

Figure S1: Wind rose centred on measurement location. The prevailing wind passing up the Rhine valley into the COPS region at around 5m/s average wind speed (measured at 1166m ABSL, 2m AGL).



Figure S2: Relative Humidity (RH), organic:sulphate ratio, organic:inorganic ratio and dNdlogDp for the measurement period, segregated into "cloud periods" (CP) and "no cloud" periods (NC). Cloud periods are defined by $RH \ge 85\%$ and characterised by a low organic:inorganic ratio and low number concentrations. No cloud periods are characterised by higher a organic:sulphate ratio and tend to start with high concentrations of small (≤ 40 nm particles, which grows into a large (≈ 150 nm), strongly monomodal size distribution.



Figure S3: Size resolved AMS data for both a) $\leq 4:1$ and b) $\geq 4:1$ organic:sulphate ratio periods. The high organic to sulphate ratio periods (b) show a dramatic relative increase in organics above 100nm, with the majority of the organic fraction around 300nm.



Figure S4: A graph showing $S_{c,D_0} - S_{set}$ vs D_0 . The two data points straddling the zero line are linearly interpolated between, with the intercept defining the physical threshold diameter of the aerosol, $D_{thres(S_{c,D_0})}$. The errors on $D_{thres(S_{c,D_0})}$ are propagated using standard procedure.