower} by $\sim 5\,\%$.

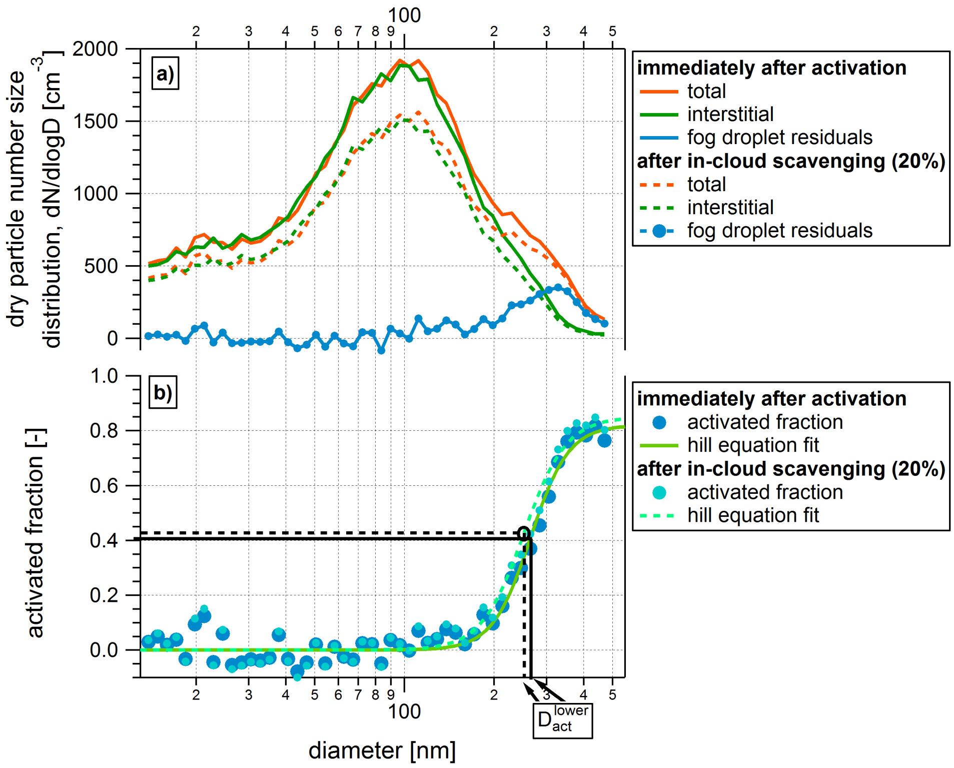

Figure 4: Same as Fig. 1 but with additionally showing the size distributions resulting after in-cloud scavenging of interstitial particles.

S 1.4 Fog droplet processing

Fog droplets can acquire additional particulate matter through different types of droplet processing, including condensation of semi-volatile aerosol components or aqueous phase heterogeneous formation of secondary aerosol within the droplet. We simulate the droplet processing as a 50 % increase of the fog droplet diameters (> 3 times the mass), while the interstitial particles remain unchanged. The processed total size distribution is obtained as the sum of the processed interstitial and droplet residual particle size distributions. Figure 5 shows that such droplet processing increases the measured $D_{\mathrm{act}}^{\mathrm{lower}}$ by ∼ 16 % in our example. The top panel further shows how the droplet processing can lead to the Hoppel minimum in the processed size distribution in the range of the dry activation diameter.

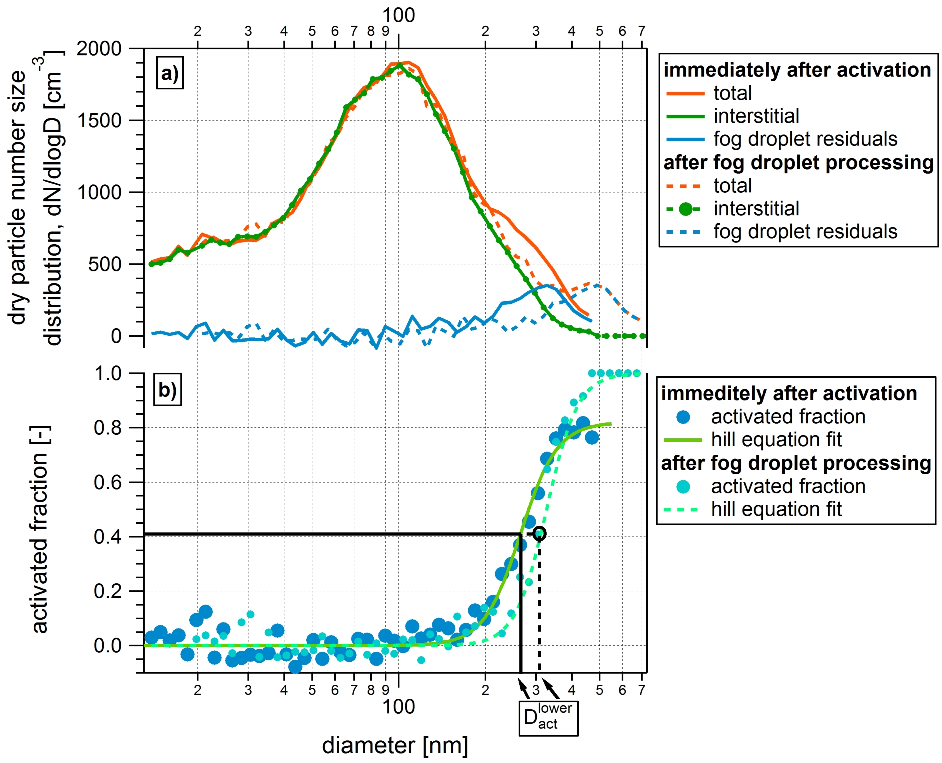

Figure 5: Same as Fig. 1 but additionally showing the size distributions resulting after fog droplet processing, i.e. condensation of semi-volatile aerosol components into the droplet and/or heterogeneous production of secondary aerosol components within the droplet.

S 1.5 Coalescence of fog droplets

Coalescence is the process of merging two or more droplets during contact to form a single droplet. Coalescence removes a constant fraction of the fog droplet residuals across all sizes, assuming that the fog droplet size is not related to the size of the CCN that formed the droplet. The residual particle from a droplet after coalescence will be larger than the bigger of the two original residual particles, at most by a factor of 1.26. We simulate the effect of coalescence by simply scaling the droplet residual size distribution by a factor of 0.8 (20 % coalescence), while the size change is ignored, as it only has a small additional effect. The interstitial particle size distribution remains unchanged and the new total size distribution is obtained as the sum of the new interstitial and droplet residual size distributions. Figure 6 shows that 20 % coalescence of fog droplets increases the $D_{\mathrm{act}}^{\mathrm{lower}}$ by ∼ 3 % in our example.

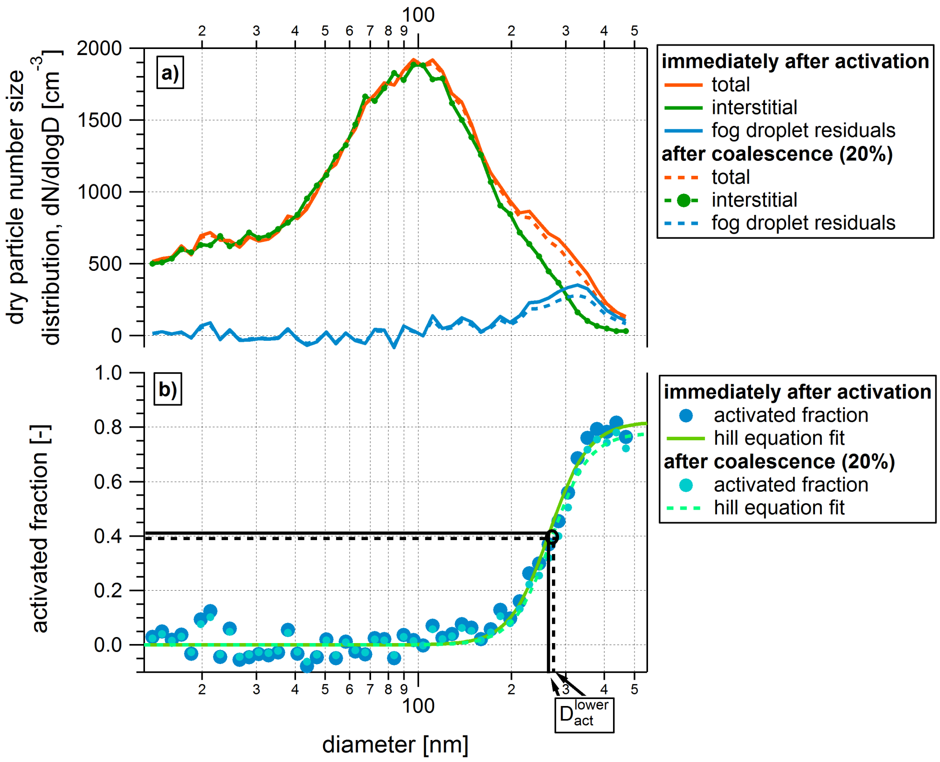

Figure 6: Same as Fig. 1 but additionally showing the size distributions resulting after coalescence of fog droplets