The impact of cloud processing on the ice nucleation abilities of soot particles at cirrus temperatures

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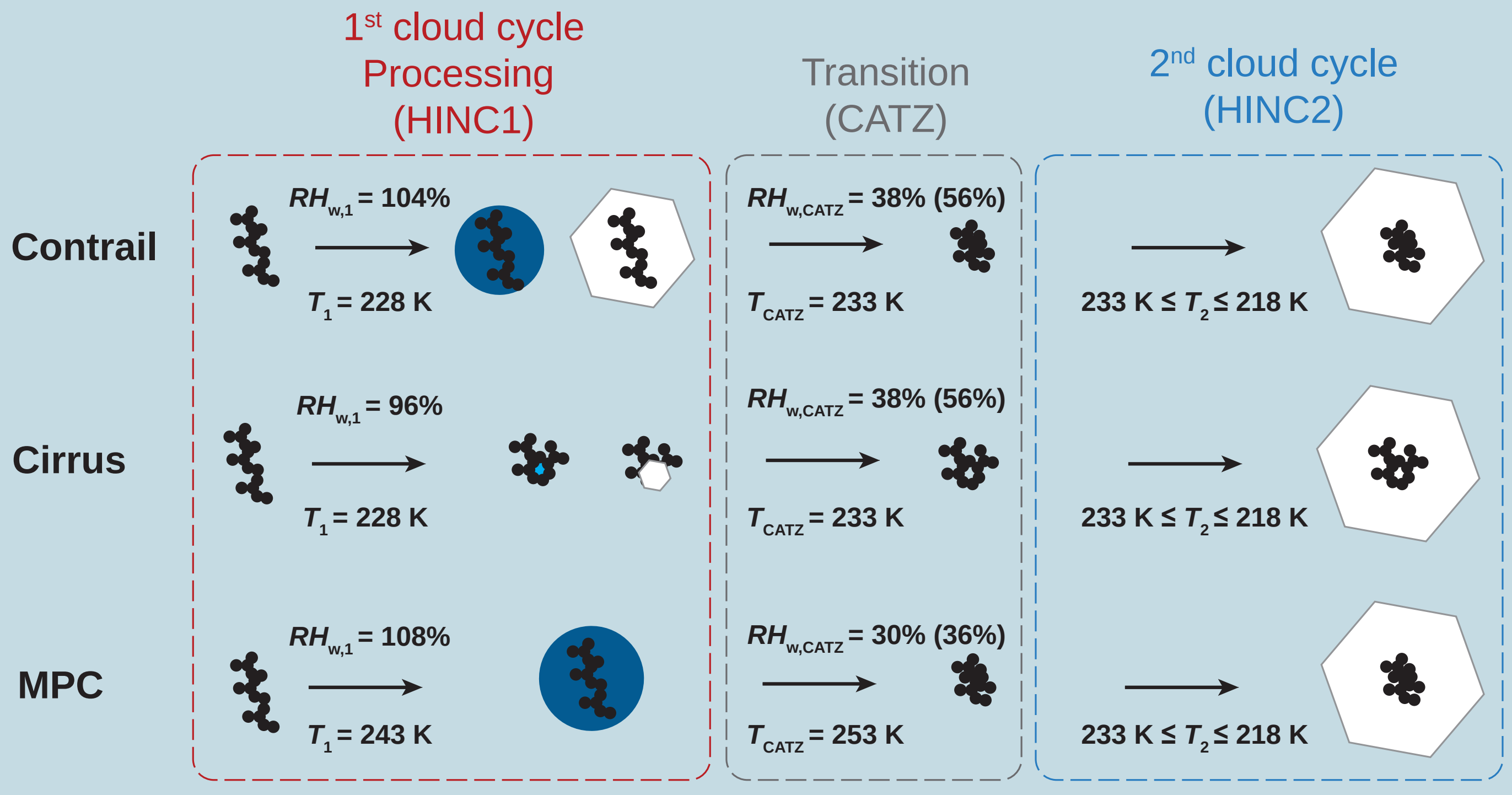
How can soot particles form ice clouds?

Soot aerosols are primary particles produced by incomplete combustion of both fossil fuels and biomass burning. Freshly emitted soot particles are generally found to be hydrophobic and poor ice nucleating particles (INPs)[1,2]. However, during their **atmospheric lifetime** of approximately a week[3] soot particles can be involved into different **aging process**. One aging process is the **involvement into cloud mircosphysical processes**[4]. Here we present results from a laboratory study that investigates the impact of such cloud processing on the ice nucleation ability of soot.

We show that cloud processing significantly enhances the ice nucleation activity of soot and propose hydrometeor formation on the soot to be the key factor. In the atmosphere such scenarios could be important for e.g. contrail formation on **aviation emitted particles**.



Soot aggregate · Ice crystal · Supercooled cloud droplet

Fig. 1: Overview of the different cloud processing types investigated, namely contrail, cirrus and mixed-phase cloud (MPC) processing. The processing pathways differ in terms of thermodynamic conditions experienced by the soot particles (temperature, T; and relative humidity w.r.t. water; RH_w).

Dashed boxes mark distinct steps in our experimental setup (Fig. 2): Cloud processing step (red box), transition step (grey box) and ice nucleation step (blue box).

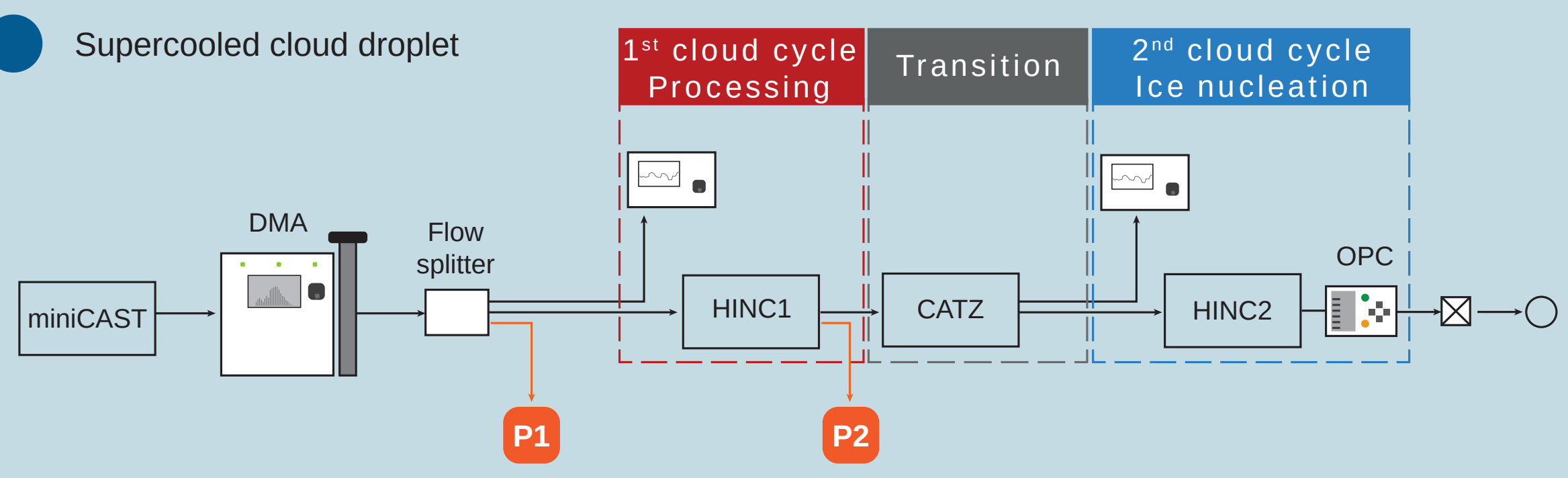


Fig. 2: Schematic of the experimental setup used to investigate cloud processing. Soot was generated from a propane burner (miniCAST). DMA size selected soot was then cloud processed in a first cloud chamber (HINC1)[5]. The processed soot was transported through a temperature controlled flow tube (CATZ) into a second cloud chamber (HINC2), where the ice nucleation potential was tested at cirrus cloud temperatures.

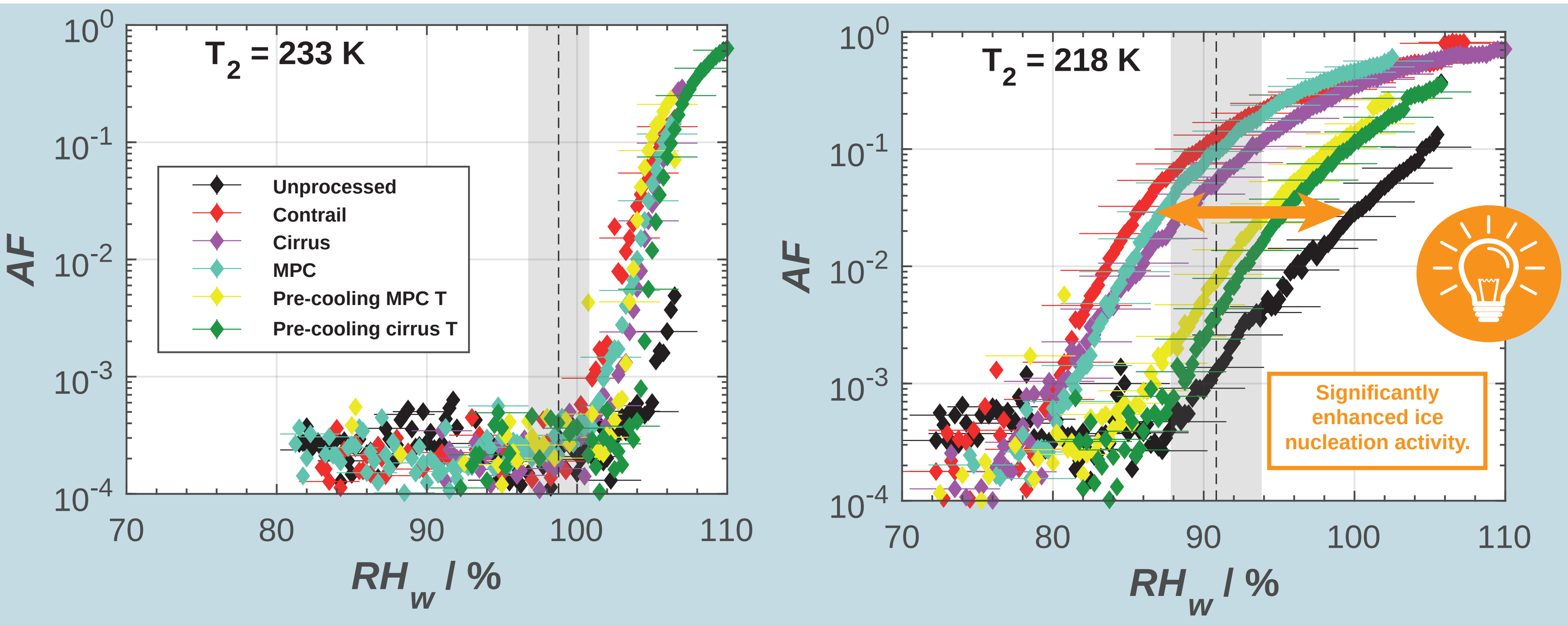


Fig. 3: Mean *RH* scans of 400 nm mobility diameter cloud processed soot particles, showing *AF* as a function of RH_w for different temperatures in the cirrus regime. Black dashed lines represent expected homogeneous freezing conditions according to Koop et al. (2000)[10], and the gray shaded regions indicates the calculated RH_w variation across the aerosol lamina in HINC. Uncertainties are given for every 5th data point. Different colors represent different processing scenarios (see Fig. 1). Also shown are the ice nucleation results of unprocessed soot (black symbols). During pre-cooling experiments RH/T conditions where such that no hydrometeors formed on the soot particles during the processing step (HINC1, Figs. 1 and 2).

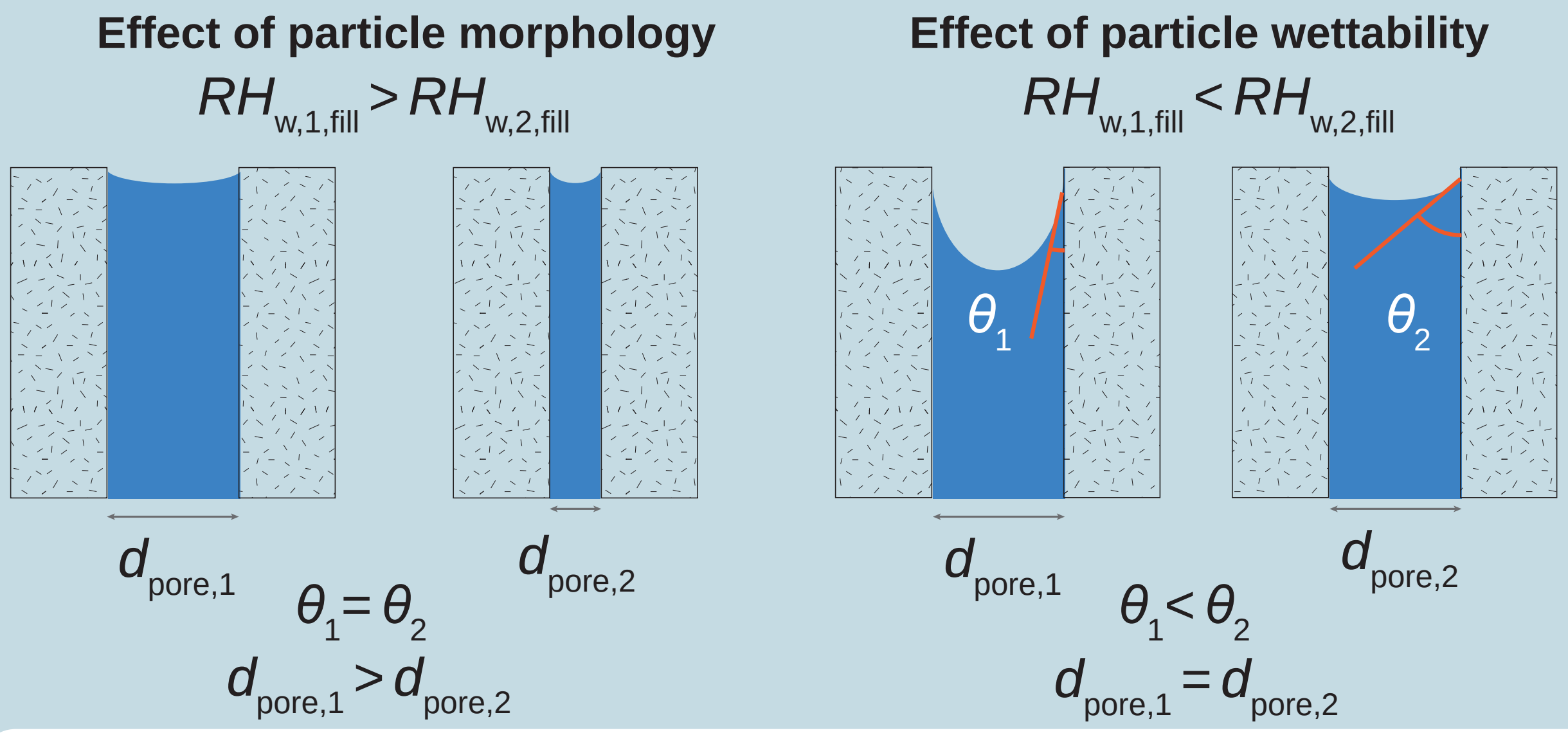


Fig. 4: Illustrattion of the two main effects controlling capillary condensation of water: i) pore diameter (left) and ii) pore wettability/contact angle (right)[11]. Smaller pores can take up water at relatively lower RH_w. Similarly, the lower the contact angle, the more hydrophilic a pore is and the lower the filling RH_w. If soot particles are cooled to T < 235 K the pore water can freeze homogeneously, allowing soot to form ice ice via pore condensation and freezing (PCF).

▨ Pore material
🟦 Pore water

Cloud processed soot is significantly more ice active.

Intermediate effect when no hydrometeor formed on soot particles during processing step (pre-cooling experiments in Fig. 3).

Enhanced ice nucleation not due to pre-activation (not shown) but from enhanced ice formation by pore condensation and freezing[6].

What particle property controls enhanced ice nucleation?

We conducted measurements of particle hydrophilicity and morphology for unprocessed and cloud processed particles in order to determine what particle property controls the enhanced PCF ice nucleation activity. Particle hydrophilicity was investigated using dynamic water vapor sorption measurements and particle morphology was examined by means of transmission electron microscopy.

Effect of particle wettability

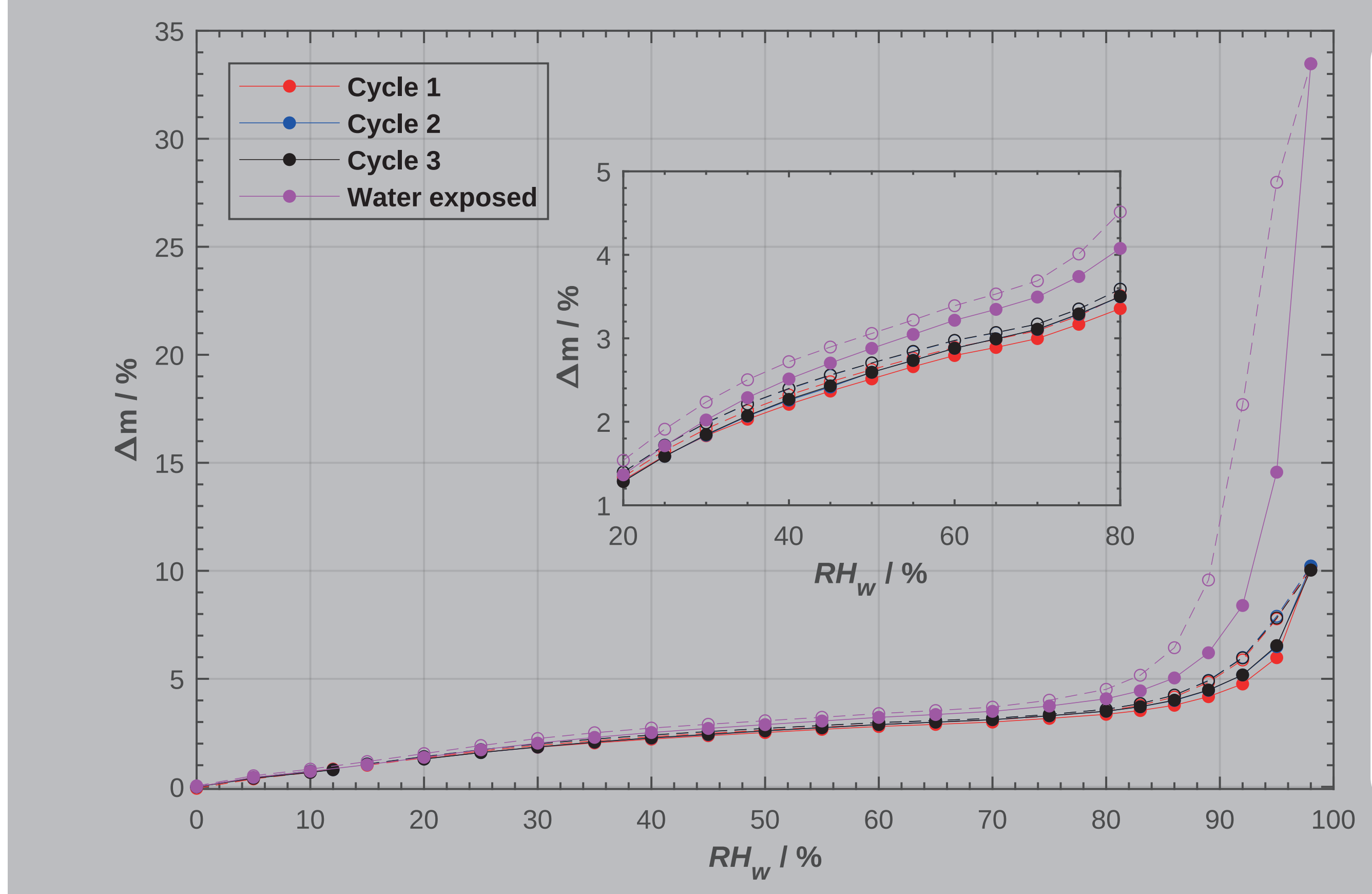


Fig. 5: Dynamic water vapor sorption measurements showing the water uptake (solid lines, filled symbols) and water loss (dashed lines, open symbols) in terms of mass change relative to sample dry weight, as a function of RH_w.

Cloud processed/water exposed soot shows enhanced water uptake capacity.

The increased water uptake at high RH_w reveals the presence of mesopores[7].

Enhanced water uptake can result from change of pore size distribution and or contact angle.

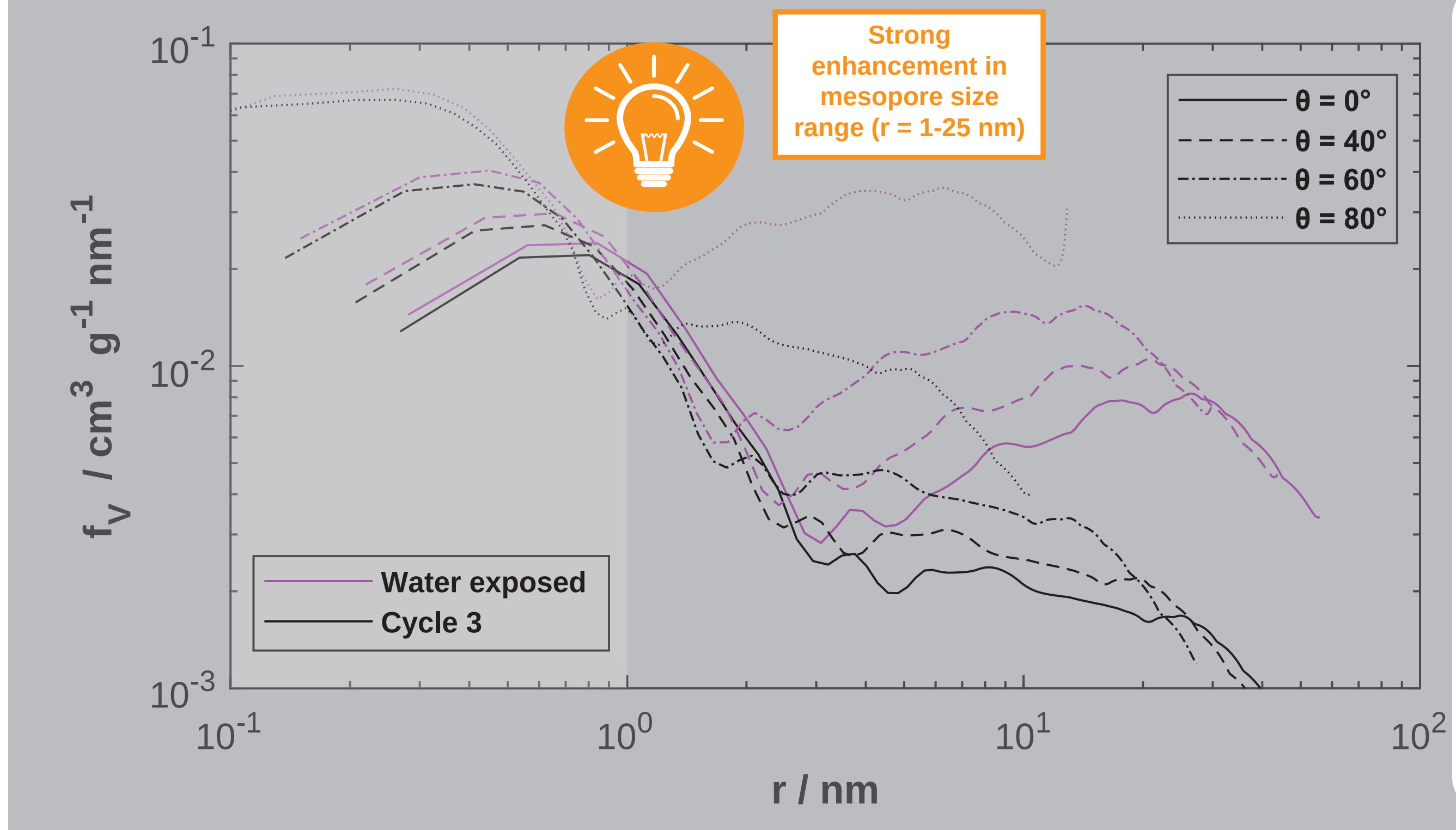


Fig. 6: Pore size distribution (PSD), calculated from water uptake curves (see Fig. 5)[8], using the inverse Kelvin equation and assuming different soot-water contact angles. Contact angles of 0° and 180° denote a perfectly wettable and inwettable surface, respectively. Typical soot-water contact angles are between 40-80°. The shaded area indicates pores outside the mesopore range, where no capillary condensation takes palce and our method to estimate the PSD becomes invalid.

Cloud processed soot shows increased mesopore water volume, independet of contact angle.

FTIR measurements reveal no differene in hydrophilic function groups between unprocessed and processed soot (not shown).

Assumption of different contact angle not warranted.

Effect of particle morphology

Cloud processed soots are more compact.

Morphology change favours ice nucleation through pore condensation and freezing.

Hydrometeor formation on soot particles during processing is key for compaction and enhanced ice nucleation activity.

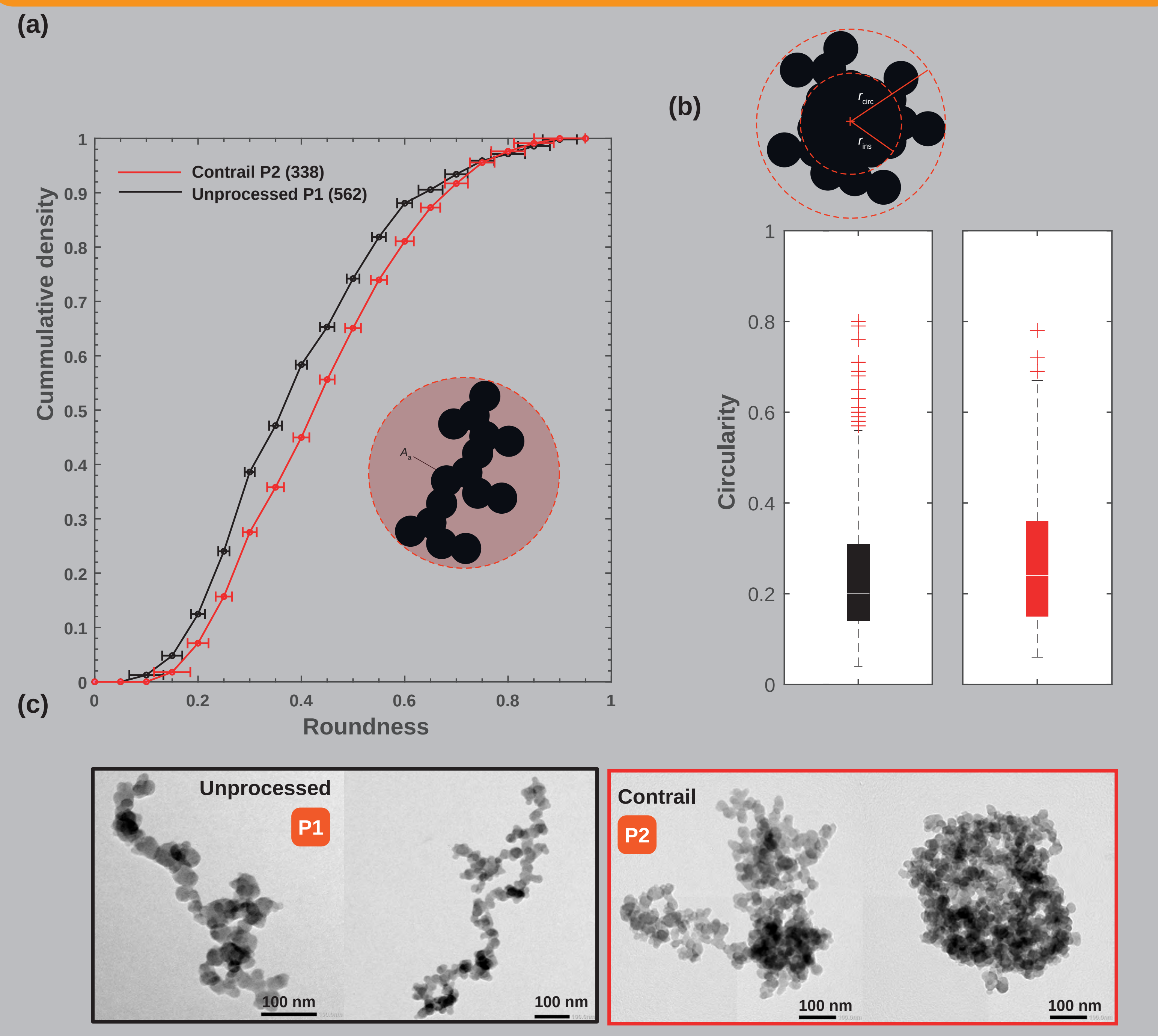


Fig. 7: Morphological anaylsis[9] of unprocessed and (contrail) processed soot particles, sampled at position P1 and P2, respectively (see Fig. 2). Panels show (a) particle roundness and (b) circularity. (c) shows representative TEM images of unprocessed and processed soot aggregartes, sampled at location P1 and P2, respectively (see Fig. 2). Sampling at P2 was achieved using a pumped counterflow viertual impactor. Particle roundness is defined as the ratio of the area of the soot aggregate to the area of a cirecle circimscribing the aggregate, as indicated by the inset. Particle circularity is defined by the ratio of two circles inscribing and circumscribing the aggeragate, as indiacated by the schematic.

Summary and Conclusions

Cloud processing can effectively increase the ability of soot to nucleate ice in cirrus clouds.

Increased ice nucleation results from pure physical changes of particle morphology (compaction).

Cloud processed soot could be an important source of INPs at upper troposheric conditions, e.g. from aircraft emissions, where water vapor condensed on soot aggregates and freezes homogeneously (contrail processing scenario).

Acknowldgement:
The authors gratefully acknowledge the great technical support of H. Wydler in overseeing the maintenenace of HINC and improving both the CFDC and associated software.

Read more on soot ice nucleation

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