

\*\*\*Influence of the dry aerosol particle size distribution and morphology on the cloud condensation nuclei activation\*\*\*

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\*\*\*General introduction\*\*\*

Combustion and other high temperature processes frequently result in the emission of aerosols in the form of polydisperse fractal like aggregates made of condensed phase nanoparticles (soot for instance). If certain conditions are met, the emitted aerosol particles are known to evolve into important cloud condensation nuclei (CCN) in the atmosphere. In this work, the hygroscopic parameter  $\kappa$  of complex morphology aggregates is calculated from the supersaturation dependent activated fraction  $F_a = F_a(SS)$  in the frame of  $\kappa$ -Köhler theory. The particle size distribution is approximated with the morphology corrected volume equivalent diameter  $d_{ve}$  calculated from the electrical mobility diameter  $d_m$  by taking into account the diameter of the primary particle  $d_{pp}$  and the fractal dimension of the aggregate  $D_f$  experimentally obtained from transmission electron microscopy measurements. Activation experiments are performed in water supersaturation conditions using a commercial CCN-100 condensation nuclei counter. The model is tested in close to ideal conditions of size selected, isolated spherical particles (ammonium sulfate nanoparticles dispersed in nitrogen), then with complex polydisperse fractal like aggregates (soot particles activated by exposure to ozone with  $\kappa$  as low as  $5 \times 10^{-5}$ ) that represent realistic anthropogenic emissions in the atmosphere.

This database contains the experimental and calculated data used in the development of the model to predict the efficiency as cloud condensation nuclei (CCN) of fresh and chemically aged soot particles that includes their size distribution and morphology. Experimental data were collected during the PhD of Dr. J. Wu (2016-2019) in the laboratories of Lille University (PC2A group) and during measurement campaign at Créteil University (LISA group).

\*\*\*Description of the data in this data set\*\*\*

Each tab corresponds to a different case and to one specific figure discussed in the paper. In this file, both the raw experimental data and the calculated functions are listed and described case by case.

Fig.1. Simulations of  $d_{ve} = d_{ve}(d_{pp}, D_f, d_m)$  of soot particles having complex morphology. The DMA transfer function (black solid line) results from the size selection at 150 nm of soot particles sampled from the kerosene jet diffusion flame at 130 mm HAB. For each series of simulations, (a)  $d_m$  and  $D_f$ , or

alternatively (b)  $d_m$  and  $d_{pp}$  are set as constant and the remaining parameter is varied in the range indicated in the table.

Fig.3. (b) diameter of the particle projection  $d_p$  obtained from TEM measurements and fitting function. (c) electrical mobility diameter  $d_m$  obtained from SMPS measurements. The contributions of +1 and +2 charged particles to the fitting function are included.

Fig.4. Dry ammonium sulfate particles: (a) activation data obtained from CCNc experiments (black dots) and calculated  $F_a = F_a(SS)$  using SMPS and TEM data as  $d_{ve}$ . (b) corresponding  $p(\kappa)$  distributions.

Fig. 5. Soot sampled from the turbulent jet flame supplied with liquid kerosene at 70 mm HAB. (b)  $d_{pp}$  size distribution and lognormal fit,  $d_{pp} = 13.0$  nm (mass equivalent  $d_{pp} = 14.1$  nm). (c)  $\ln(N_{pp})$  vs.  $\ln(L_{2D}/d_{pp})$  plot from which  $D_f$  is obtained (42 projections). (d) normalized SMPS data after size selection at 150 nm and morphology corrected  $d_{ve}$ .

Fig. 6. Soot sampled from the turbulent jet flame supplied with liquid kerosene at 130 mm HAB. (b)  $d_{pp}$  size distribution and lognormal fit,  $d_{pp} = 16.7$  nm (mass equivalent  $d_{pp} = 17.7$  nm). (c)  $\ln(N_{pp})$  vs.  $\ln(L_{2D}/d_{pp})$  plot from which  $D_f$  is obtained (100 projections). (d) normalized SMPS data after size selection at 150 nm and morphology corrected  $d_{ve}$ .

Fig.7. Soot sampled from the turbulent jet flame supplied with liquid kerosene at 70 mm HAB. CCNc activation curves of the soot particles obtained after chemical aging with ozone, comparison of the experimental data (data points) to activation curves calculated under four different hypotheses: single  $\kappa$  value,  $p(\kappa)$ , DMA transfer function, and morphology corrected  $d_{ve}$ .

Fig. 8. Soot sampled from the turbulent jet flame supplied with liquid kerosene at 130 mm HAB. CCNc activation curves of the soot particles obtained after chemical aging with ozone, comparison of the experimental data to activation curves calculated by using  $p(\kappa)$  the and morphology corrected  $d_{ve}$ .

Fig. 9. Evolution of  $p(\kappa)$  vs. ozone exposure of (a) young soot particles sampled at 70 mm HAB, and (b) mature soot particles sampled at 130 mm HAB.