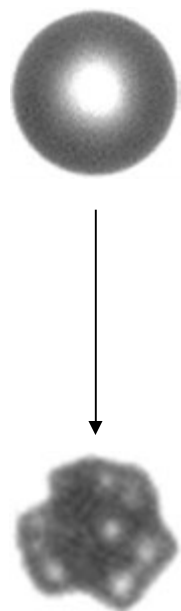
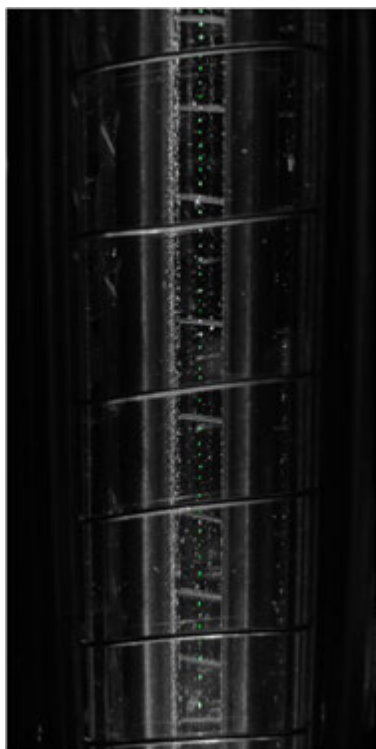
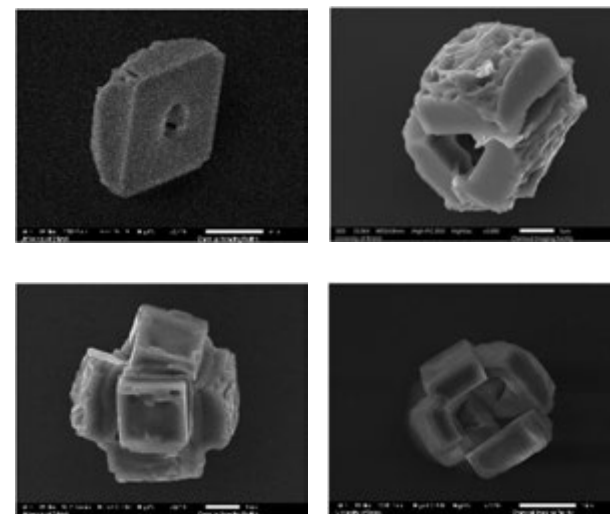


## High Time Resolution Measurements of Droplet Evaporation Kinetics and Particle Crystallization



Dan Hardy

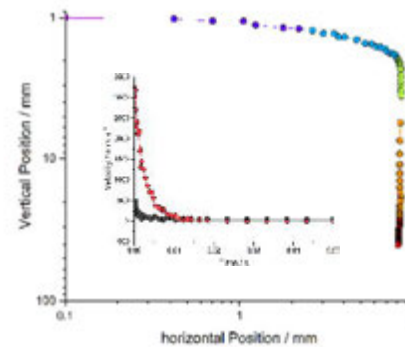


## A bit about me

- My history and journey to present day

## Some science

- Introduction to my project
- Some results



How did I end up where I am?



# Profile

13 March 2021





Electronics



Maths



Chemistry



Physics





# EPSRC

Engineering and Physical Sciences  
Research Council



WEB OF SCIENCE

Scopus®

All Types of Produce	Apple vs. Other	Fruits vs. Vegetables
Apple	=if(A2="Apple")	
Banana		
Carrot		
Watermelon		







Undergraduate at  
University of Bristol



Chemistry

Chemical Physics

Physics





## Finding a PhD:

1. Chance email
2. Basic criteria:
  - a. Interesting
  - b. In Bristol





### Finding a PhD:

1. Chance email
2. Basic criteria:
  - a. Interesting 
  - b. In Bristol 
3. Application process progressed
  - Reassess criteria



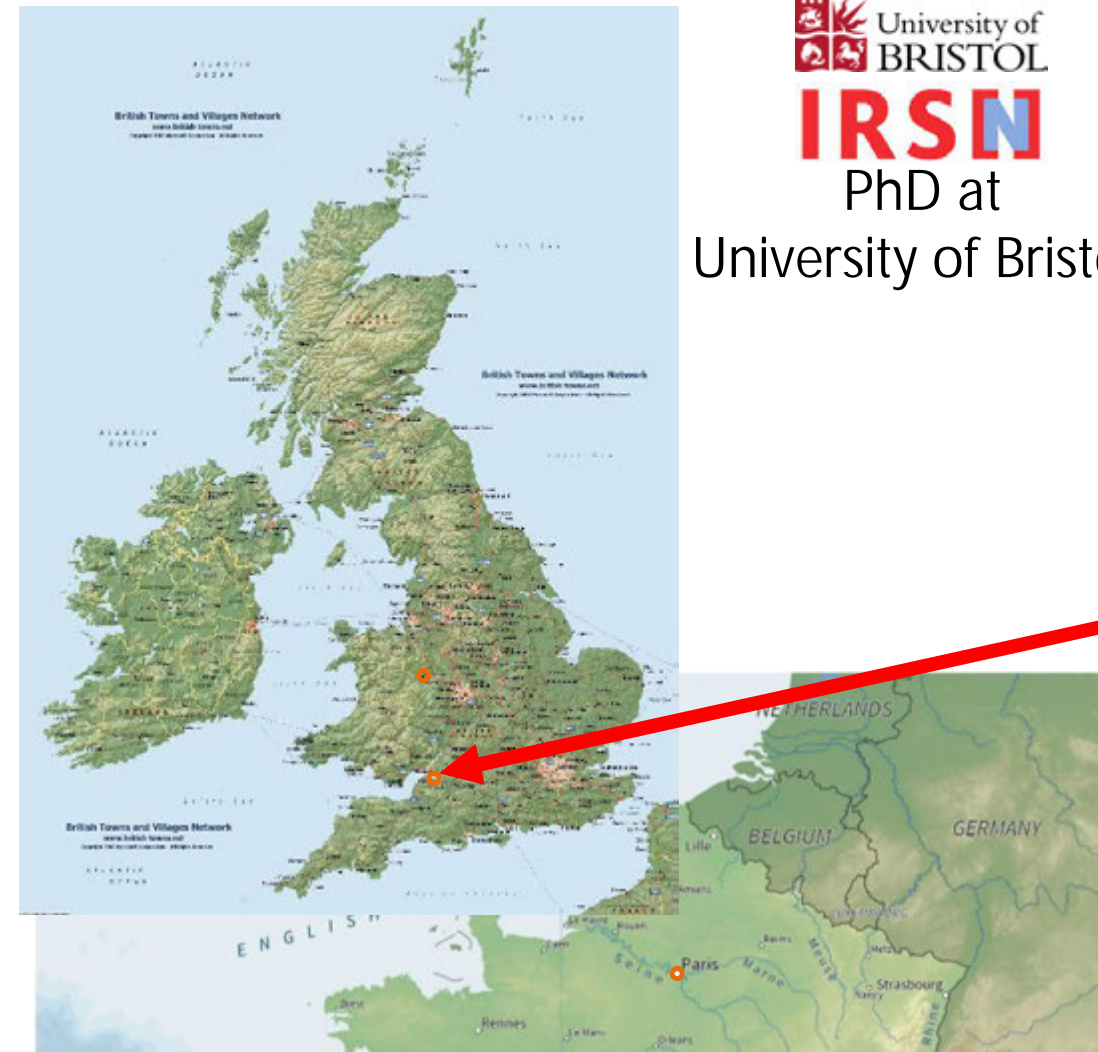
**IRSN**





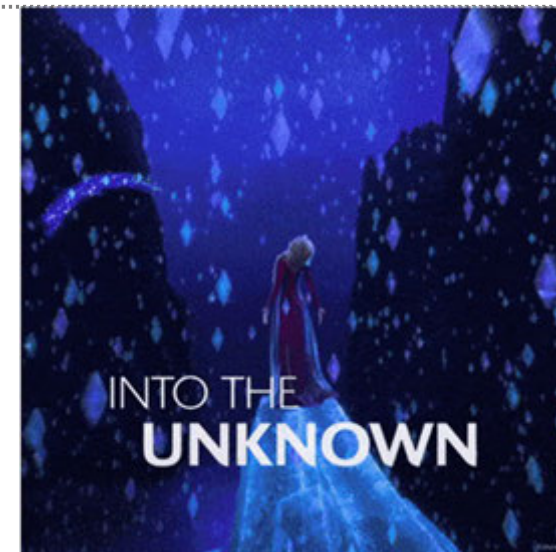
PhD at  
University of Bristol

Bristol Aerosol Research Centre  
+  
Institute for Radiological Protection and Nuclear Safety





# What next?



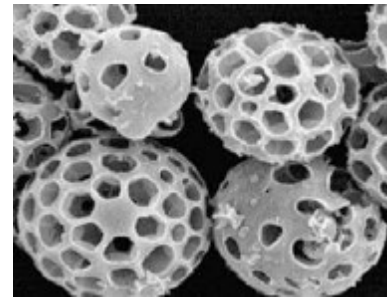
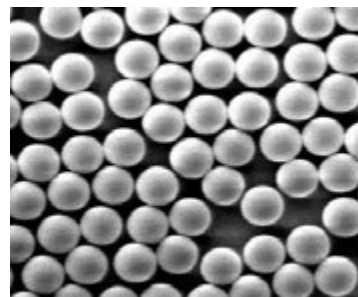
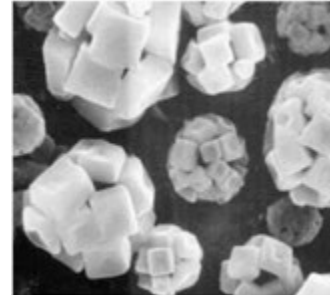
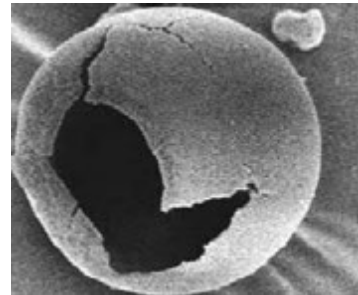
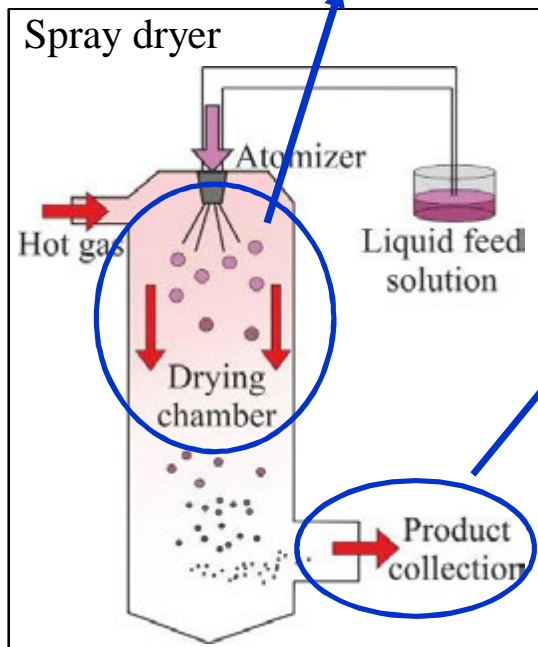
Things I have enjoyed:

- Instrument development
- Data analysis and modelling
- Aerosol Science
- Bristol





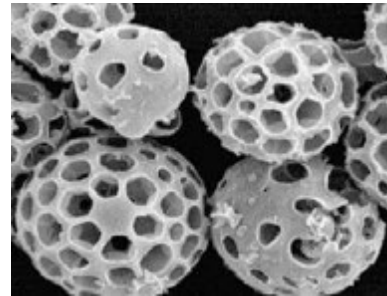
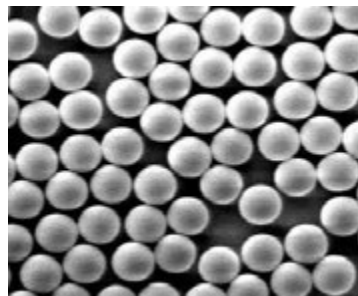
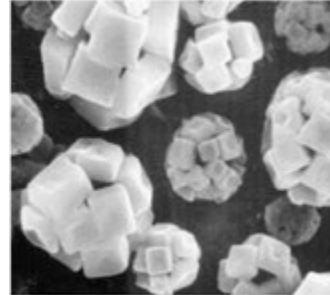
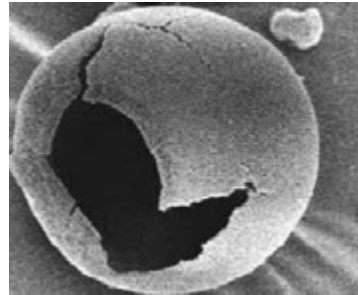
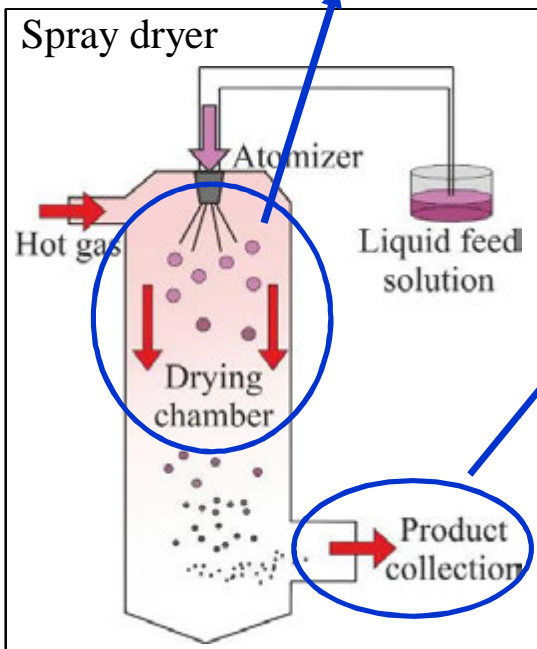
## Formation of micro-structured particles





# Spray Drying – Food and Drink

13 March 2021



### Pharmaceutical Industry

- Powder production
- Drug delivery to the lungs



## Spray Drying – Other Scenarios

13 March 2021

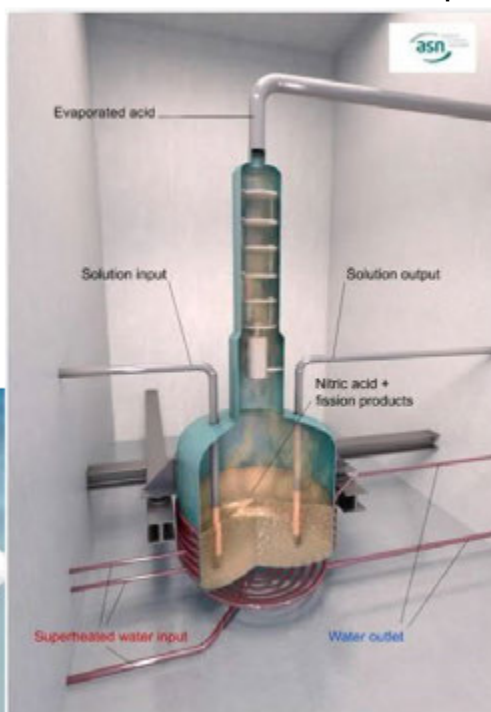
### Pharmaceutical Industry

- Powder production
- Drug delivery to the lungs



### Nuclear Industry

- Release of contamination via aerosol
- Waste processing
  - Fission product evaporators





## Spray Drying – Other Scenarios

13 March 2021

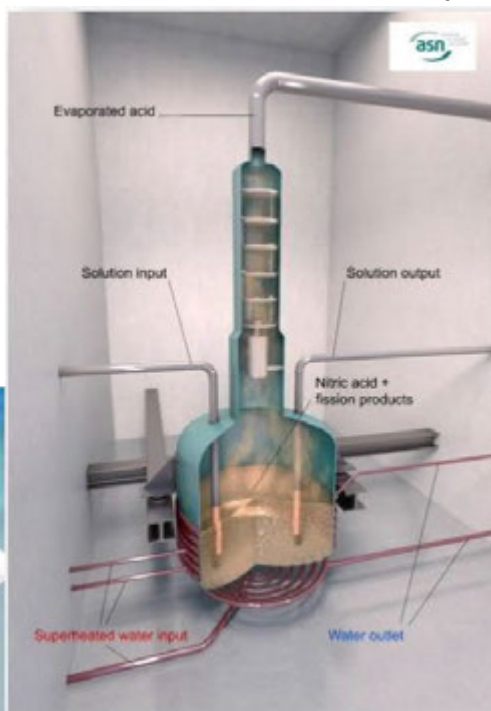
### Pharmaceutical Industry

- Powder production
- Drug delivery to the lungs



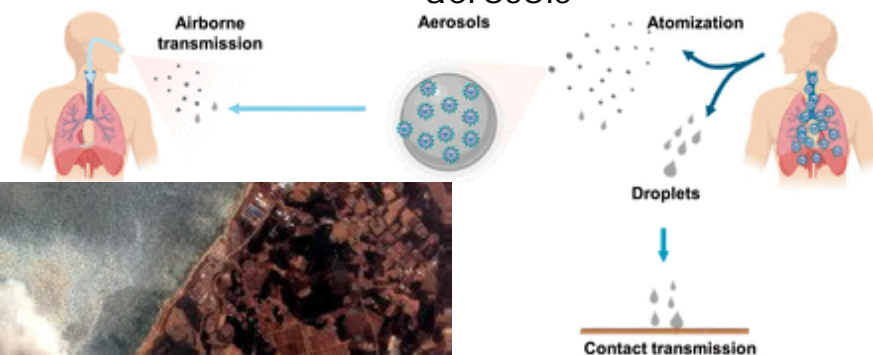
### Nuclear Industry

- Release of contamination via aerosol
- Waste processing
  - Fission product evaporators



### Airborne transmission of disease

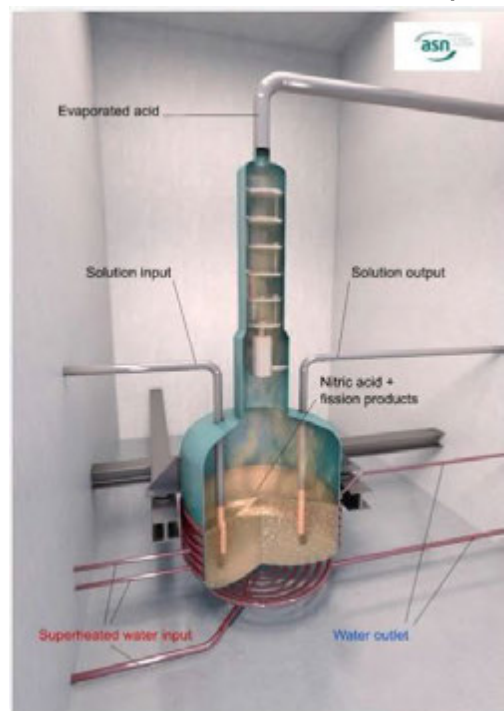
- Release of bioaerosol
- Viability of pathogens in aerosols



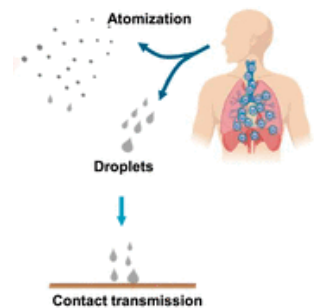
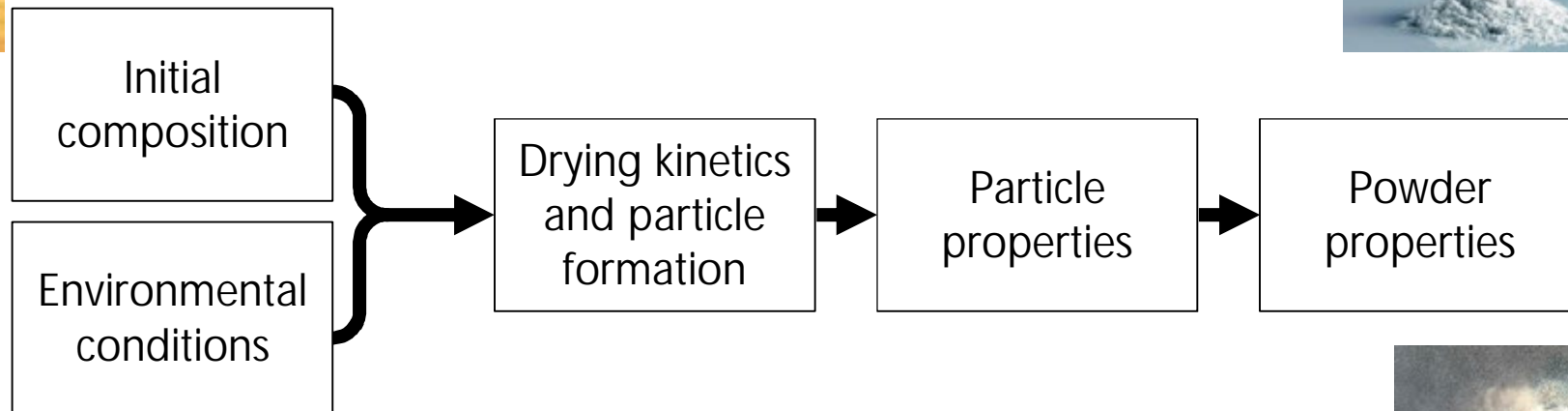


### Nuclear Industry

- Release of contamination via aerosol
- Waste processing
  - Fission product evaporators



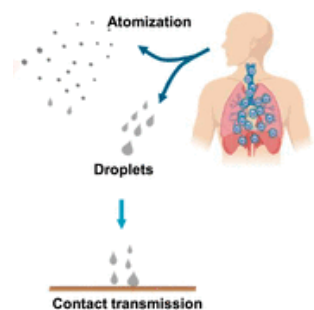
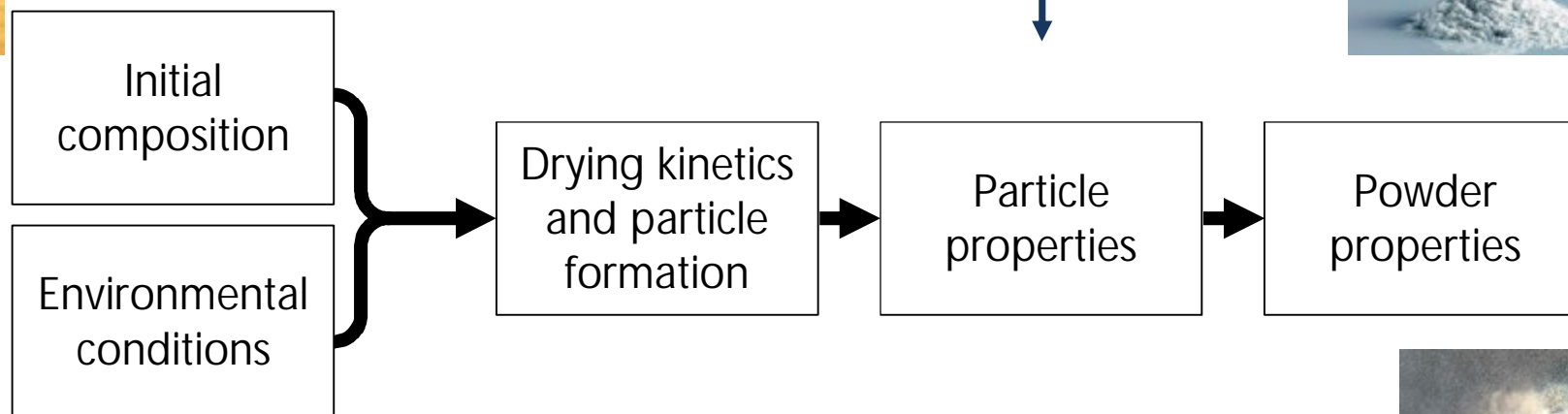
*Determination of the transport properties of aerosol particles produced by droplet drying*



# Spray Drying - processes

13 March 2021

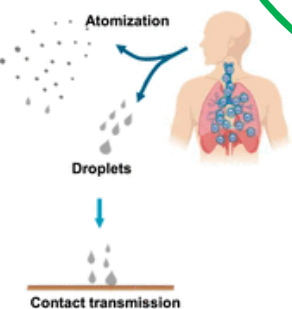
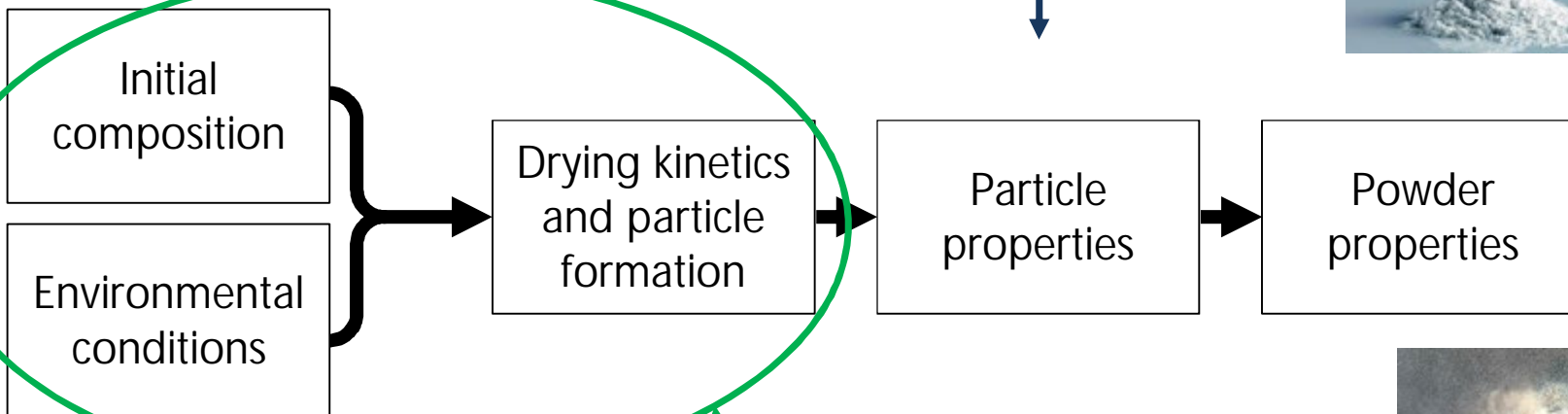
Aim: Understanding and control of particle properties



# Spray Drying - processes

13 March 2021

Aim: Understanding and control of particle properties



To achieve this, we must understand the preceding stages





## Spray Drying – physical driving forces

---

- Droplet evaporation  $d(t)^2 = d_0^2 - \kappa t$ 
  - Solvent properties
  - Drying conditions
  - Particle composition

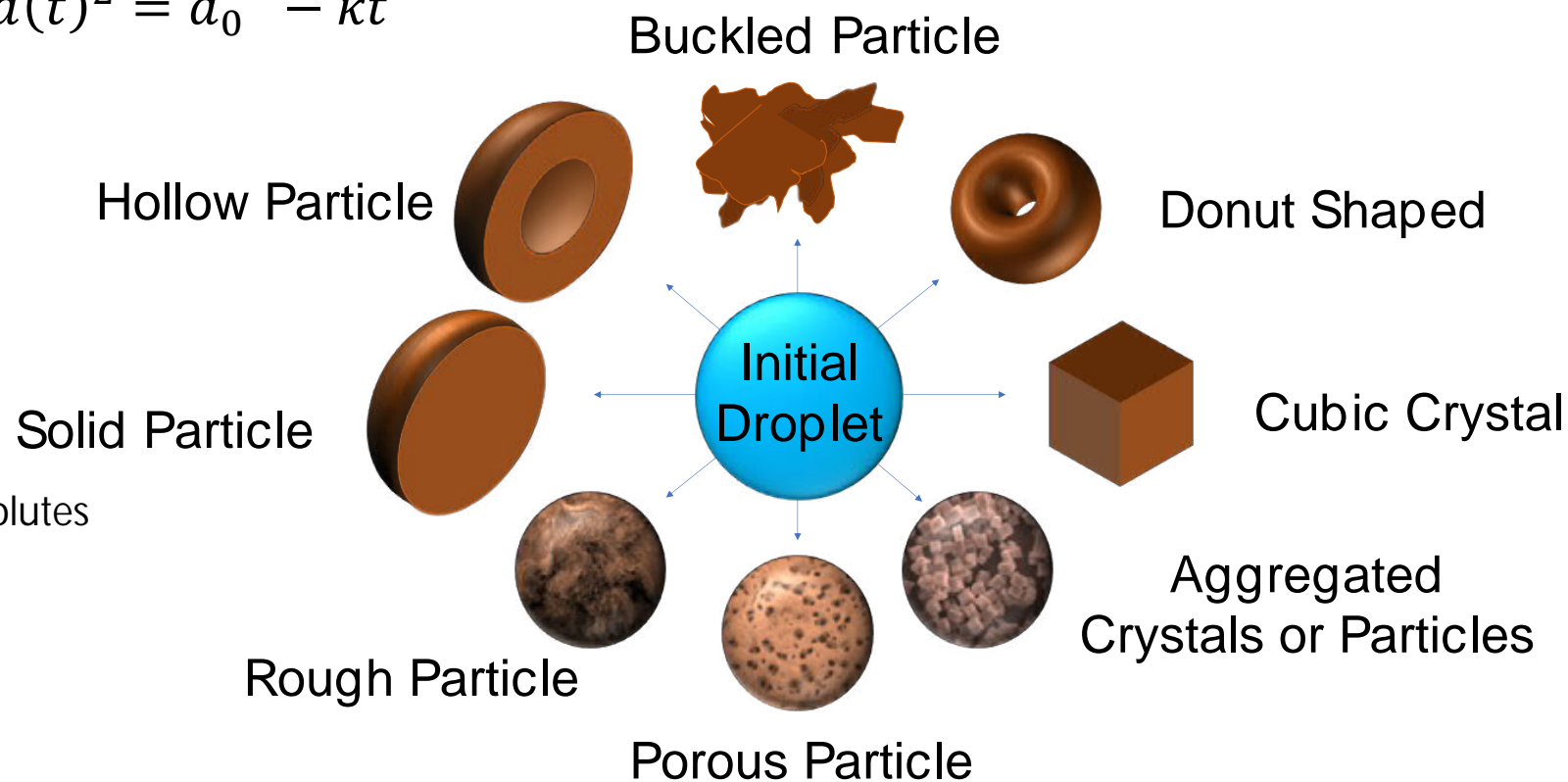
- Droplet evaporation

$$d(t)^2 = d_0^2 - \kappa t$$

- Solvent properties
- Drying conditions
- Particle composition

- Particle formation

- Solidification behavior of solutes or colloidal particles
- Solvent properties
- Solvent/solute interactions



### Study dry particle formation from aerosol droplets

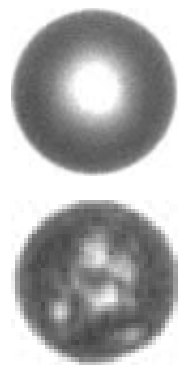
- Contact free behaviour
- Droplet dimensions
- Droplet density





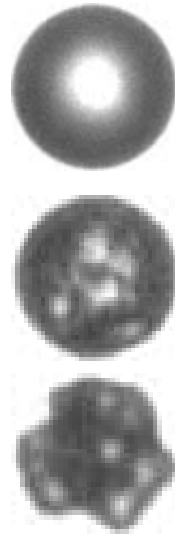
### Study dry particle formation from aerosol droplets

- Contact free behaviour
- Droplet dimensions
- Droplet density
- Droplet phase



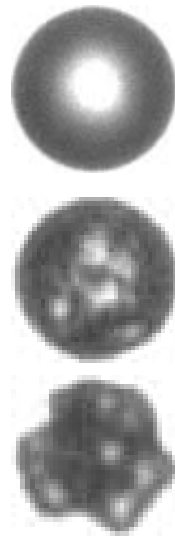
### Study dry particle formation from aerosol droplets

- Contact free behaviour
- Droplet dimensions
- Droplet density
- Droplet phase
- Droplet structure



### Study dry particle formation from aerosol droplets

- Contact free behaviour
- Droplet dimensions
- Droplet density
- Droplet phase
- Droplet structure



### Falling Droplet Chain Instrument

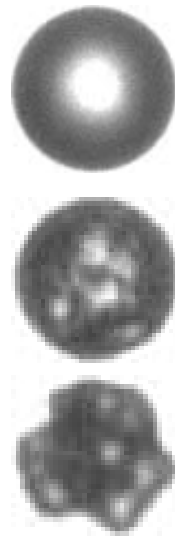
- Monodisperse droplet chain





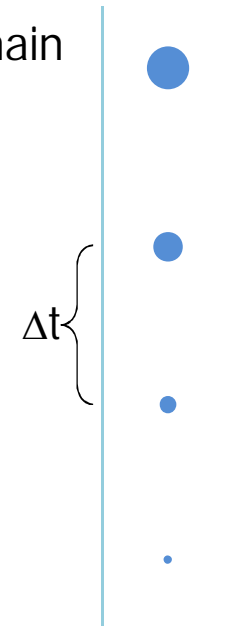
### Study dry particle formation from aerosol droplets

- Contact free behaviour
- Droplet dimensions
- Droplet density
- Droplet phase
- Droplet structure



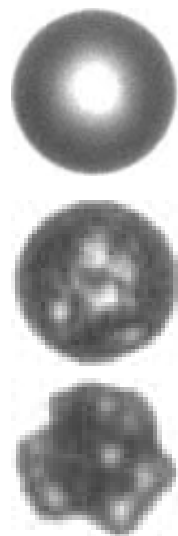
### Falling Droplet Chain Instrument

- Monodisperse droplet chain
- Stroboscopic imaging



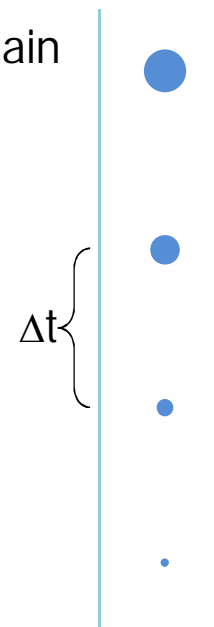
### Study dry particle formation from aerosol droplets

- Contact free behaviour
- Droplet dimensions
- Droplet density
- Droplet phase
- Droplet structure



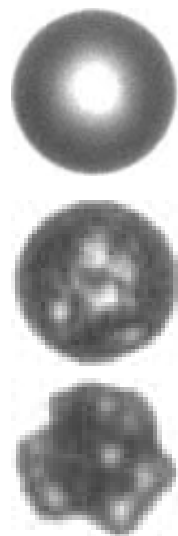
### Falling Droplet Chain Instrument

- Monodisperse droplet chain
- Stroboscopic imaging
- Observation throughout evaporative lifetime



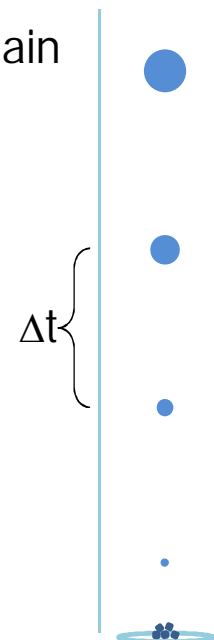
### Study dry particle formation from aerosol droplets

- Contact free behaviour
- Droplet dimensions
- Droplet density
- Droplet phase
- Droplet structure



### Falling Droplet Chain Instrument

- Monodisperse droplet chain
- Stroboscopic imaging
- Observation throughout evaporative lifetime
- Collect final particles for SEM imaging

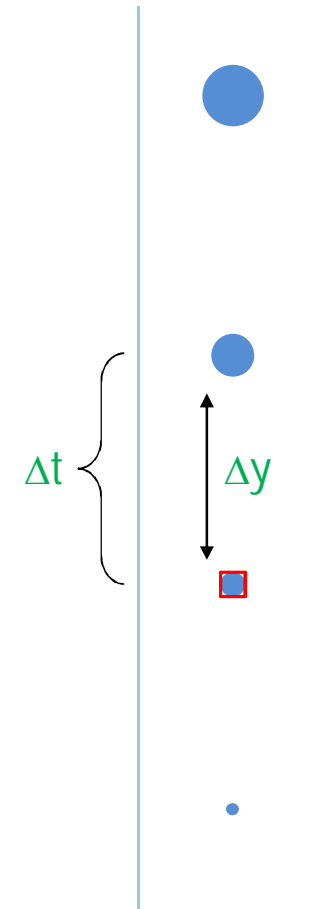




## Aerodynamic Diameter

Diameter of a sphere with a density of  $1 \text{ gcm}^{-3}$  that has a settling velocity,  $v_s$ , equal to a droplet in question.

$$\frac{\Delta y}{\Delta t} = v_s = \frac{\rho g d(t)^2}{18\mu} \quad d_a = \sqrt{\frac{18\mu v_s}{\rho^* g}}$$



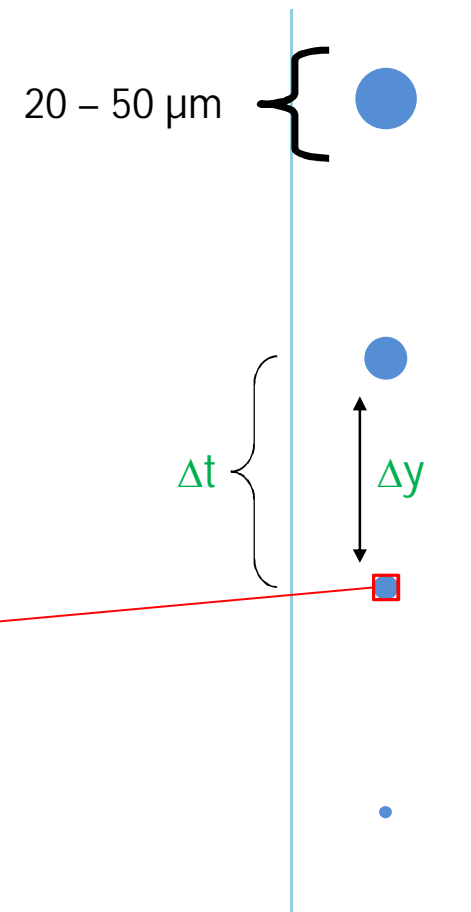
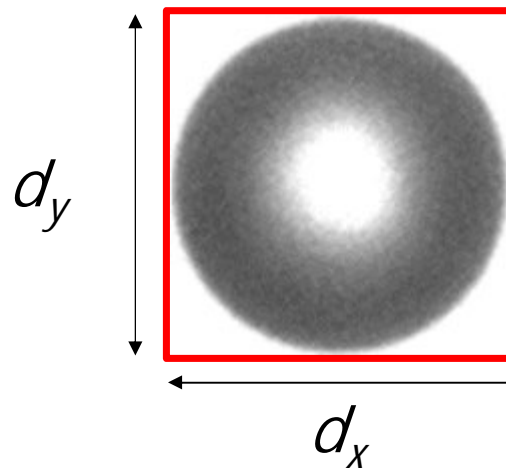
# Measuring Droplet Diameter

## Aerodynamic Diameter

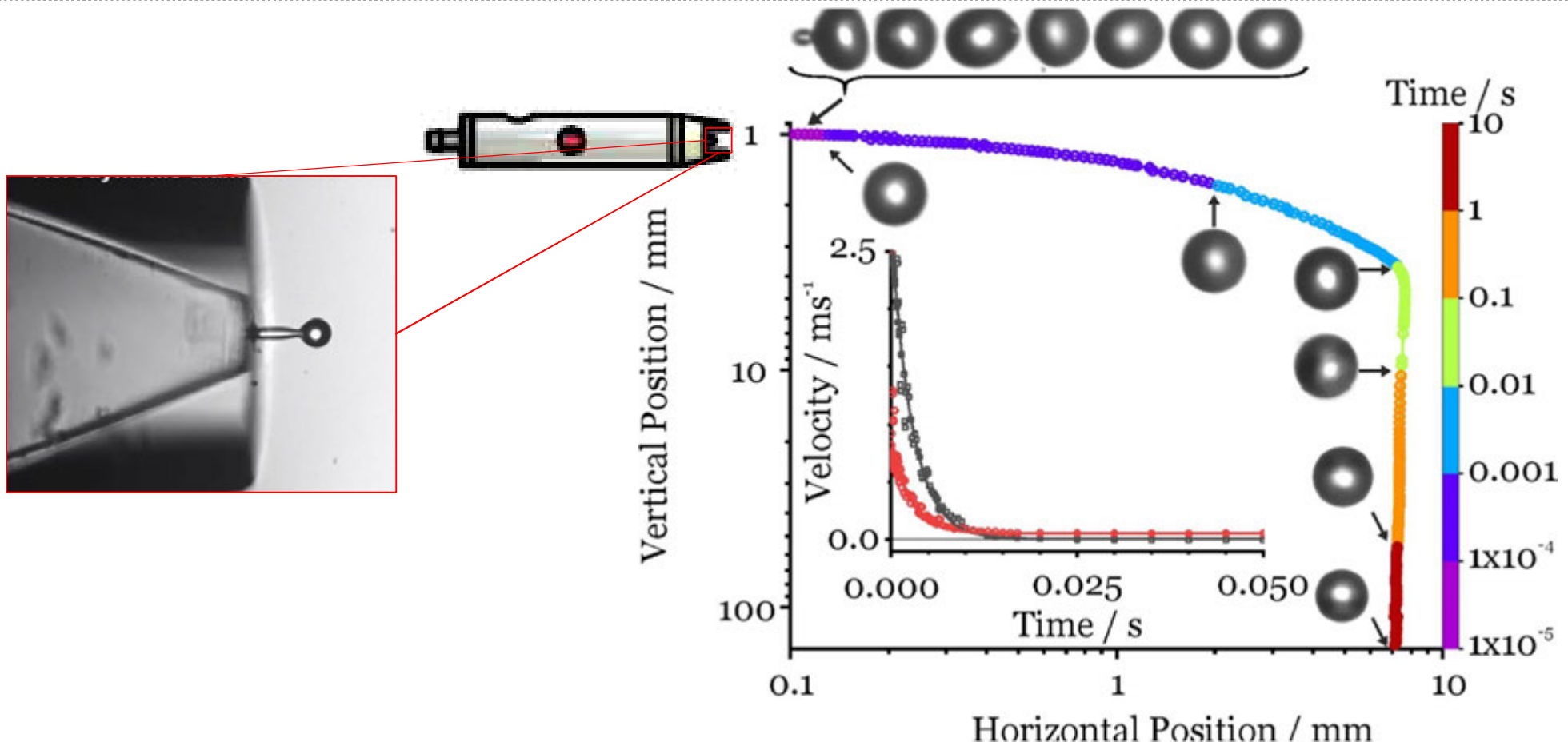
Diameter of a sphere with a density of  $1 \text{ gcm}^{-3}$  that has a settling velocity,  $v_s$ , equal to a droplet in question.

$$\frac{\Delta y}{\Delta t} = v_s = \frac{\rho g d(t)^2}{18\mu} \quad d_a = \sqrt{\frac{18\mu v_s}{\rho^* g}}$$

## Geometric diameter



# Observing Droplet Trajectories



Sample: Water, 75% RH, 294K,  $V_{\text{gas}} = 20.8\text{mms}^{-1}$   
 Dispenser Nozzle position (0.1, 1)

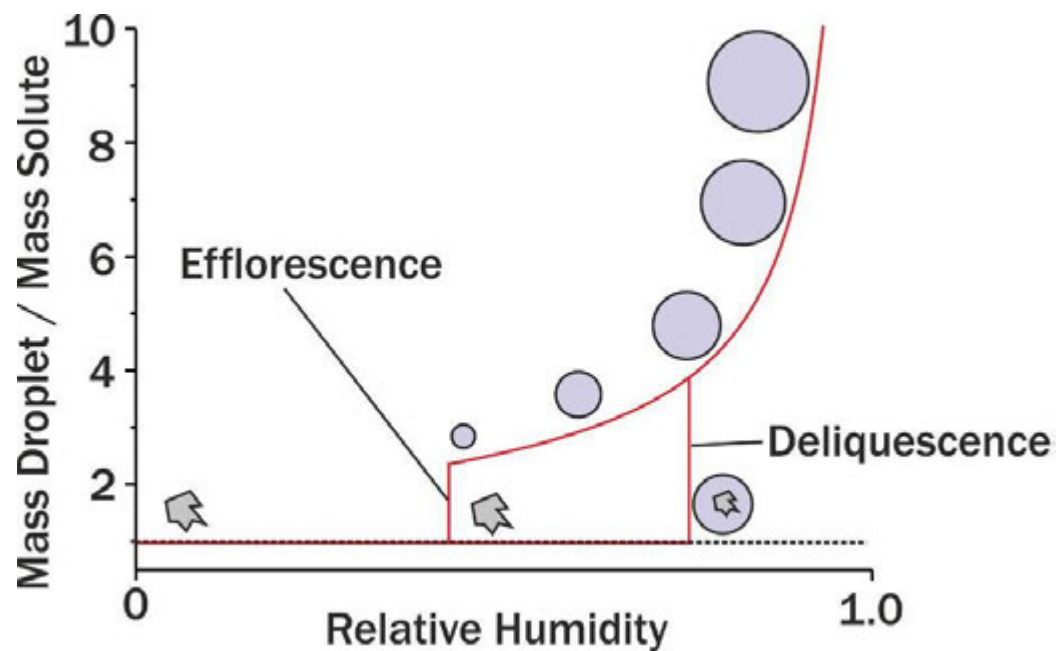


Sodium chloride:

- Solute, not colloidal
- Ideal solute

## Sodium chloride:

- Solute, not colloidal
- Ideal solute
- Well known behaviour in aerosol phase
- Clear solidification behaviour



### Sodium chloride:

- Solute, not colloidal
- Ideal solute
- Well known behaviour in aerosol phase
- Clear solidification behaviour

- 
- Understand the phenomena that occur

### Sodium chloride:

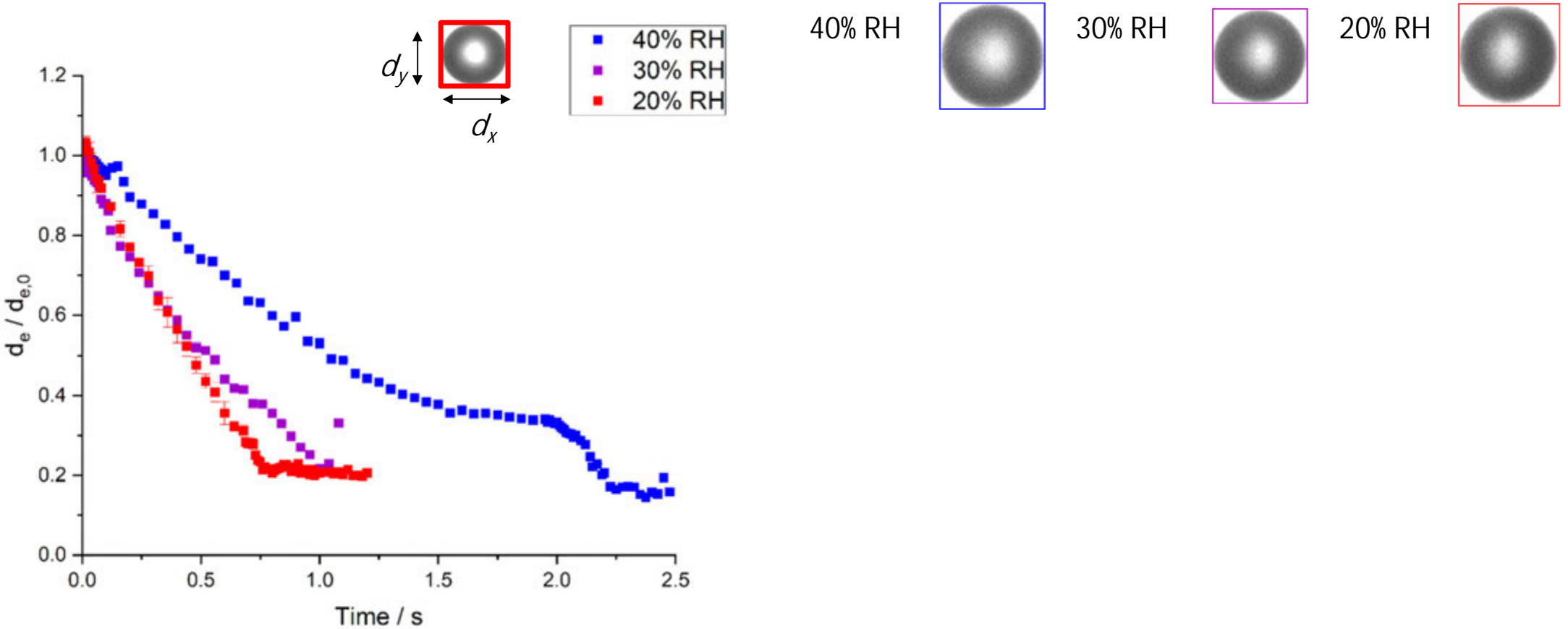
- Solute, not colloidal
- Ideal solute
- Well known behaviour in aerosol phase
- Clear solidification behaviour
  
- Relevant to food the food industry
- Nuclear industry involves mixed solutions

• Understand the phenomena that occur

• Apply to more complex scenarios

# 'In-flight' images of NaCl crystallisation

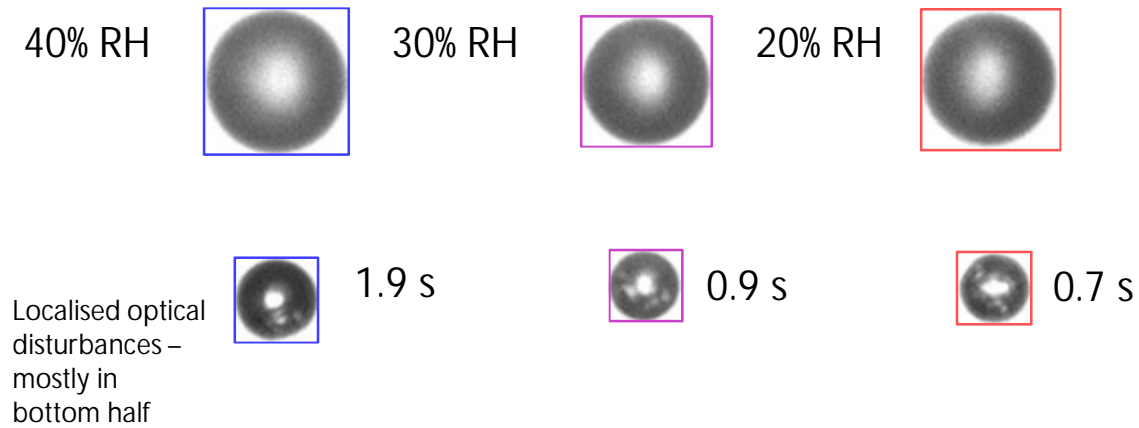
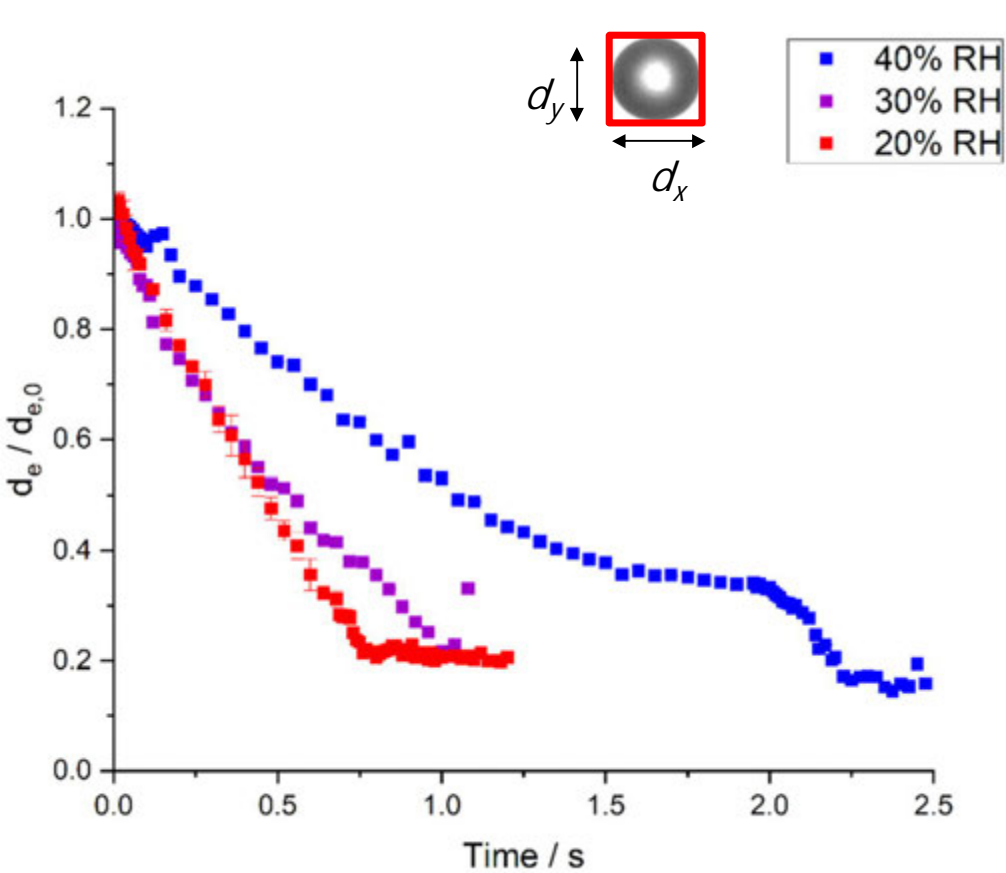
13 March 2021





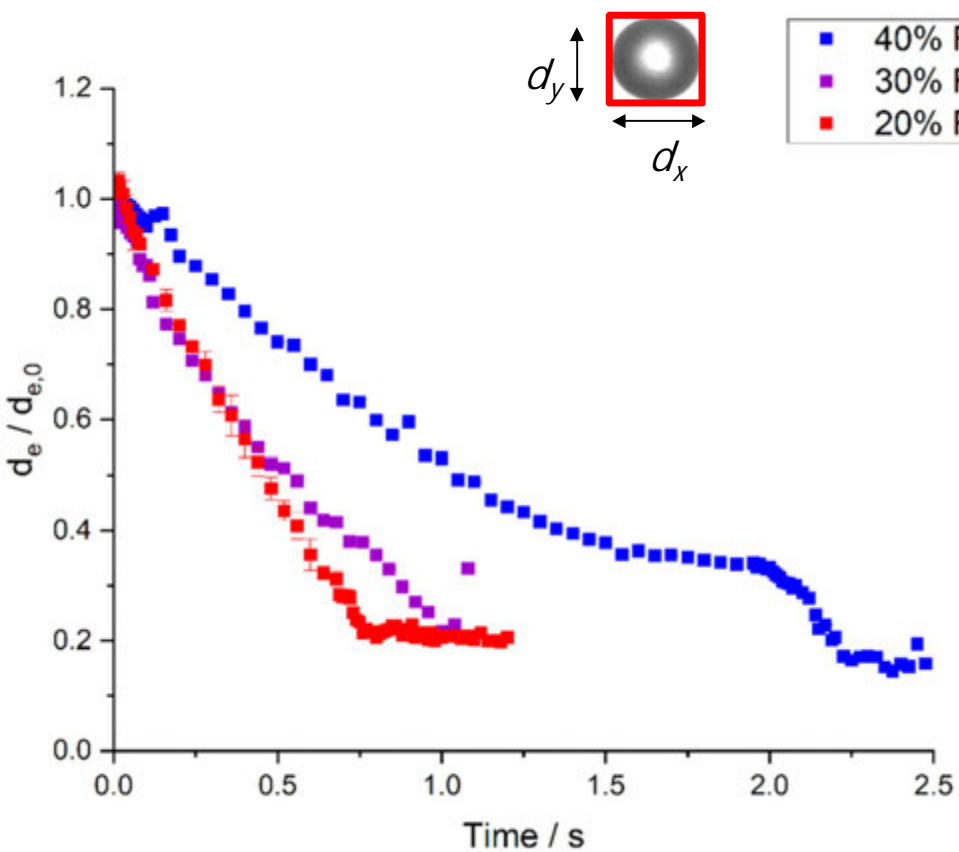
# 'In-flight' images of NaCl crystallisation

13 March 2021

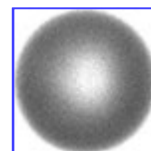


# 'In-flight' images of NaCl crystallisation

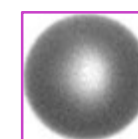
13 March 2021



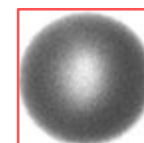
40% RH



30% RH



20% RH



Localised optical disturbances – mostly in bottom half



1.9 s



0.9 s



0.7 s

Loss of curvature from one side "half droplet, half crystal"



2.2 s



1.1 s

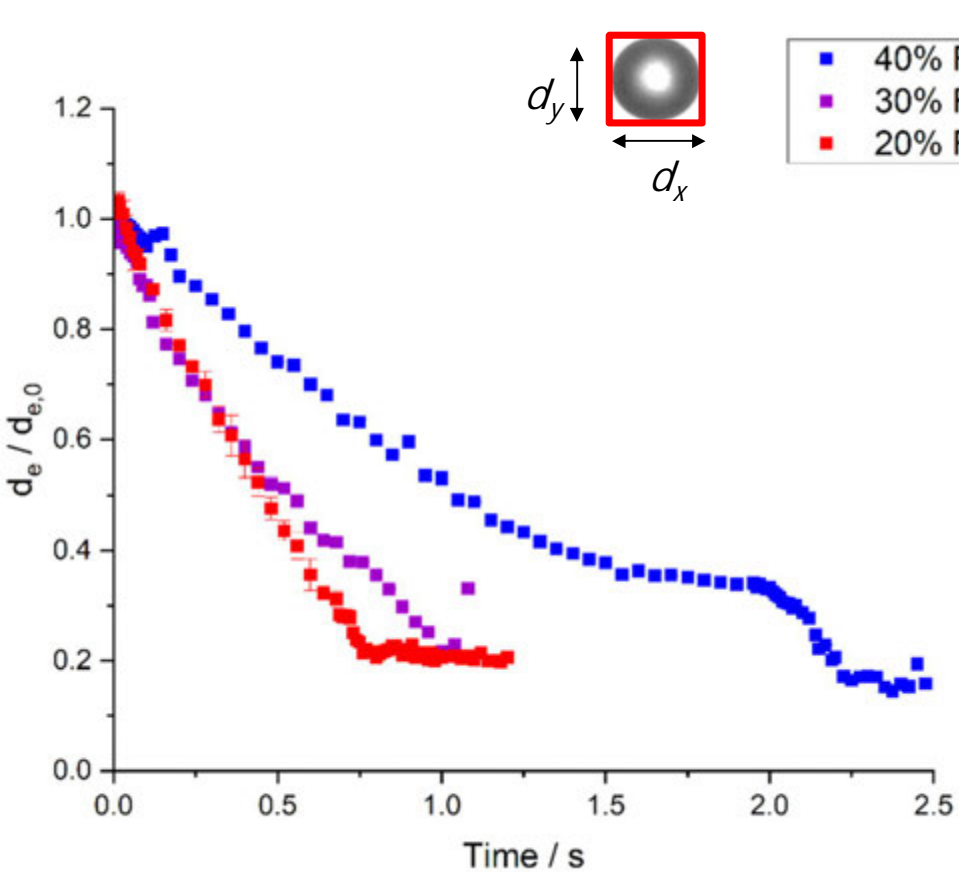
Optical disturbances all over droplet



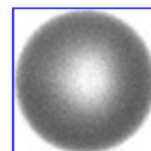
0.8 s

# 'In-flight' images of NaCl crystallisation

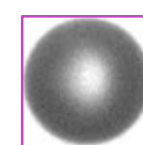
13 March 2021



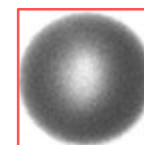
40% RH



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Localised optical disturbances – mostly in bottom half



1.9 s

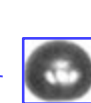


0.9 s



0.7 s

Loss of curvature from one side  
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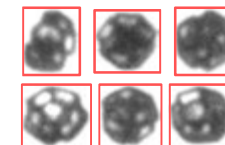
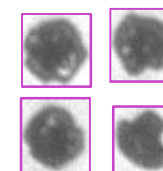
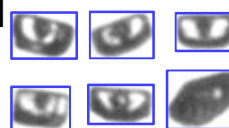
1.1 s



0.8 s

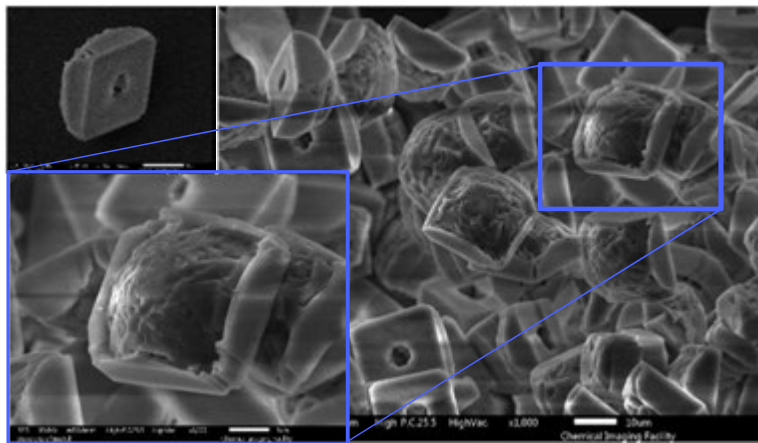
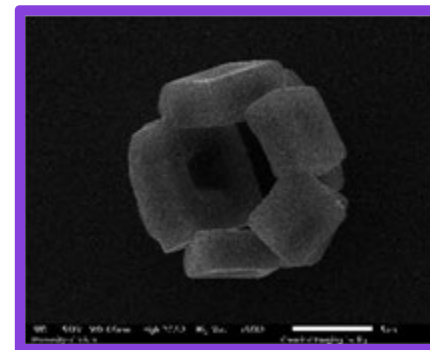
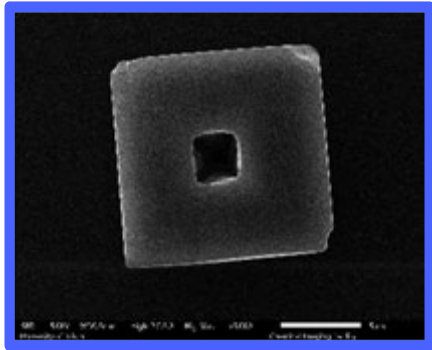
Single crystal structure

Typically curved side down



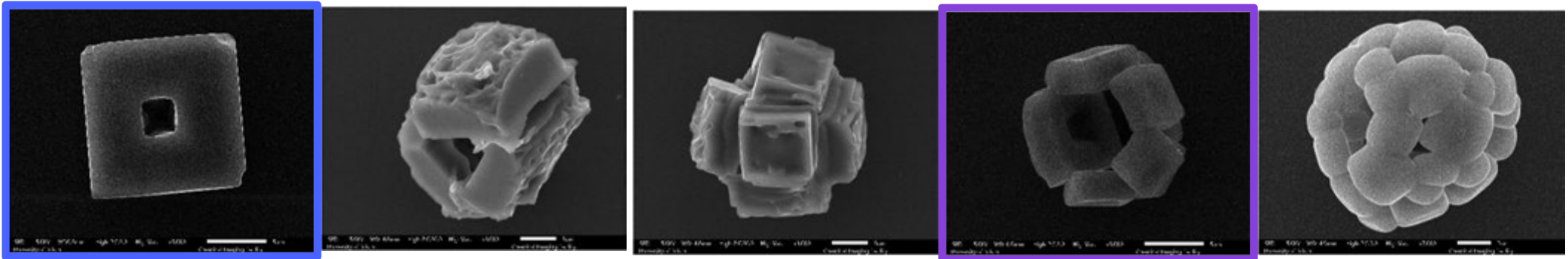
# SEM Analysis

## - Understanding Morphology



# SEM Analysis

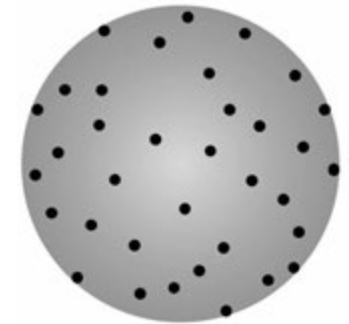
## – Understanding Morphology



Increasing evaporation rate,  $\kappa$

Peclet Number

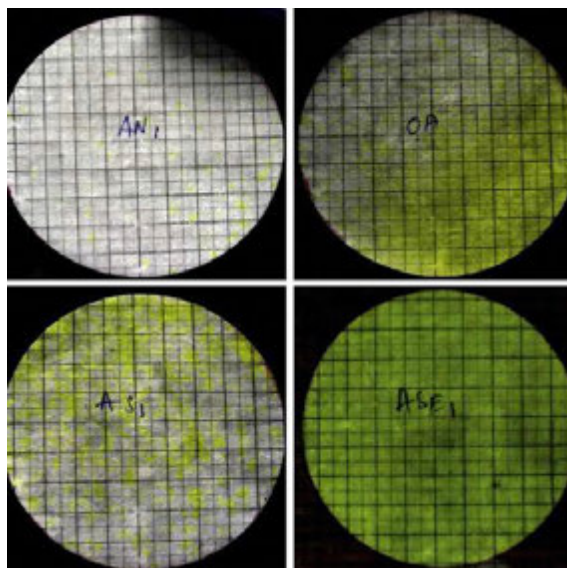
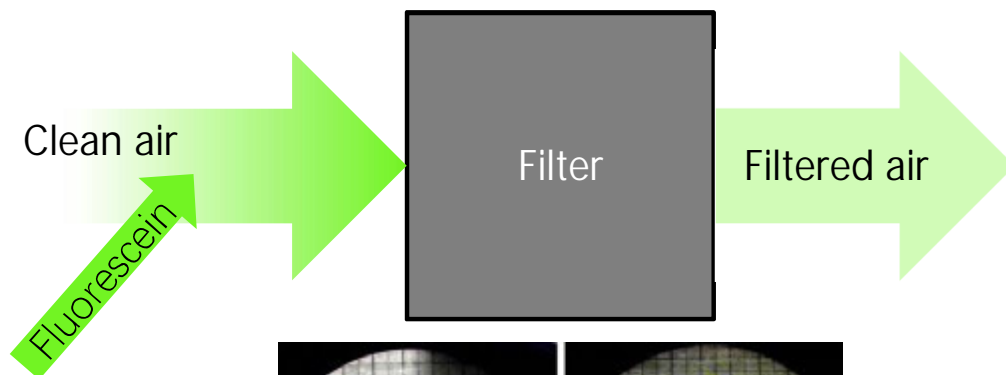
$$Pe = \frac{\kappa(RH, T)}{8D(T)}$$





# Real World Systems - Sodium Fluorescein

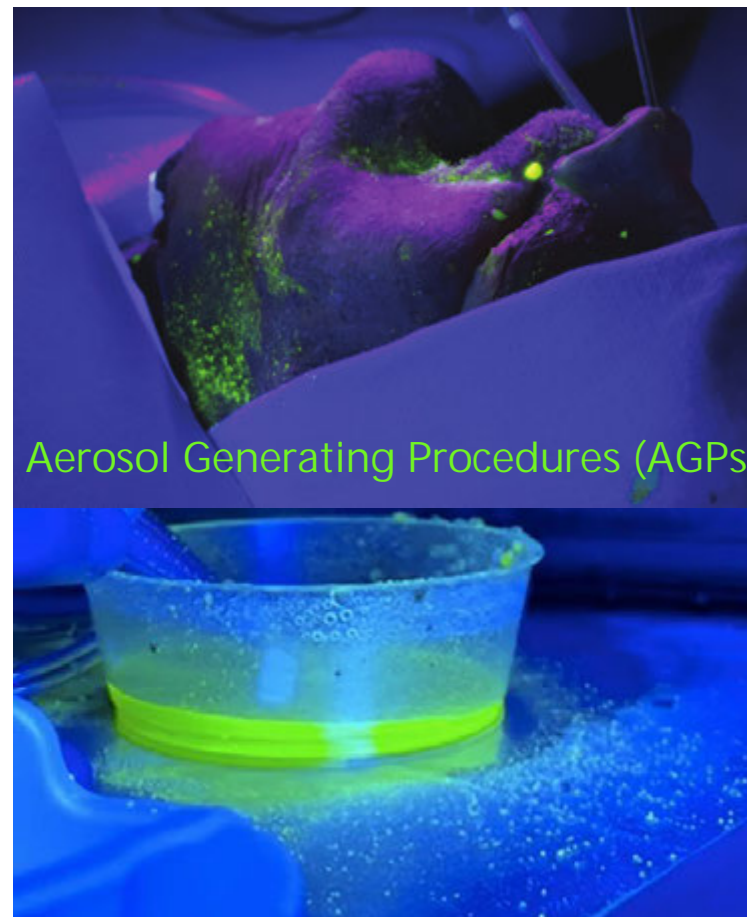
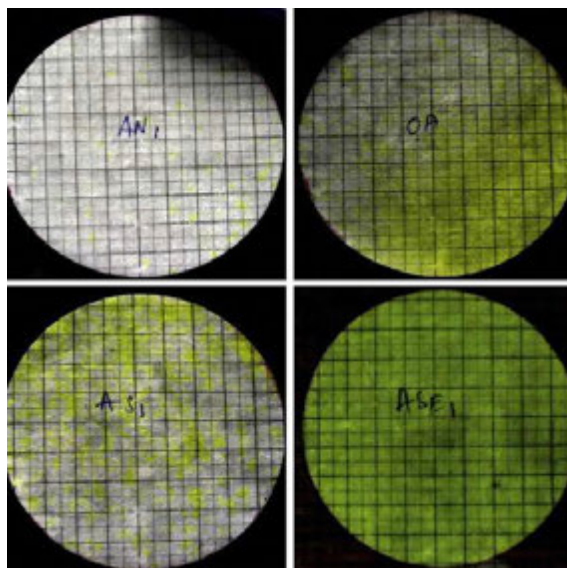
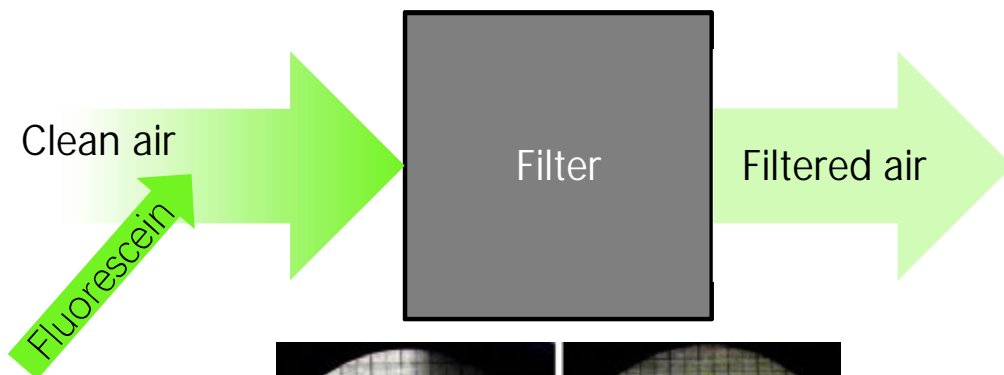
13 March 2021



RH 20% ± 5%  
15°C ± 2°C

# Real World Systems - Sodium Fluorescein

13 March 2021



RH 20% ± 5%  
15°C ± 2°C

## Sodium fluorescein:

- Less ideal solute
- Slower diffusion

## Sodium fluorescein:

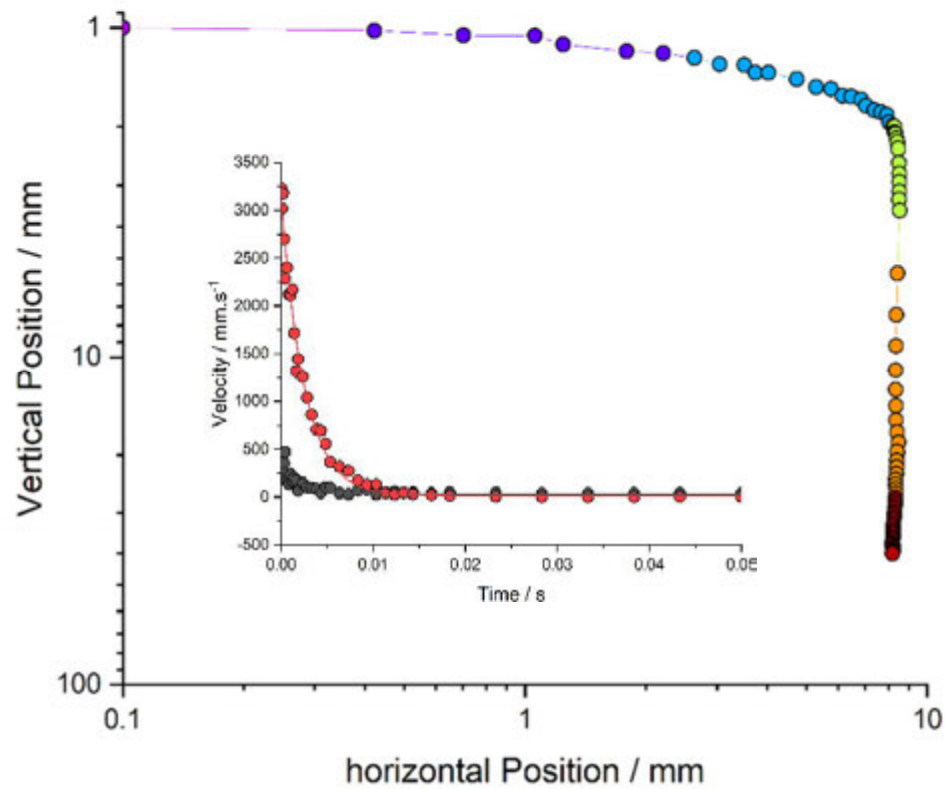
- Less ideal solute
- Slower diffusion
- Less well known behaviour in aerosol phase

## Sodium fluorescein:

- Less ideal solute
- Slower diffusion
- Less well known behaviour in aerosol phase
- Poorly understood solidification behaviour
  - Amorphous?
  - Solid?
  - Ruptured?



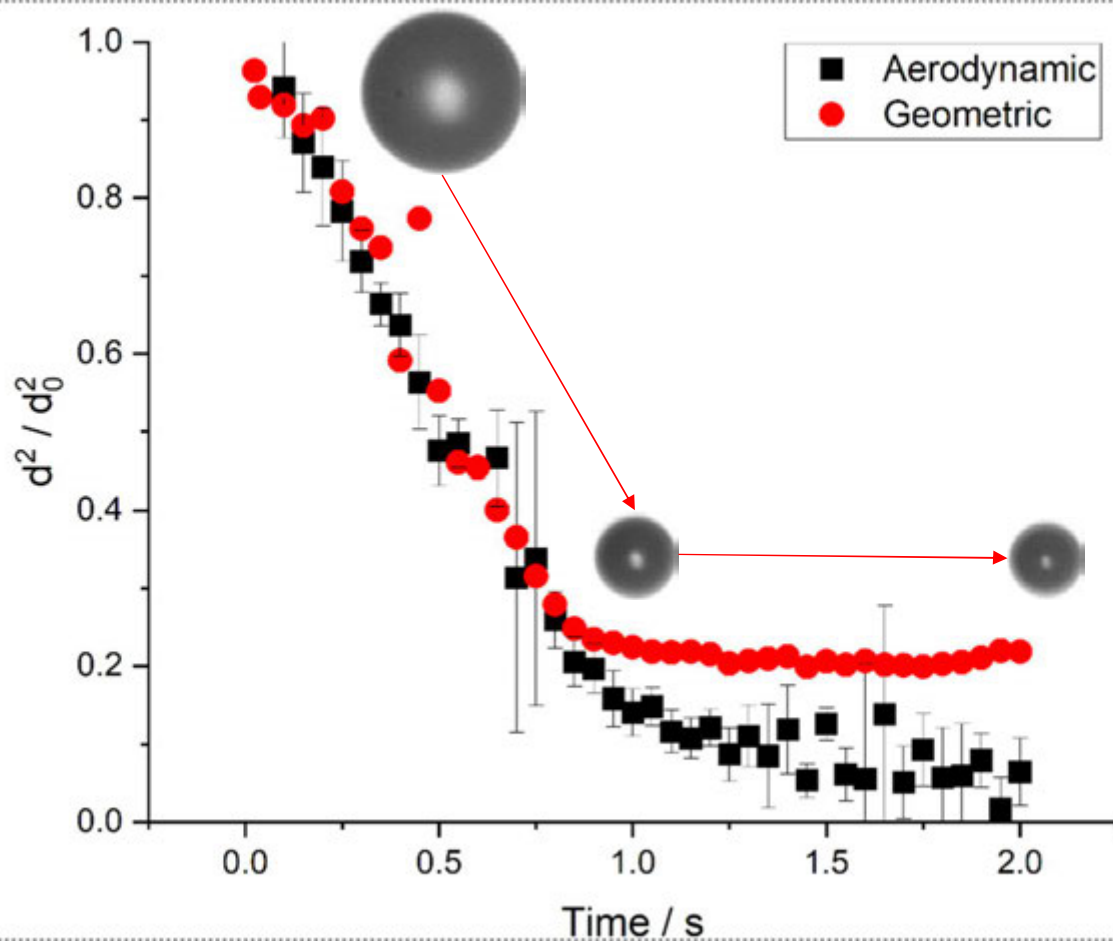
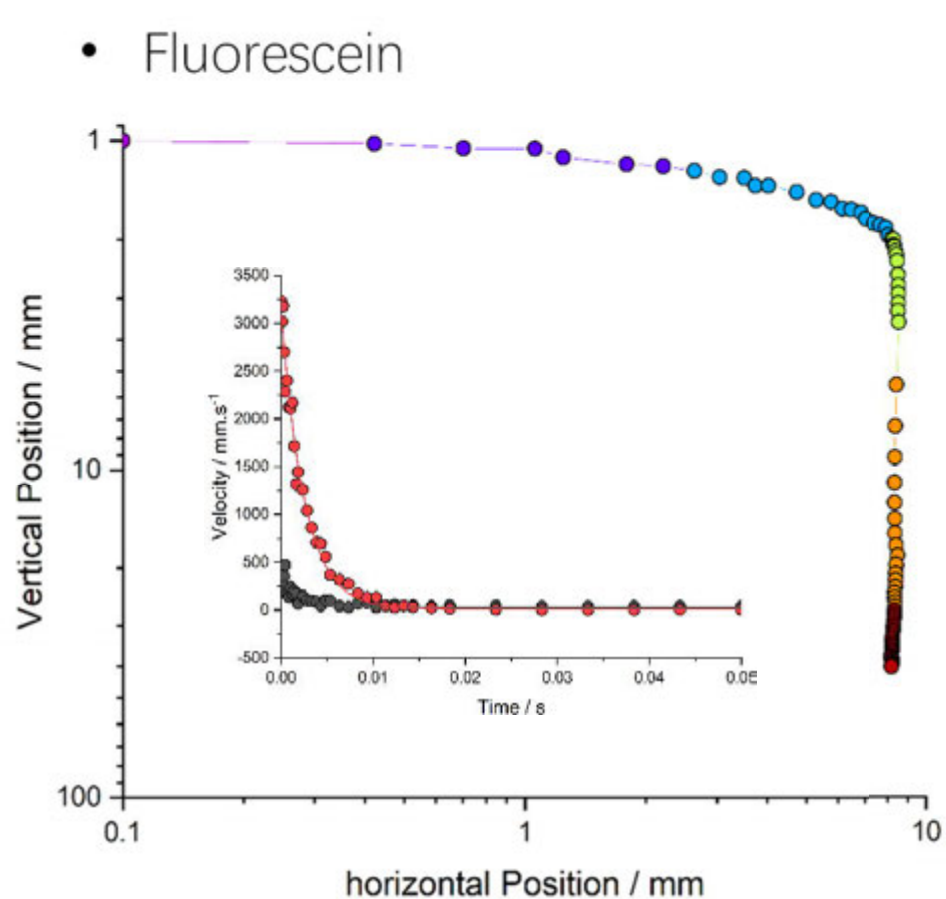
- Fluorescein



# Real World Systems - Sodium Fluorescein

13 March 2021

- Fluorescein



## Accurate Representations of the Microphysical Processes Occurring during the Transport of Exhaled Aerosols and Droplets

Jim S. Walker, Justice Archer, Florence K. A. Gregson, Sarah E. S. Michel, Bryan R. Bzdek, and Jonathan P. Reid\*

Cite This: <https://dx.doi.org/10.1021/acscentsci.0c01522>

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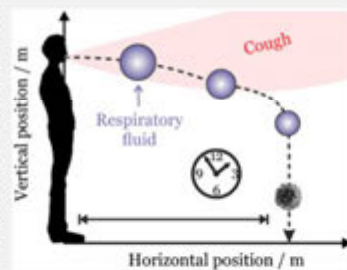
ACCESS |

Metrics & More

Article Recommendations

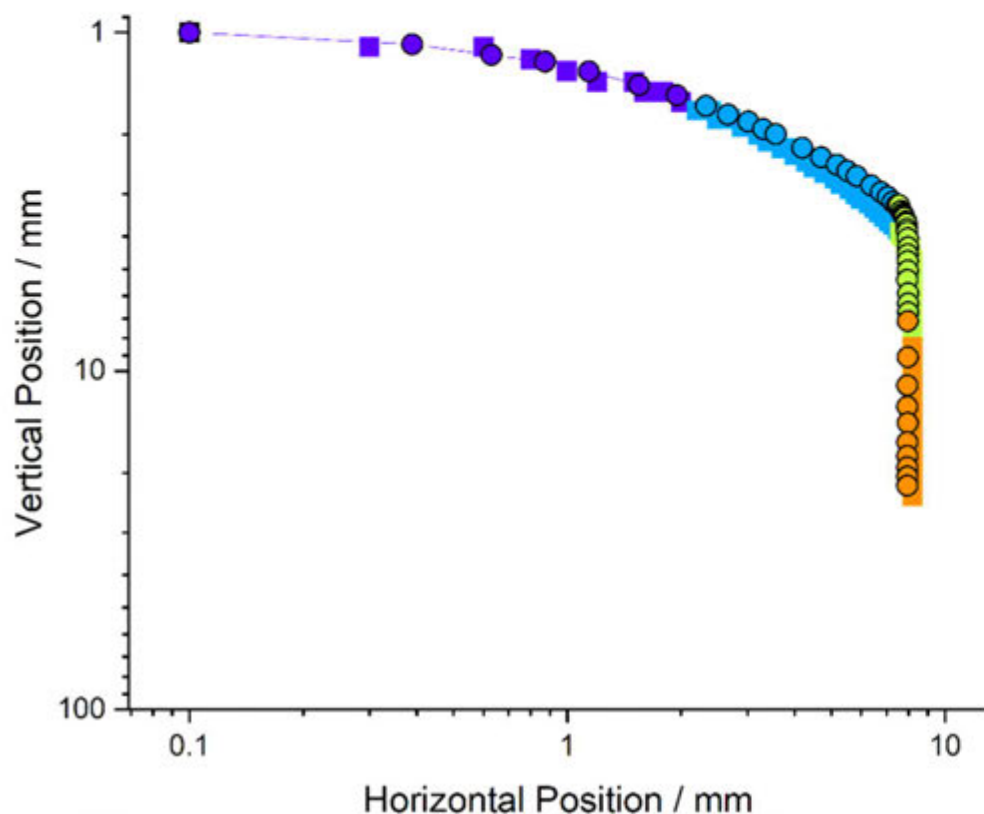
Supporting Information

**ABSTRACT:** Aerosols and droplets from expiratory events play an integral role in transmitting pathogens such as SARS-CoV-2 from an infected individual to a susceptible host. However, there remain significant uncertainties in our understanding of the aerosol droplet microphysics occurring during drying and sedimentation and the effect on the sedimentation outcomes. Here, we apply a new treatment for the microphysical behavior of respiratory fluid droplets to a droplet evaporation/sedimentation model and assess the impact on sedimentation distance, time scale, and particle phase. Above a 100  $\mu\text{m}$  initial diameter, the sedimentation outcome for a respiratory droplet is insensitive to composition and ambient conditions. Below 100  $\mu\text{m}$ , and particularly below 80  $\mu\text{m}$ , the increased settling time allows the exact nature of the evaporation process to play a significant role in influencing the sedimentation outcome. For this size range, an incorrect treatment of the droplet composition, or imprecise use of RH or temperature, can lead to large discrepancies in sedimentation distance (with representative values >1 m, >2 m, and >2 m, respectively). Additionally, a respiratory droplet is likely to undergo a phase change prior to sedimenting if initially <100  $\mu\text{m}$  in diameter, provided that the RH is below the measured phase change RH. Calculations of the potential exposure versus distance from the infected source show that the volume fraction of the initial respiratory droplet distribution, in this size range, which remains elevated above 1 m decreases from 1 at 1 m to 0.125 at 2 m.



$$\left\{ \begin{aligned} \frac{dr_p}{dt} &= \frac{CM_v D_\infty p Sh}{\rho_p r_p RT_\infty} \ln\left(\frac{p - p_{va}}{p - p_{v\infty}}\right) = f_1(r_p, T_p, \vec{V}_p) \\ \frac{dT_p}{dt} &= 3K_g \frac{T_\infty - T_p}{c_p r_p^2} Nu - \frac{L_v I}{m_p c_p} - \frac{3\Gamma(T_p^4 - T_\infty^4)}{r_p c_p} = f_2(r_p, T_p, \vec{V}_p) \\ \frac{d\vec{V}_p}{dt} &= \vec{g} \left(1 - \frac{\rho_p}{\rho_g}\right) - \frac{3C_d \rho_g |\vec{V}_p - \vec{V}_g| (\vec{V}_p - \vec{V}_g)}{8\rho_p r_p} = f_3(r_p, T_p, \vec{V}_p) \\ \frac{d\vec{x}_p}{dt} &= \vec{V}_p = f_4(\vec{V}_p) \end{aligned} \right.$$

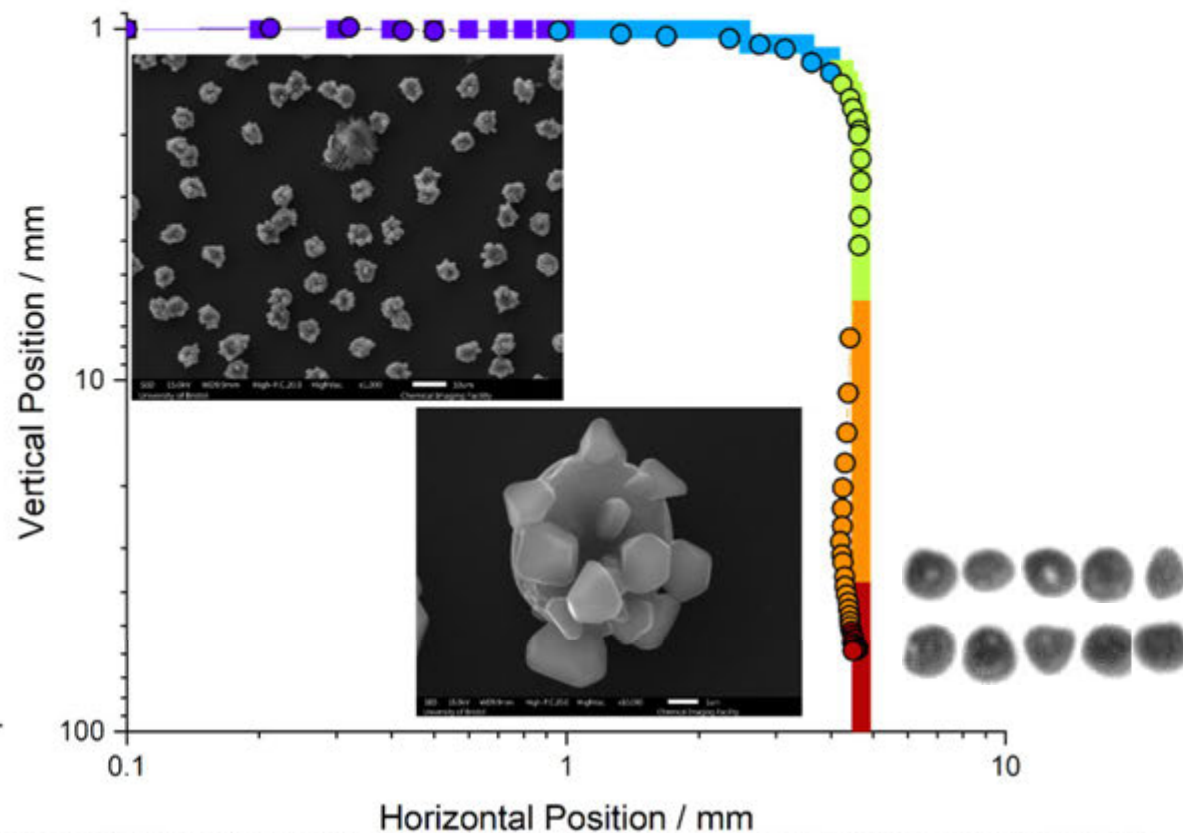
- Artificial Saliva



RH 20% ± 5%  
15°C ± 2°C

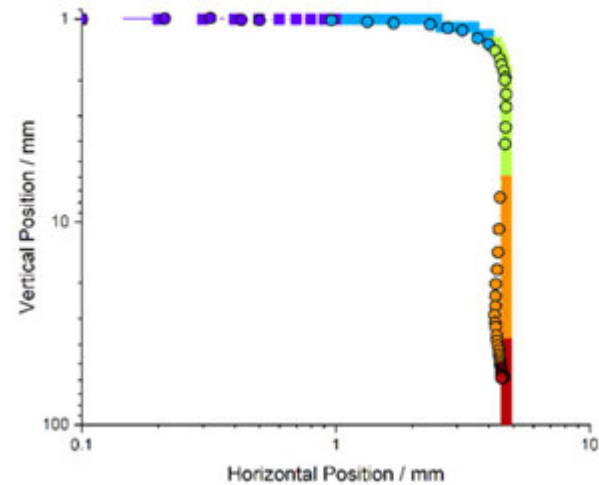
RH 5% ± 5%  
20°C ± 2°C

- Deep Lung Fluid



## Falling Droplet Column Capability

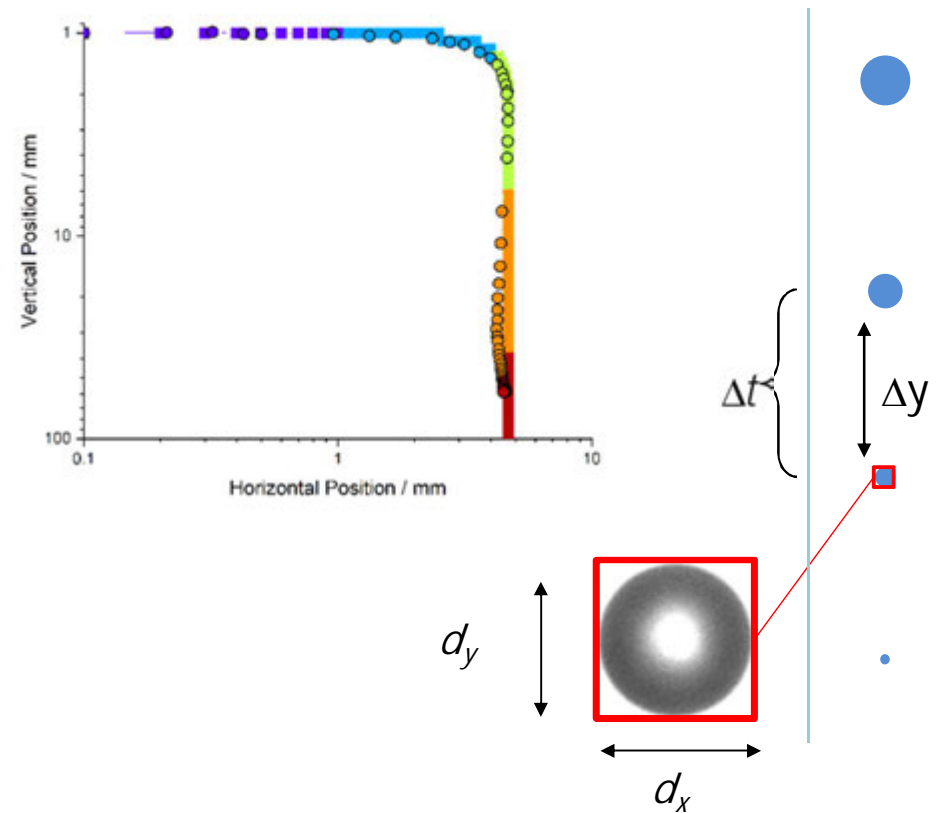
- Observe and model freefalling droplet trajectories





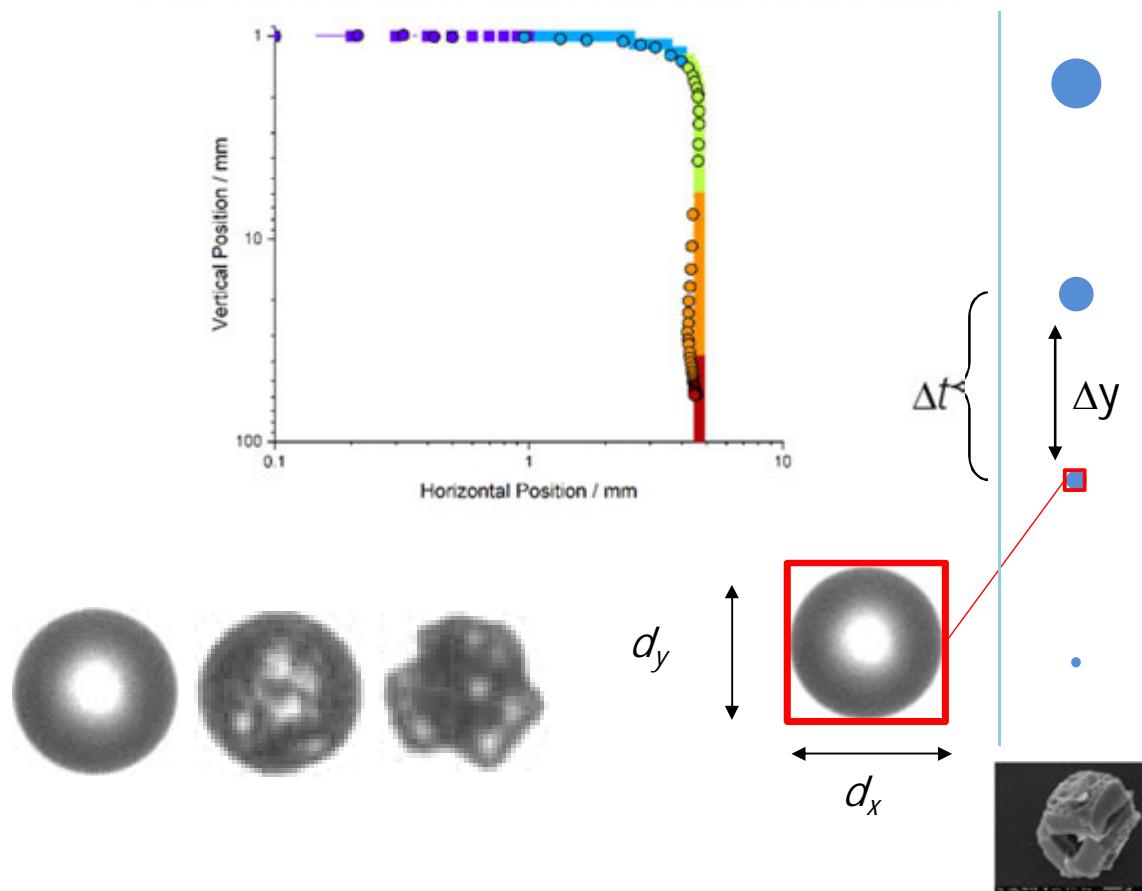
## Falling Droplet Column Capability

- Observe and model freefalling droplet trajectories
- Image droplets throughout drying process
- Measure geometric and aerodynamic diameter



## Falling Droplet Column Capability

- Observe and model freefalling droplet trajectories
- Image droplets throughout drying process
- Measure geometric and aerodynamic diameter
- Observe phase changes
- SEM analysis of dry particles



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Jim Walker

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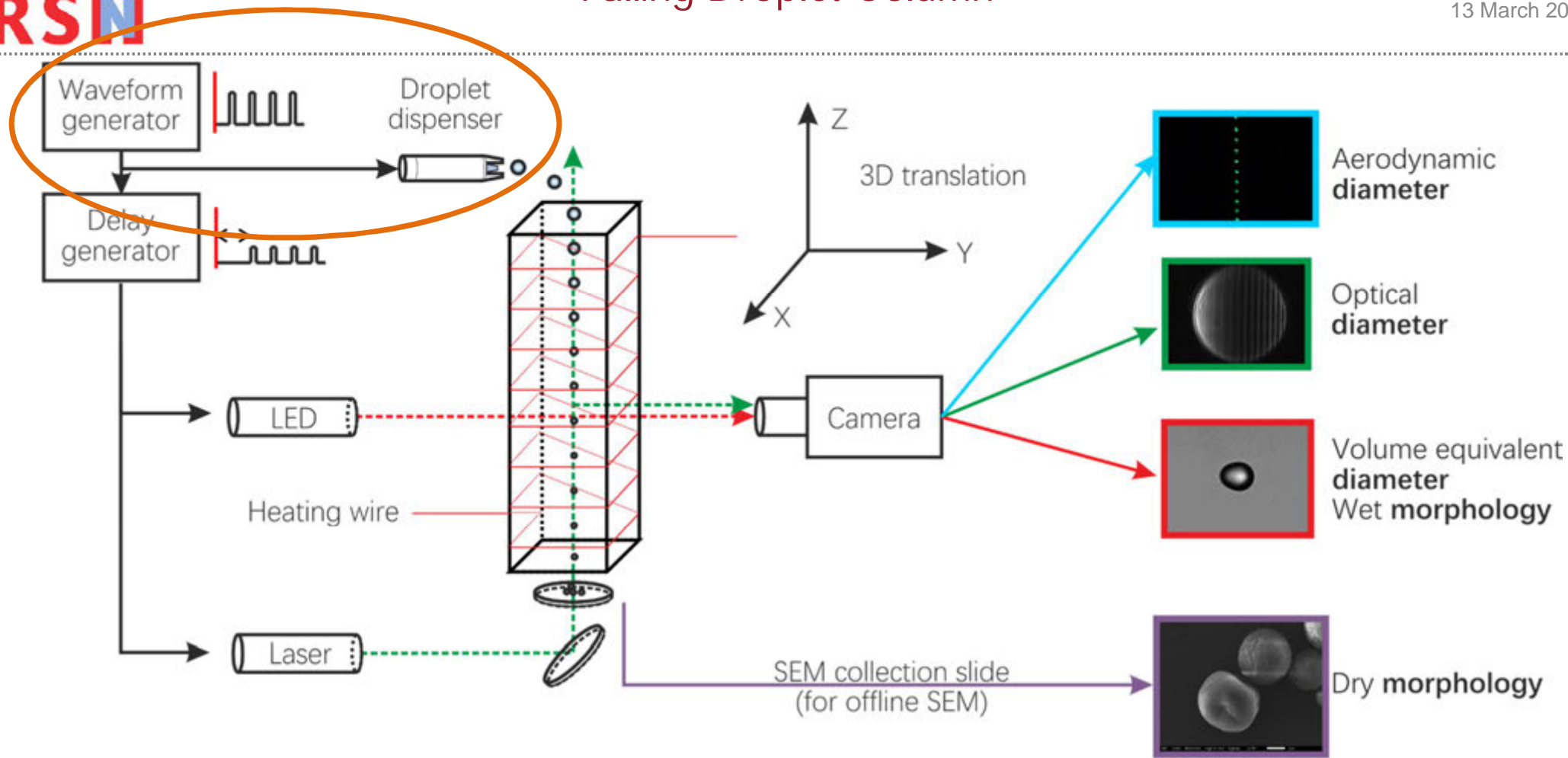


Thank you for listening



# Falling Droplet Column

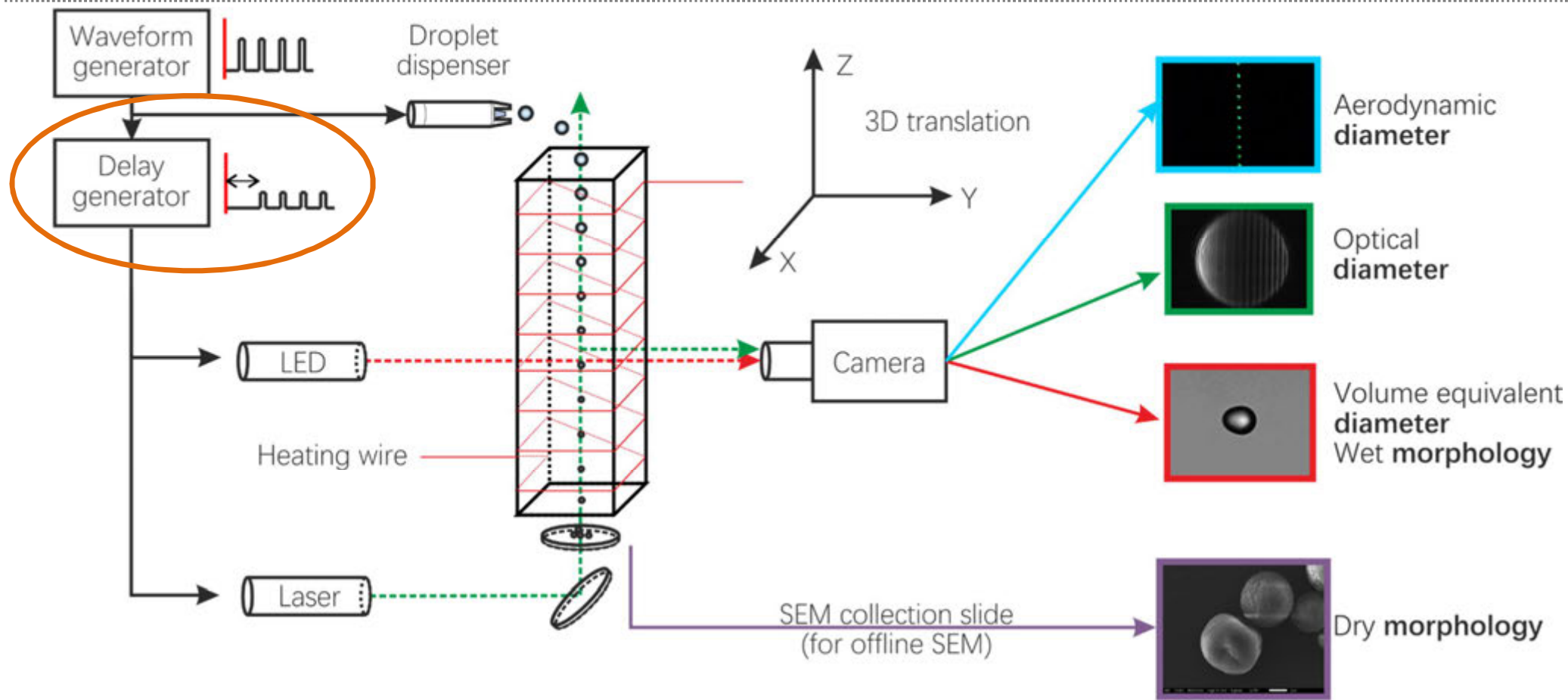
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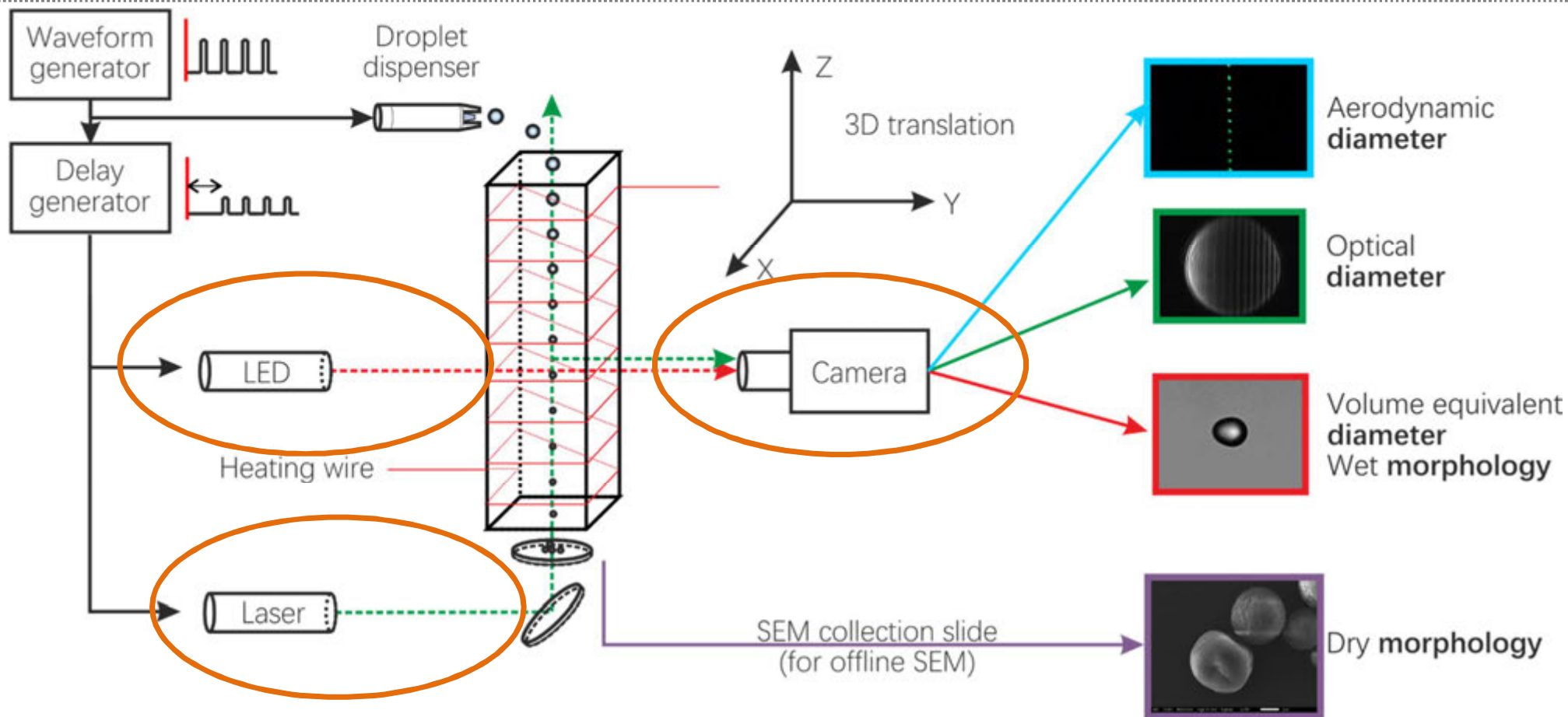
## Falling Droplet Column

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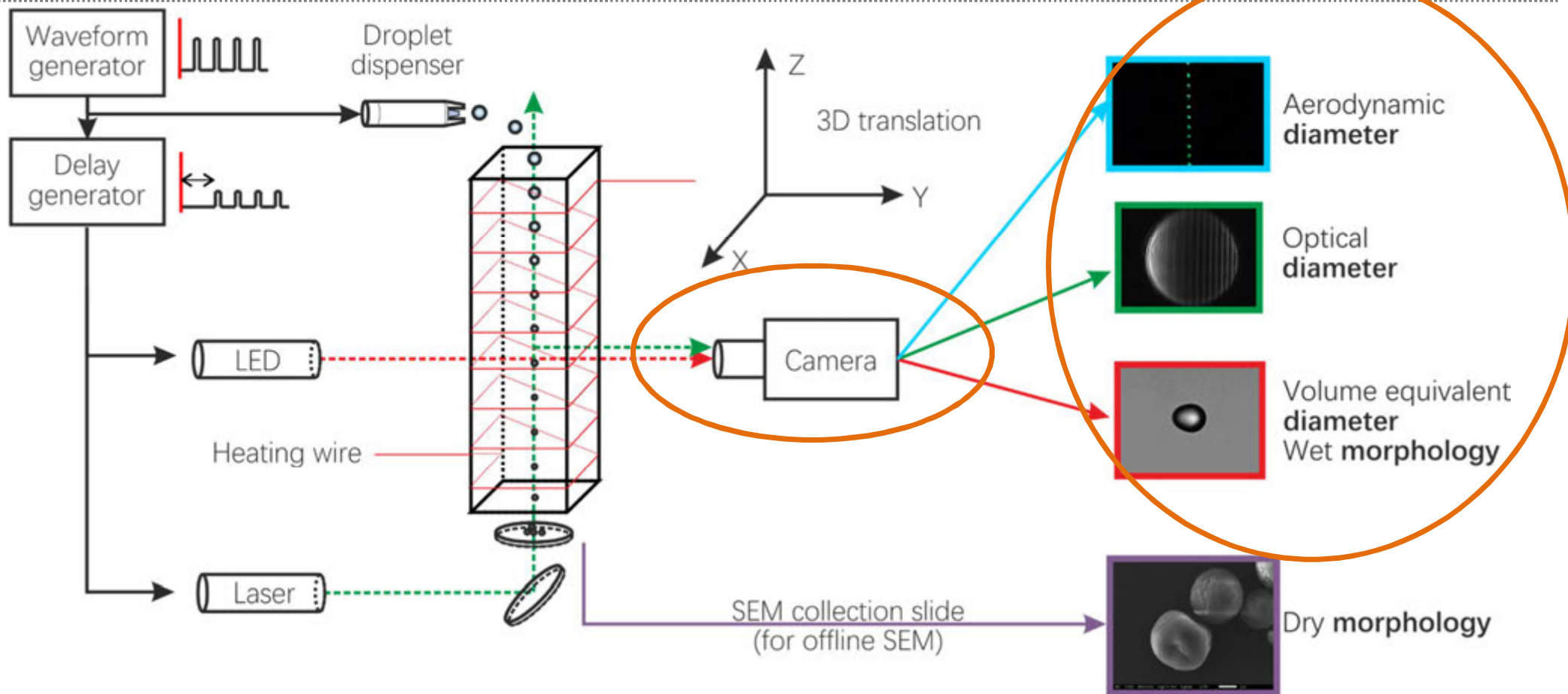
## Falling Droplet Column

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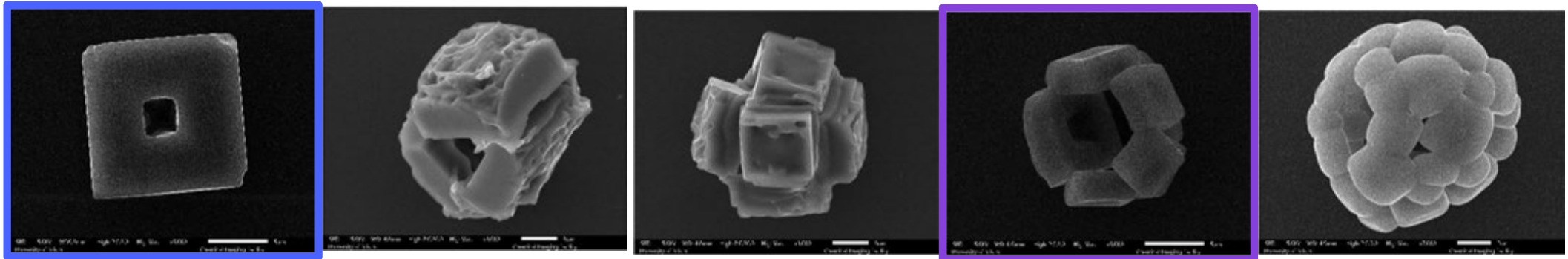
## Falling Droplet Column

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# SEM Analysis

## – Understanding Morphology



Increasing evaporation rate,  $\kappa$      $\uparrow$  Temperature     $\downarrow$  Relative Humidity

Pe < 1 Diffusion dominates, homogeneity maintained

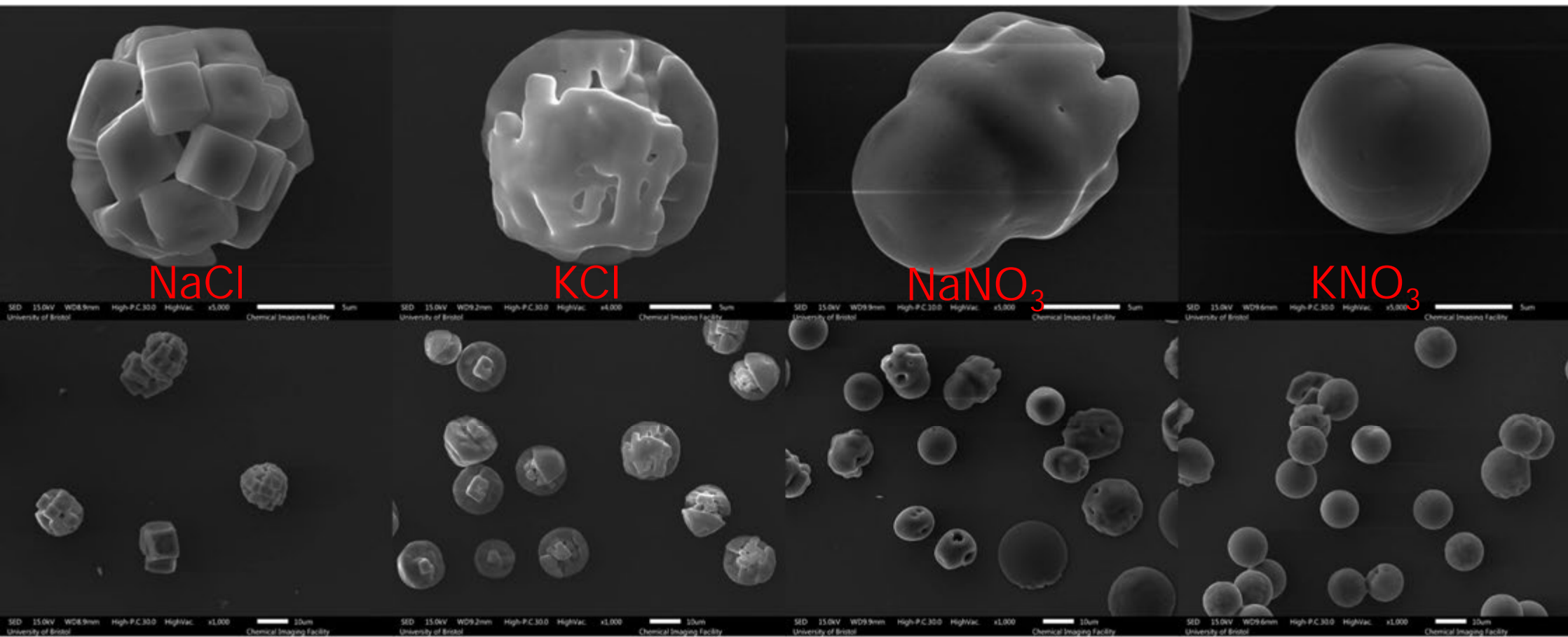
- Fewer nucleation sites
- Crystal growth on earliest nucleation site

Peclet Number

$$Pe = \frac{\kappa(RH, T)}{8D(T)}$$

Pe > 1 Leads to surface enrichment

- Many nucleation sites
- Parallel crystal growth
- Surface crust / shell formation



- 
- Further experiments on:
    - Industrially relevant compounds
    - Compounds forming amorphous particles
  - Parameterisation of sodium fluorescein aerosol properties
    - For use in model
  - Further development of model
    - To include surface enrichment



## Future Work

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- Image analysis
  - Particle morphology
  
- Particle orientation

