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Online Classification of Soot Aggregate Aerosols by Morphology

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T.J. Johnson[1], Xiao Zhang[1], Robert Nishida[1], J.P.R Symonds[2], J.S. Olfert[3] and A.M. Boies[1]

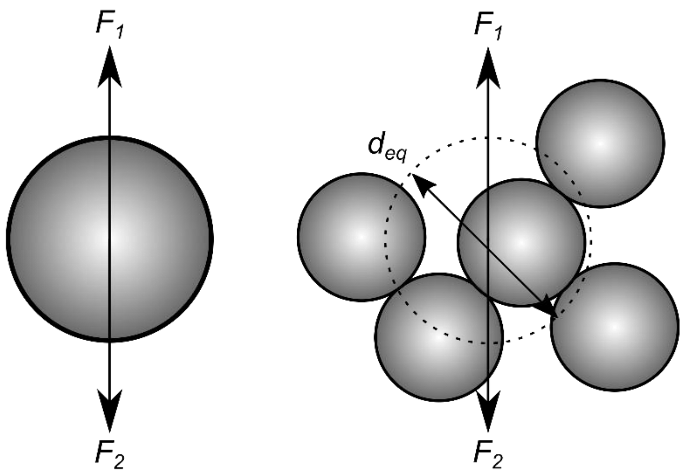
[1]Division A: Energy, Fluid Mechanics and Turbomachinery

Motivations

- Particles of different sizes, masses or morphologies often have different volatility, charging, chemical or hygroscopic properties.
- Benefits of a truly monodispersed source:
  - Probe physics of non-spherical particles on-line;
  - Characterize structure of non-spherical particles; and
  - Calibrate other devices using non-spherical particles.

Monodisperse Classification	On-Line Classifier	Truly monodispersed, considering:	
		Spherical	Non-Spherical
Electrical Mobility, d_m	DMA	✗ **(Multiple-Charging)**	✗
Aerodynamic, d_a	AAC	✓	✗
Mass, m	CPMA	✗ **(Multiple-Charging)**	✗
"Morphology" = d_m, d_a and m	AAC-DMA	✓	✓

Equivalent Particle Diameters



Equivalent Diameter, d_{eq}	Force 1, F_1	Force 2, F_2
Aerodynamic Diameter, d_a	Centrifugal (mass)	Drag
Electrical Mobility Diameter, d_m	Electrostatic (charge)	Drag

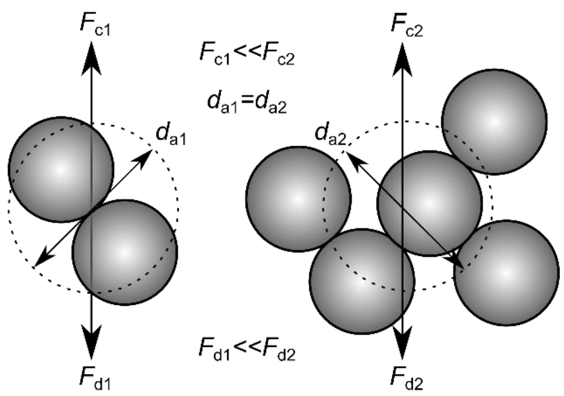
Particle Relaxation Time (τ):

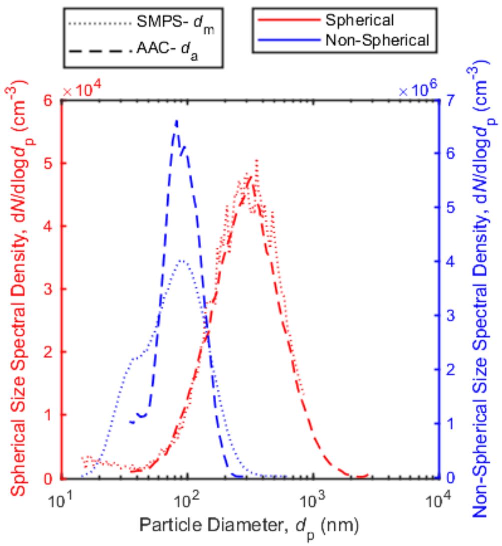
$$\tau = m \cdot B = \frac{C_c(d_a) \cdot \rho_o \cdot d_a^2}{18\mu} = \frac{C_c(d_m) \cdot \rho_{eff} \cdot d_m^2}{18\mu} = \frac{C_c(d_{ve}) \cdot \rho_p \cdot d_{ve}^2}{18\mu \cdot \chi}$$

where d_{ve} is the volume equivalent diameter, the diameter of a sphere with the same volume as the particle of interest.

Challenges of Non-Spherical Particles

- For spherical, homogeneous particles being monodispersed in one domain, such as aerodynamic diameter (d_a), translates directly to others, such as particle mobility diameter (d_m) and mass (m).
- However this direct monodisperse translation between particle properties no longer occurs for non-spherical and/or non-homogenous particles.

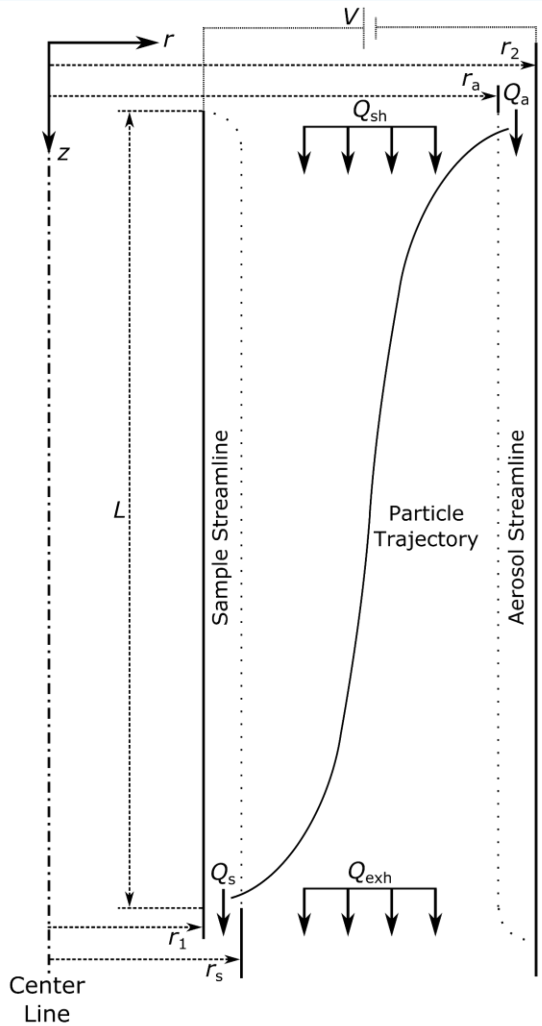




Example: Different particle masses have same aerodynamic diameter

- The effective density of aggregate morphologies often decrease with increasing particle size. Therefore, smaller particles with lower mass (∝ centrifugal force, F_c) and drag (F_d) may have the same relaxation time (τ) or aerodynamic diameter (d_a) as larger particles with higher mass and drag.
- This characteristic often results in non-spherical particle sources having a “narrow” aerodynamic size distribution.

Differential Mobility Analyzer (DMA) – Charge:Drag



- Classifier consists of two concentric cylinders with a potential voltage (V) between them.
- Classifies aerosol by particle electrical mobility (Z_p) – a particle's ability to move from an known electrostatic force. This is directly related to Mobility Diameter (d_m):

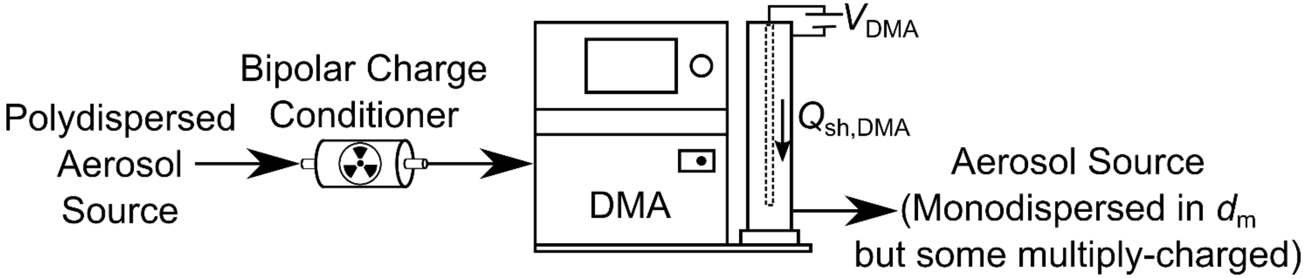
Cunningham slip

$$Z_\mathrm{p} = neB = \frac{neC_c(d_\mathrm{m})}{3\pi\mu d_\mathrm{m}} = \frac{Q_\mathrm{sh}}{2\pi VL}\ln\left(\frac{r_2}{r_1}\right)$$

\# of charges mobility

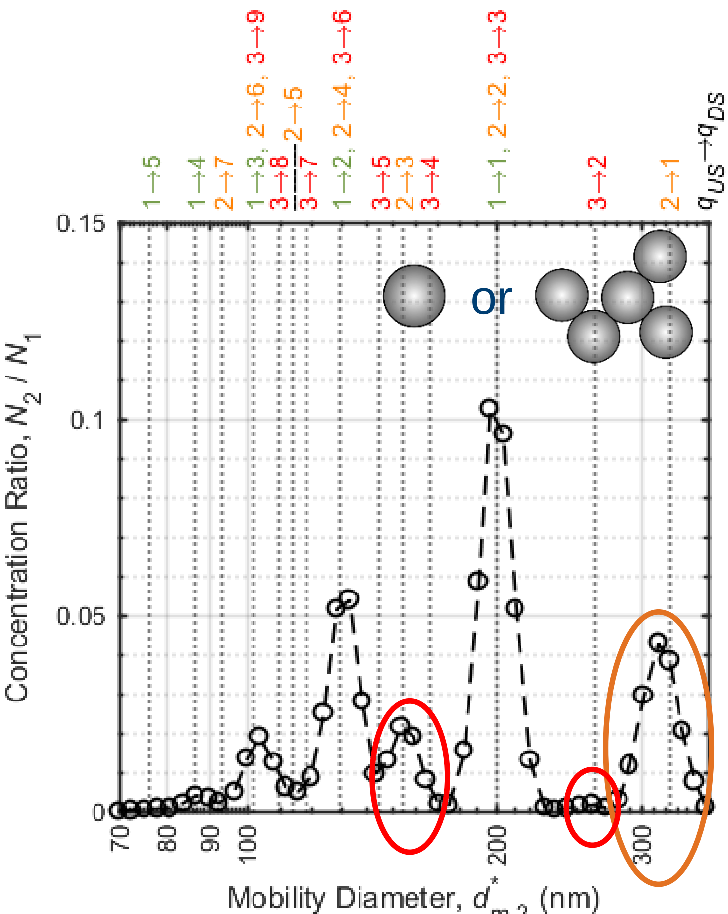
- Gas flows (dominated by sheath flow, Q_sh) move the particles axially, while the electrostatic force moves the particles radially.
- Smaller and/or higher charged particles are dominated by the electrostatic force and impact the inner cylinder.
- Bigger and/or lower charged particles are dominated by their drag, limiting their radial movements and thus remaining entrained in the sheath flow.

Common Approach to Generate "Monodisperse" Sources

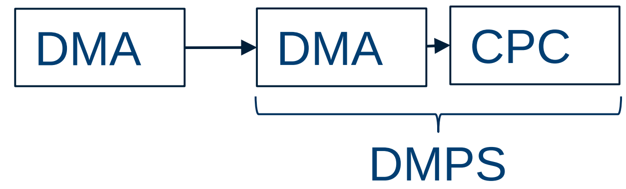


- A common solution to remove multiply-charged particle from the DMA is an impactor, but its effectiveness is limited for non-spherical particles due to:
  - Low resolution and discrete setpoints of the impactor; and
  - Narrow aerodynamic size distribution of non-spherical sources.
- Also if the particles are non-spherical being monodisperse in mobility diameter may not translate to other domains. For example, two aggregates could have the same drag, but drastically different masses.

DMA Challenges: Multiple-Charging



Measured mobility equivalent of re-neutralized particles classified by a DMA



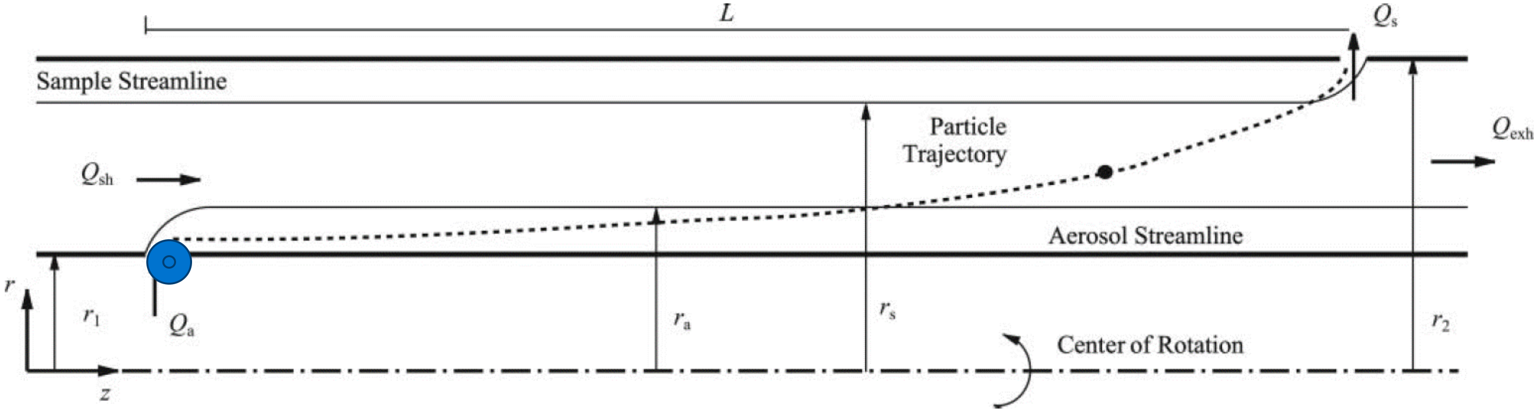
- Identify if multiply-charged particles are being classified in the DMA (i.e. orange and red labels) by:
  - Peaks above upstream DMA setpoint ($d^*_{\mathrm{m1},q=1} = 200\ \mathrm{nm}$):

    $q_{\mathrm{US}} = 2 \rightarrow q_{\mathrm{DS}} = 1$ and $q_{\mathrm{US}} = 3 \rightarrow q_{\mathrm{DS}} = 2$
  - A blended peak between the peaks for downstream charge states 1 ($d^*_{\mathrm{m2},q=1} = 200\ \mathrm{nm}$) and 2 ($d^*_{\mathrm{m2},q=2} = 129.4\ \mathrm{nm}$):

    $q_{\mathrm{US}} = 2 \rightarrow q_{\mathrm{DS}} = 3$ and $q_{\mathrm{US}} = 3 \rightarrow q_{\mathrm{DS}} = 4\ \&\ 5$
- Challenging to determine what portion of each peak are actually multiply-charged particles from the upstream DMA.

Aerodynamic Aerosol Classifier (AAC) – Mass:Drag

- Can be thought of like a “rotating DMA” – has axial sheath flow, but radial force is centrifugal not electrostatic



Adapted from F. Tavakoli & J. S. Olfert (2013)

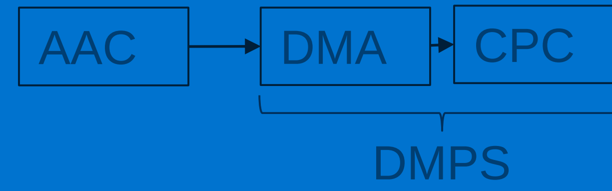
- Classifies aerosol by particle relaxation time (τ) – the time taken for a particle to match the flow to which it is introduced. This is directly related to Aerodynamic Diameter (d_a):
  - Smaller particles match the sheath flow sooner
  - Larger particles do not match the sheath flow

$$\tau = mB = \frac{C_c(d_a)\rho_0 d_a^2}{18\mu} = \frac{2Q_{sh}}{\pi\omega^2(r_i + r_o)^2 L}$$

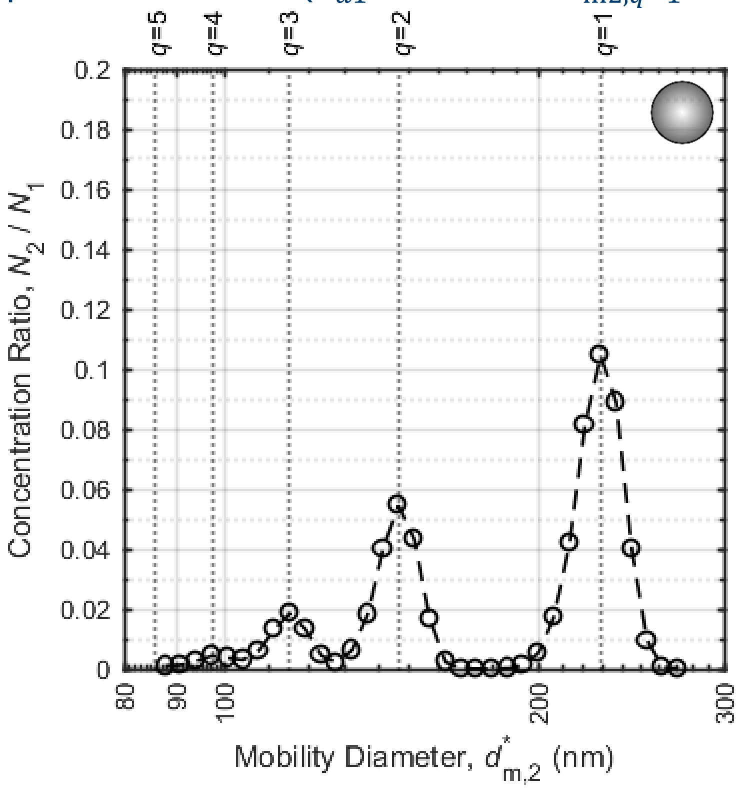
Cunningham slip; mass; mobility

- Doesn’t rely on particle charging—true monodisperse aerosol

AAC Challenges: Multiple-Masses

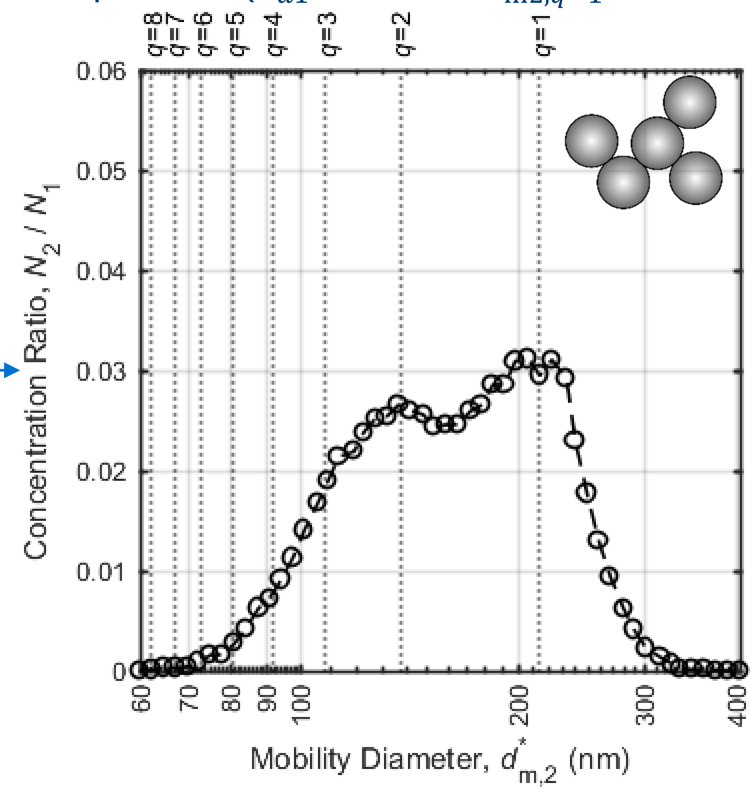


Measured Mobility Equivalent of AAC Classified Spherical Particles ($d_{a1}^{*} = 225$ nm, $d_{m2,q=1}^{*} = 229$ nm)



Broadening in Mobility Domain

Measured Mobility Equivalent of AAC Mini-CAST soot particles ($d_{a1}^{*} = 90$ nm, $d_{m2,q=1}^{*} = 214$ nm)



- As previously demonstrated, classifying non-spherical particles by d_a will be monodisperse in that domain, but may include different particle masses. These different masses causes "broadening" when the d_a monodispersed source is measured in other domains, such as mobility diameter (d_m).

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Multiple Domain Tandem Classifier System

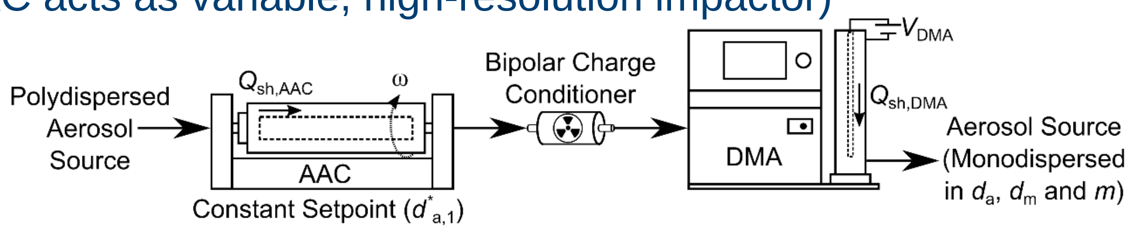
- Previous studies have demonstrated measuring the size-resolved effective density of particles with the following tandem classifier systems:
  - Tandem AAC-DMA (Tavakoli and Olfert, 2014)
  - Tandem DMA-CPMA (Olfert et al., 2007)
  - Tandem AAC-CPMA (Johnson et al., 2018)
- Any of these systems (i.e. two classifiers of different measurands in series) can also generate an aerosol source that is monodisperse in d_a, d_m and m, and therefore in morphology:

$$\tau = mB$$

AAC → τ, CPMA → m, DMA → B

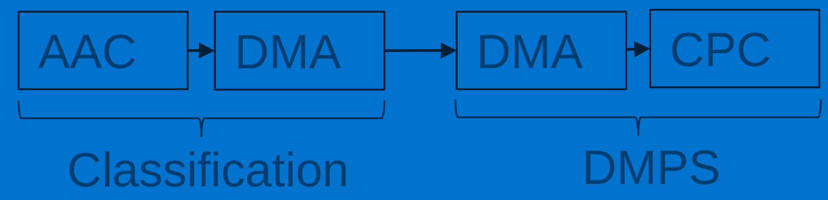
Assuming the particle charge states in the DMA and CPMA are known.

- However, **using an AAC in the tandem system is preferred** as its setpoint can be sufficiently lower than the second classifier (CPMA or DMA) **to limit multiple-charging effects** (i.e. AAC acts as variable, high-resolution impactor)



- A DMA-CPMA or CPMA-DMA system would still have multiple-charging effects

Tandem AAC-DMA System

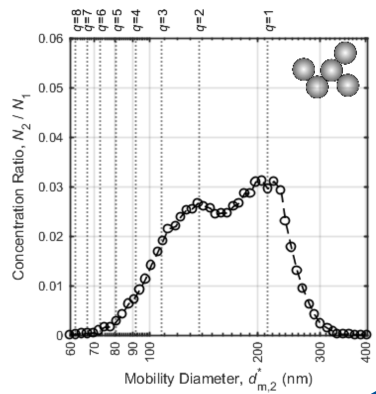


AAC Classified

Non-spherical particles classified by an AAC→Likely contain a range of particle masses

$d^*_{a1} = 90\ \mathrm{nm}$

$d^*_{\mathrm{m2},q=1} = 214\ \mathrm{nm}$



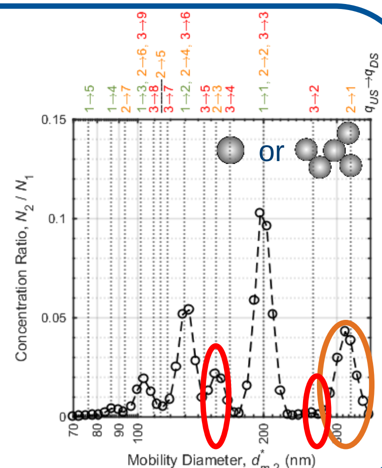
DMA Classified

Particles classified by DMA→Likely contain multiply-charged particles

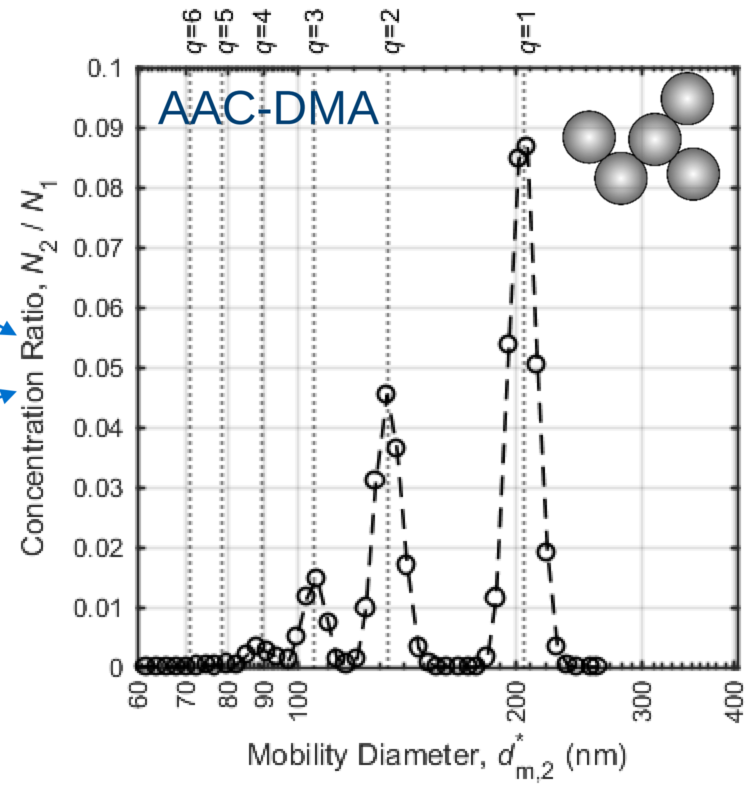
and if non-spherical→ May contain a range of particle masses

$d^*_{m1,q=1} = 200\ \mathrm{nm}$

$d^*_{\mathrm{m2},q=1} = 199\ \mathrm{nm}$



Measured mobility equivalent of re-neutralized, non-spherical particles classified by an AAC-DMA system

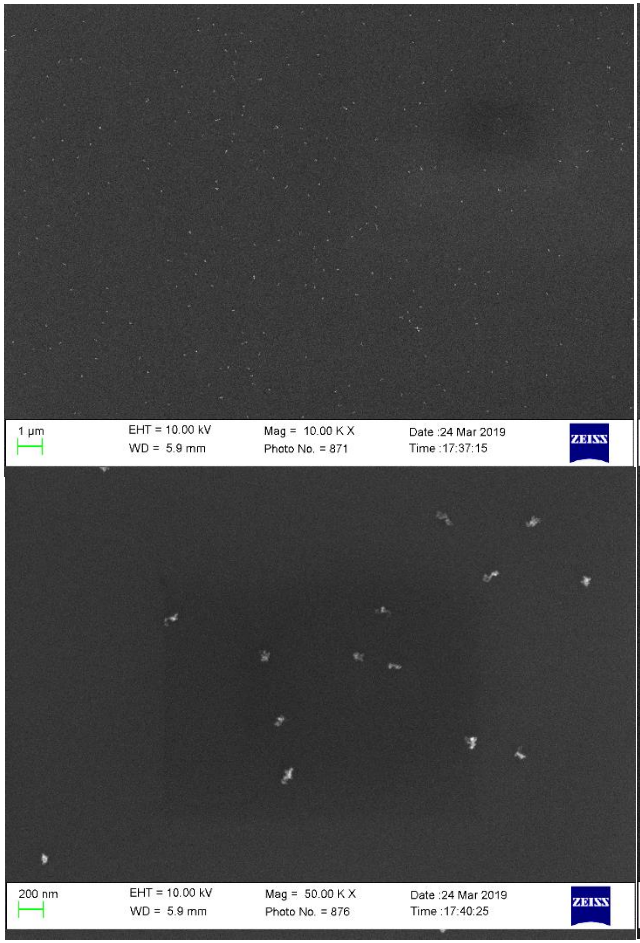


$d^*_{a1} = 56\ \mathrm{nm}$, $d^*_{\mathrm{m2},q=1} = 208\ \mathrm{nm}$ & $d^*_{\mathrm{m3},q=1} = 206\ \mathrm{nm}$

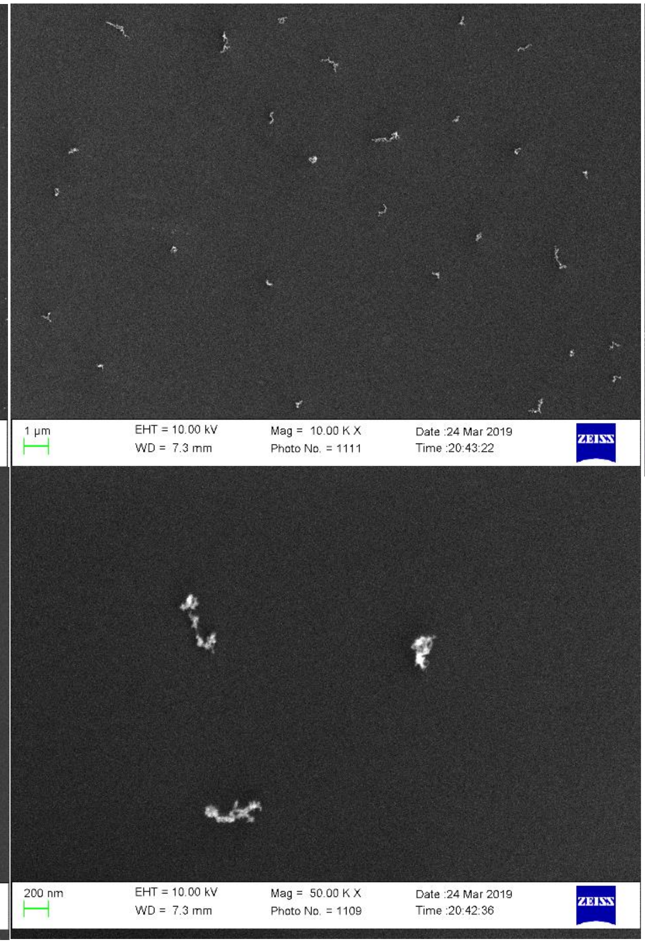
Therefore, a AAC-DMA system overcomes the classification challenges of a standalone AAC (different particle masses) or standalone DMA (multiply-charged particles and/or different particle masses).

Selecting Particle "Morphology" – SEM images of classified mini-CAST soot

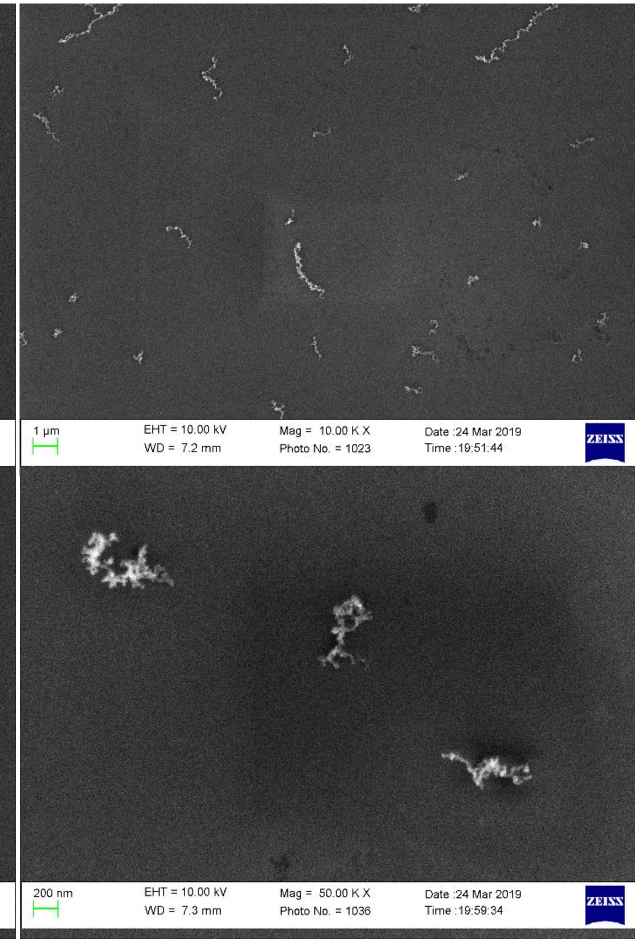
AAC 35 nm & DMA 80 nm



AAC 56 nm & DMA 150 nm

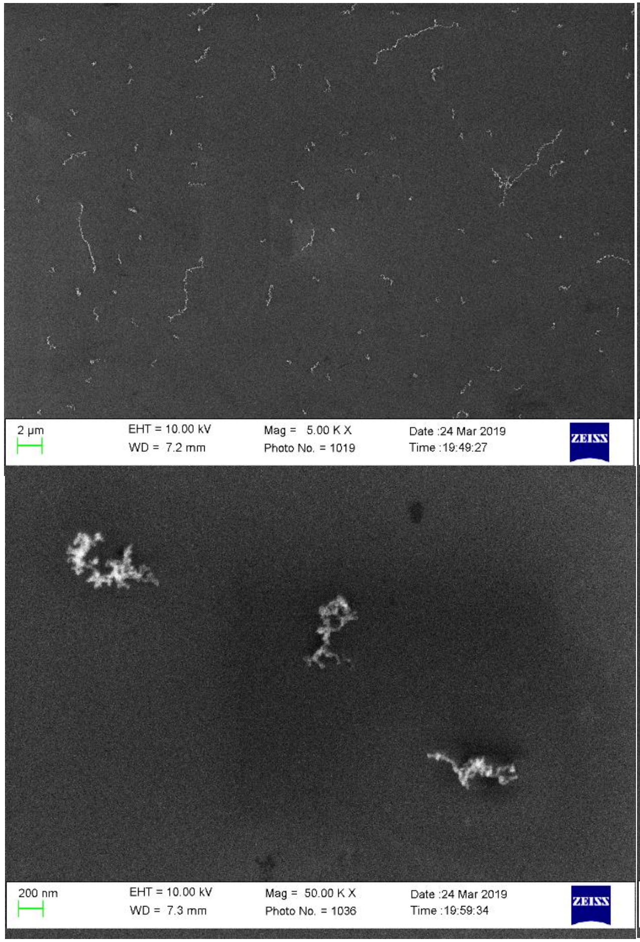


AAC 90 nm & DMA 300 nm

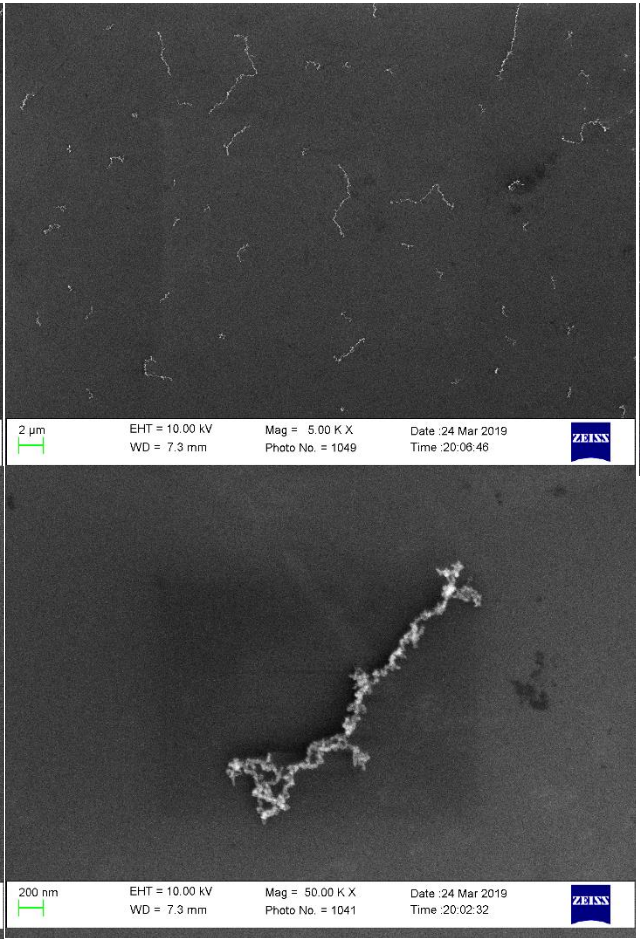


Selecting Particle "Morphology" – Same d_a

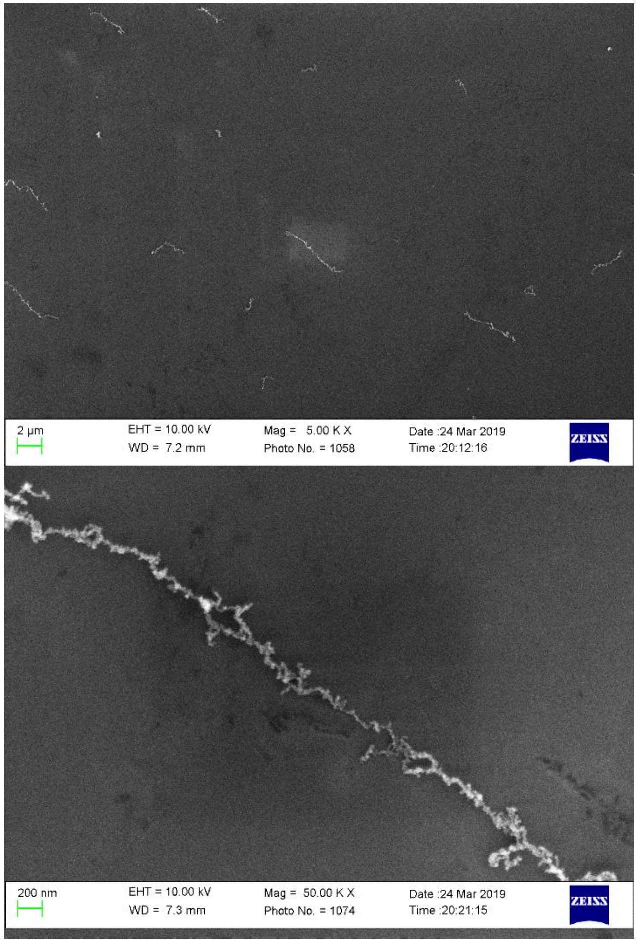
AAC 90 nm & DMA 300 nm



AAC 90 nm & DMA 360 nm

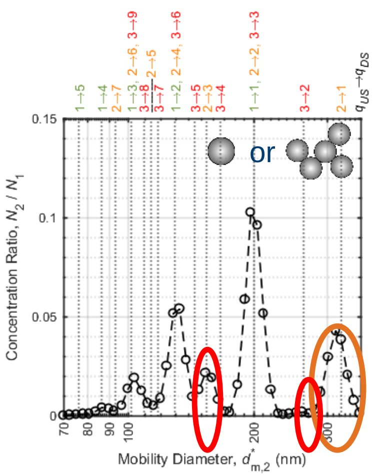


AAC 90 nm & DMA 430 nm

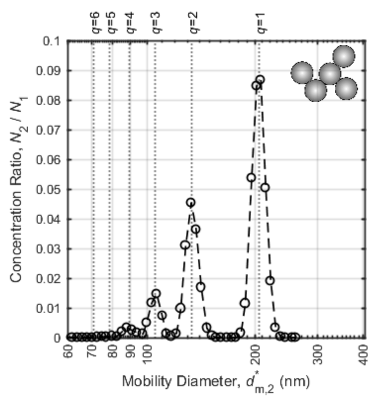


Summary

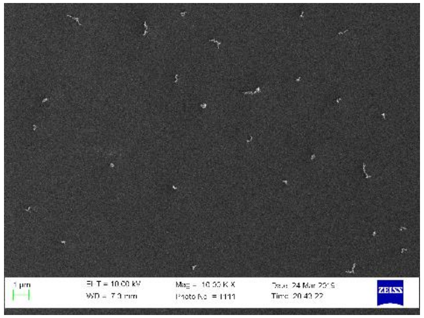
- This study demonstrates that a AAC-DMA system (i.e. two classifiers of different measurands) generates a non-spherical particle source that is monodisperse in d_a, d_m and m, and therefore morphology.
- The tandem setup overcomes many of the challenges when operating any of the aerosol classifiers as standalone instruments.
- Using an AAC in the tandem system is preferred to limit multiple-charging effects in the DMA or CPMA.
- This methodology was verified by:
    - Comparing DMA scans of the classified particles to identify additional peaks due to classifying multiply-charged particles and/or peak broadening from classifying different particle masses; and
    - SEM images of the tandem classified particles.



Standalone DMA



Tandem AAC-DMA



SEM Verification

Questions?

Jon Symonds: jps@cambustion.com

Tyler Johnson: tjj31@cam.ac.uk

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