Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

High Time Resolution Measurements of Droplet Evaporation Kinetics and Particle Crystallisation – Supplementary Information

D. A. Hardy[a], J. Archer[a], P. Lemaitre[b], R. Vehring[c], J. P. Reid[a] and J. S. Walker[a]*

A refined technique for observing the complete evaporation behaviour of free-falling droplets, from droplet generation to complete solvent evaporation, with ultra-high time resolution is introduced and benchmarked. High-resolution phase-delay stroboscopic imaging is employed to simultaneously resolve the evolving droplet morphology, geometric and aerodynamic diameters, throughout the evaporative lifetime with a user-controlled < µs timescale. This allows rapid, complex morphological changes, such as crystallisation events, to be clearly observed and the corresponding mechanisms to be inferred. The dried particles are sampled for offline SEM analysis and the observed morphologies compared to the inflight imaging. Density changes can be calculated directly from the deviation between the geometric and aerodynamic diameters.

The full capabilities of the new technique are demonstrated by examination of the different evaporation behaviours and crystallisation mechanisms for aqueous sodium chloride droplets evaporating under different ambient relative humidity (RH) conditions. The crystallisation window, defined as the time taken from initial to complete crystallisation, is shown to be RH dependent, extending from 0.03 s at 20 % RH and 0.13 s at 40% RH. The different crystallisation mechanisms observed during the experiments are also clearly reflected in final structure of the dry particles, with multi-crystal structures produced at low RH compared to single-crystal structures at higher RH.

It is anticipated that this technique will unlock measurements which explore the evaporation behaviour and crystallisation mechanisms for rapid, complex droplet drying events, and with increasingly non-ideal solutions, relevant to industrial applications.

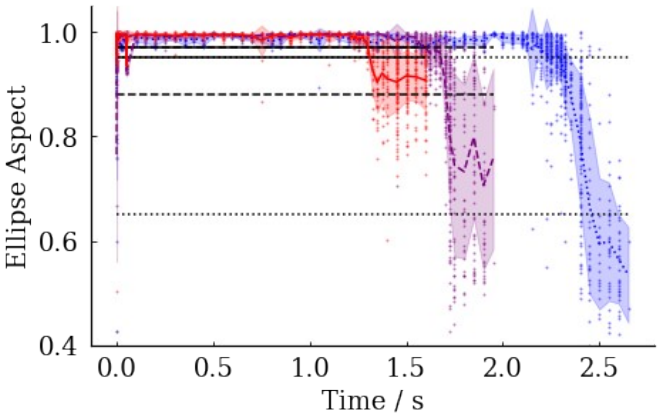

Figure 1. The aspect ratio of particles over time for NaCl droplets evaporating at 293 K in 20% RH (red), 30% RH (purple) and 40% RH (blue). The respect thresholds in aspect ratio used to determine state (either liquid droplet or final crystal form) are marked in black, 20% RH (solid line), 30% RH (dashed line) and 40% RH (dotted line).