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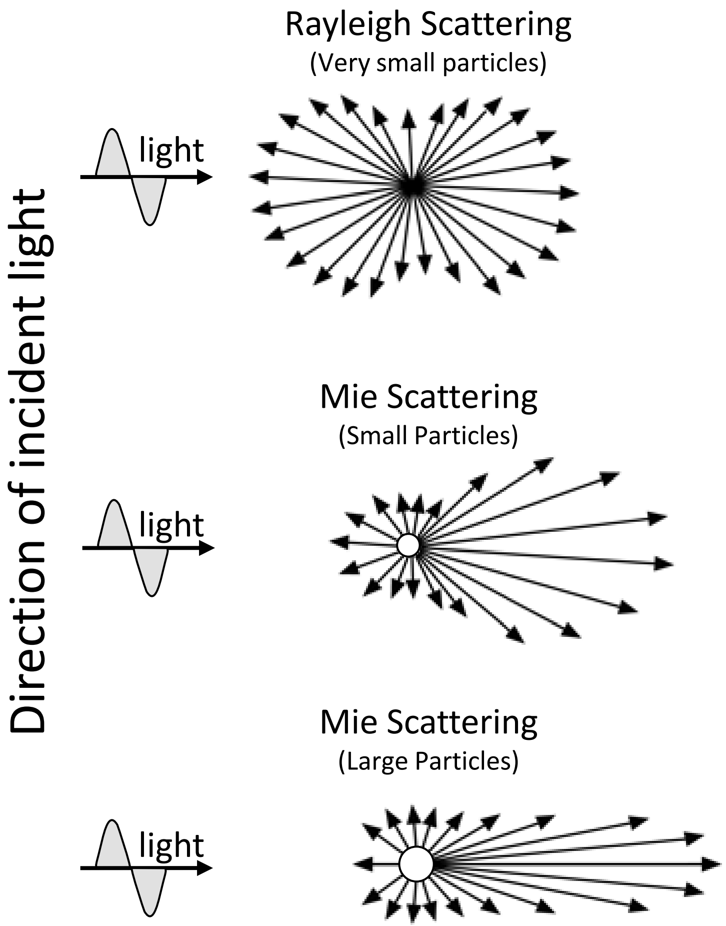
Using particles of truly monodisperse size distributions to improve the accuracy of in situ aerosol absorption measurements

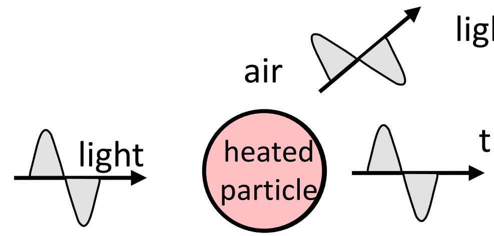
Cambridge Particle Meeting 2018

Aerosol Optical Properties

Scattering



Absorption

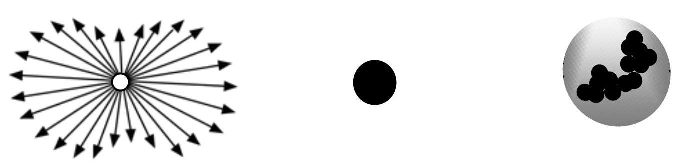


Single Scattering Albedo, *SSA*

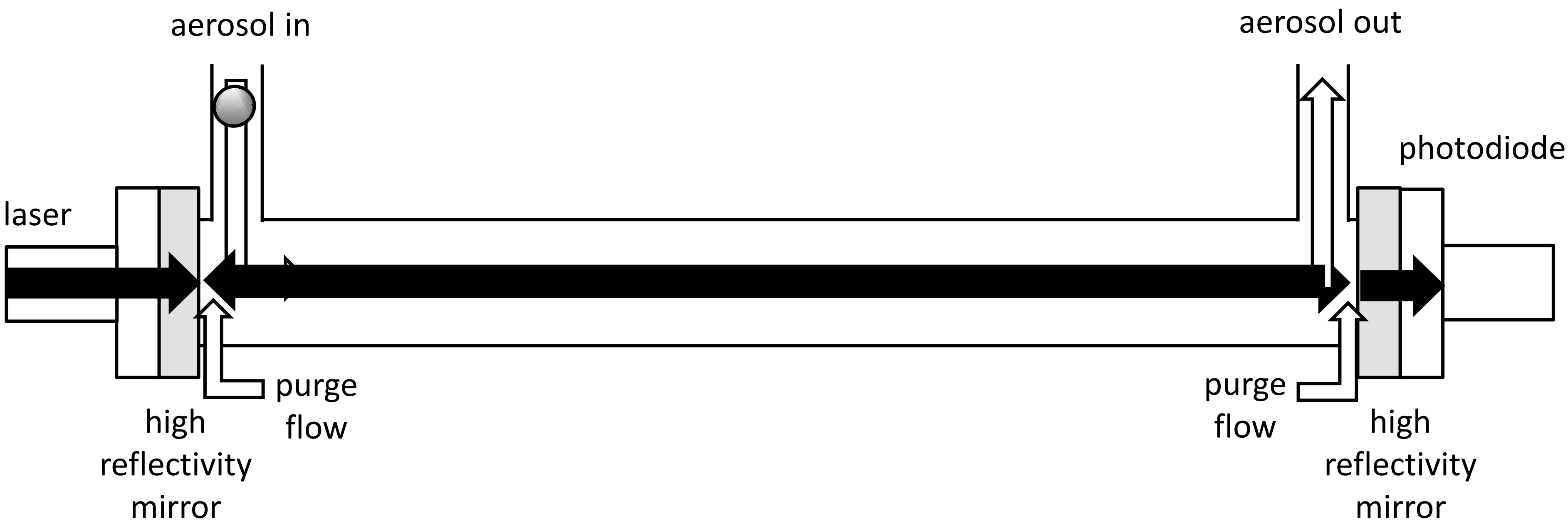
$$SSA = \frac{\textcolor{blue}{scattering}}{\textcolor{blue}{scattering} + \textcolor{red}{absorption}}$$

$$= \frac{\textcolor{blue}{scattering}}{extinction}$$

$SSA = 1?$ $\quad SSA = 0?$ $\quad SSA = ??$



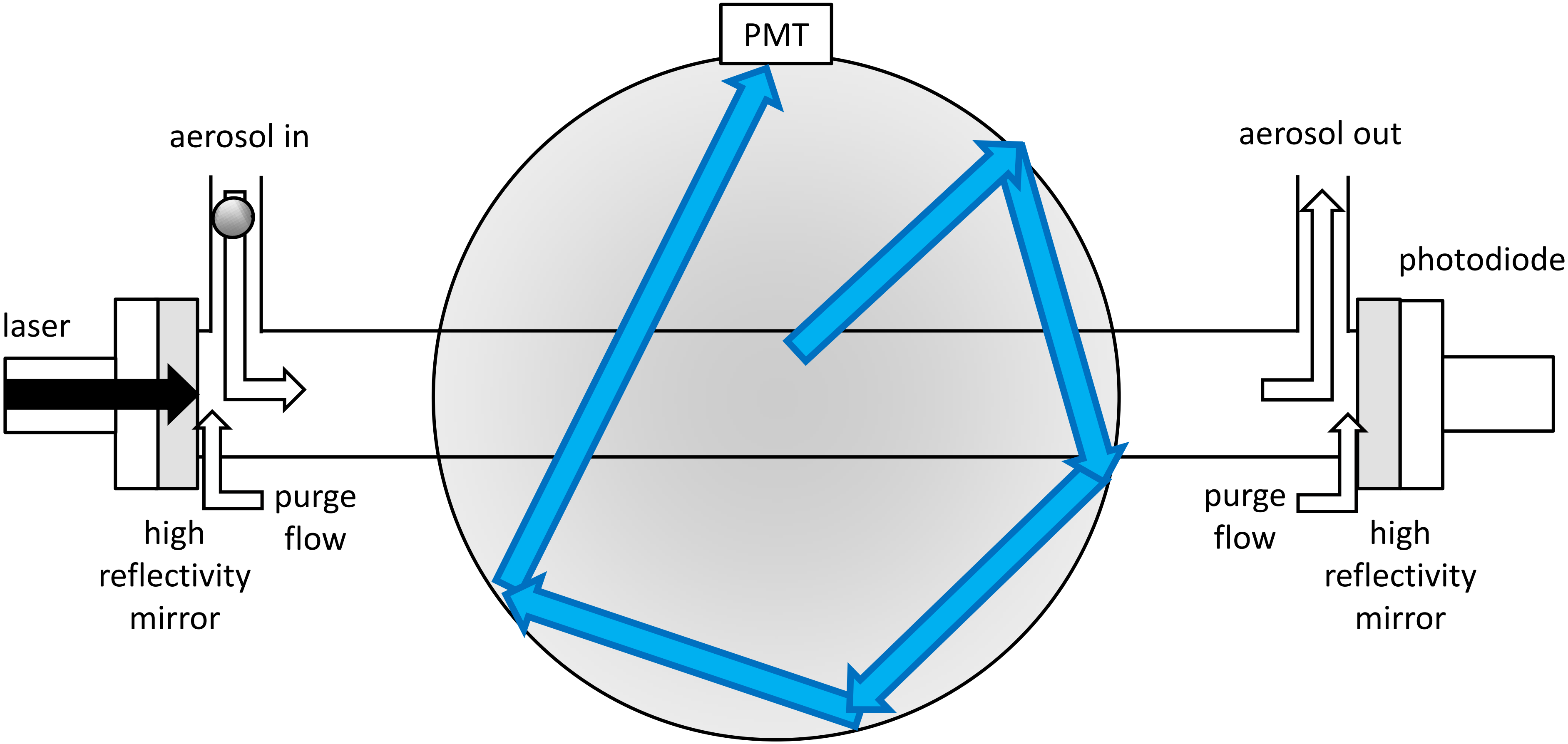
CAPS PM$_{SSA}$ - EXTINCTION



Extinction measurement:

- Modulated light source; effective path length several km
- Phase shift occurs due to presence of aerosol
- Geometric correction factor of 1.27 to account for mirror purge flow
- ~1s time response

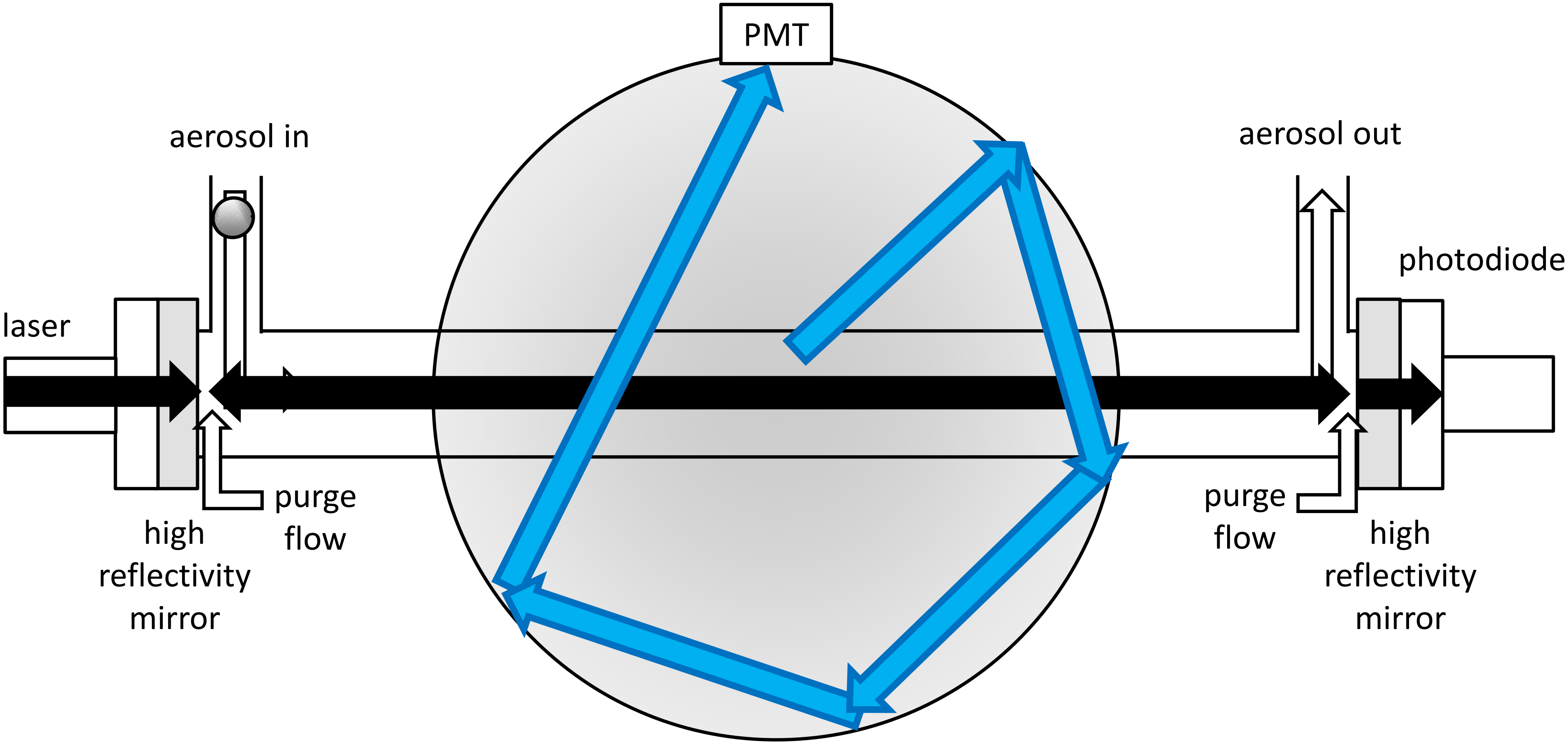
CAPS PM_{SSA} - SCATTERING



Scattering measurement:

- Integrating sphere captures scattered light
- “Truncation” of scattering signal due to entrance and exit of within the sphere
- Combine with the scattering measurement to obtain *Single Scattering Albedo, SSA*

CAPS PM_{SSA} – Single Scattering Albedo



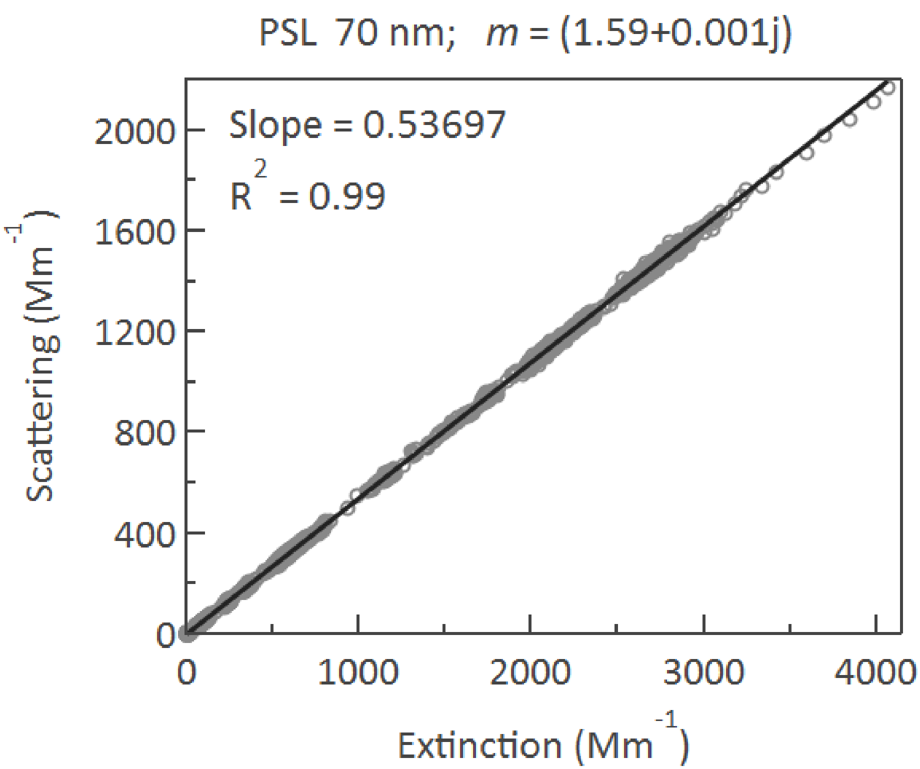
$$SSA = \frac{scattering}{extinction}$$

CAPS PM_{SSA} cross-calibration

AIM:

Using a *monodisperse* scattering aerosol, cross-calibrate the scattering and extinction channels

$$SSA = \frac{scattering}{scattering + absorption} = \frac{1}{1+0}$$



→ Monodisperse and in the Rayleigh regime (i.e. D << λ)

→ Very high SSA (i.e. white scattering aerosol such as PSL or $(NH_4)_2SO_4$)

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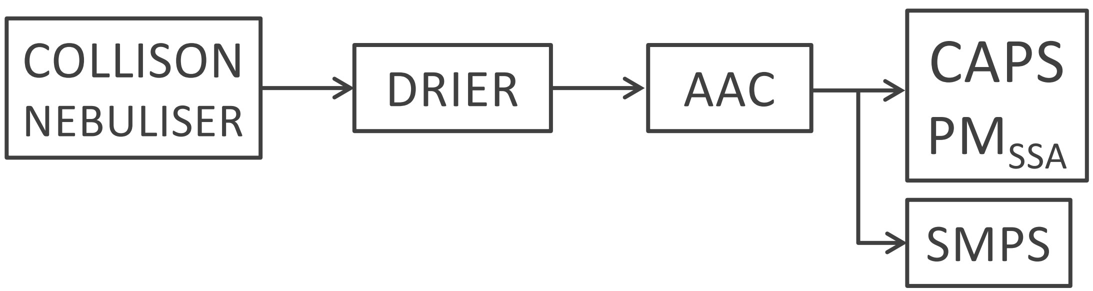
Experimental setup

SMPS suffers from:

→ multiple charging interference

→ loss of concentration due to charge distribution



Aerodynamic Aerosol Classifier (AAC):

- Truly monodisperse aerosol selection
- High throughput
- Wide size range 25nm - > 5µm

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Truly monodisperse aerosol selection

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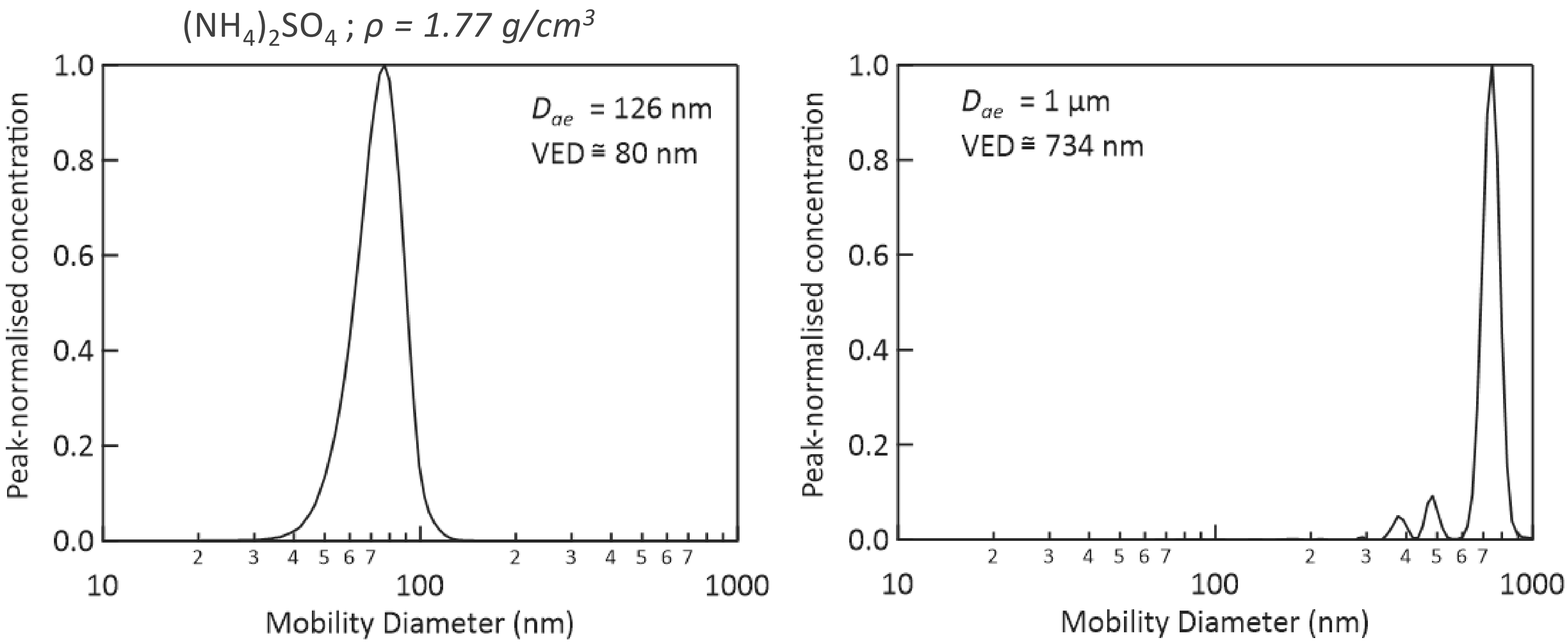
Aerodynamic Aerosol Classifier (AAC)
➔ truly monodisperse



- Optical instruments are invariant to particle charge state
- The high transmission efficiency of the AAC results in good signal:noise
- No interference due to multiply charged particles (including PSL doublets)

CAPS PM_{SSA} – deriving absorption

$$SSA = \frac{scattering}{extinction} = \frac{scattering}{scattering + absorption}$$

CAPS PM_{SSA}

Simultaneous extinction and scattering measurements with the same instrument reduce uncertainties in derived absorption

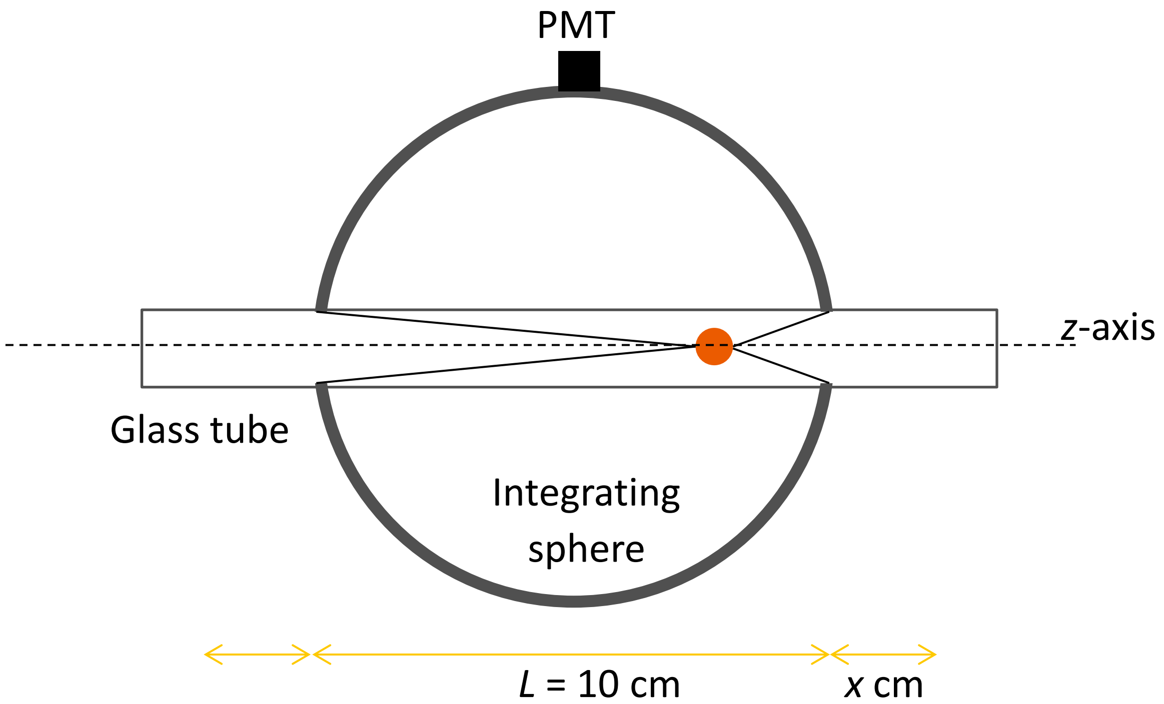
$$absorption = extinction - scattering$$

However, uncertainties in both scattering and extinction measurements have large effects on absorption uncertainty

- accurate quantification of the “truncation effects” are required…

e.g. ***5%*** *error in truncation correction factors lead to absorption errors of ~****5-20%*** *at medium to high SSA (0.5-0.8) and* ***over 50%*** *at very high SSA (0.95)*

CAPS PM$_{SSA}$ truncation issue



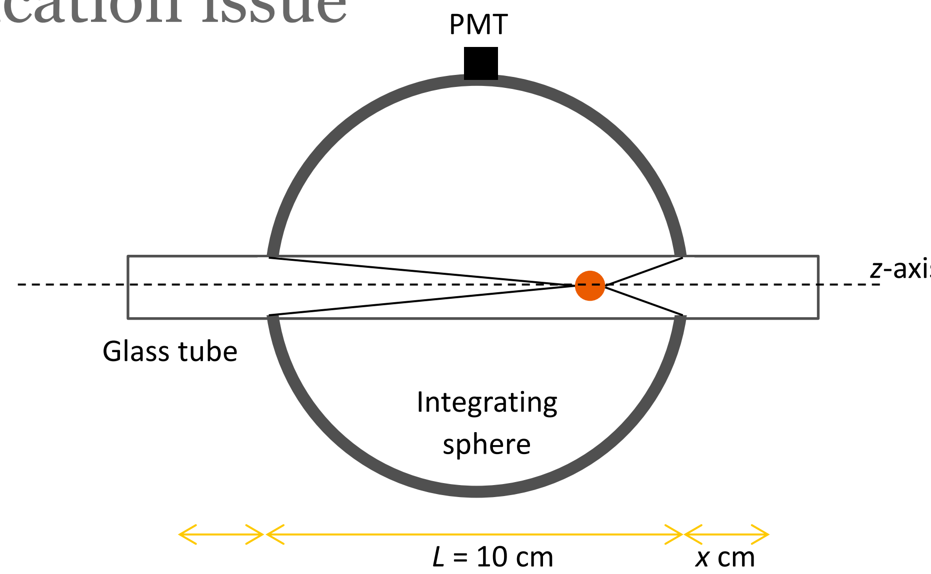
Scattering signal:

- Measured by PMT
- Certain portion of scattered light escapes the integrating sphere from the two open ends
- Light is attenuated and reflected by walls of the glass tube
- Scattering geometry and thus truncation dependent on aerosol size

CAPS PM_{SSA} truncation issue

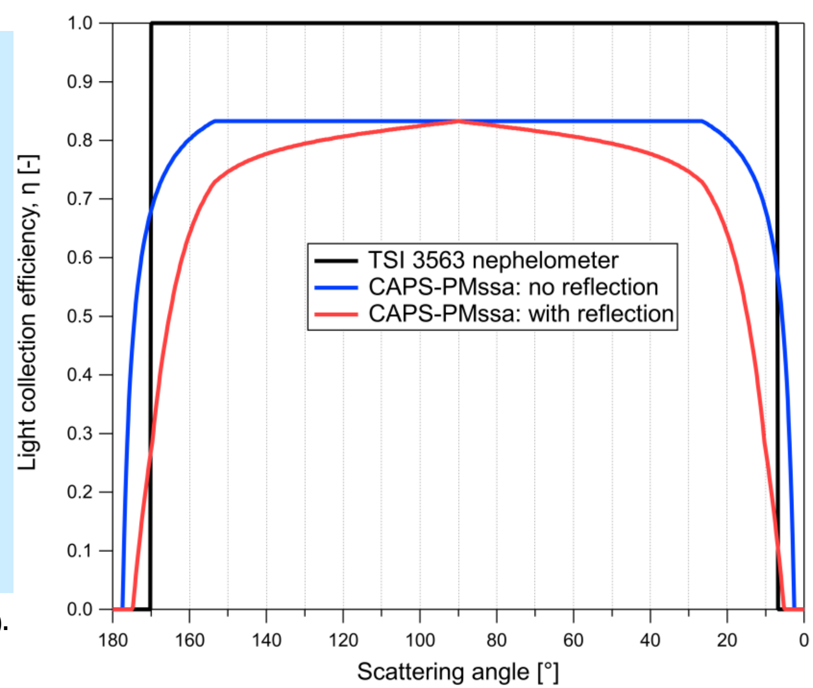
Onasch et al., (AS&T 2015) model:

- Particles distributed homogeneously along the *z*-axis over the length *L*+2*x*
- *x* was set to 1 cm to obtain best match between modeled and measured truncation for a set PSL experiments at *λ* = 450, 630 nm
- Reflection from the glass tube not accounted for



Our model builds on the Onasch model plus:

- Reformulates the problem through the light collection efficiency of the integrating sphere, *η*, a function of scattering angle and *z*-position
- *η* varies between 0 (no light collected) and 1 (all light collected)
- Allows simple introduction of the effect of reflection from the glass tube, which is calculated with Fresnel equations

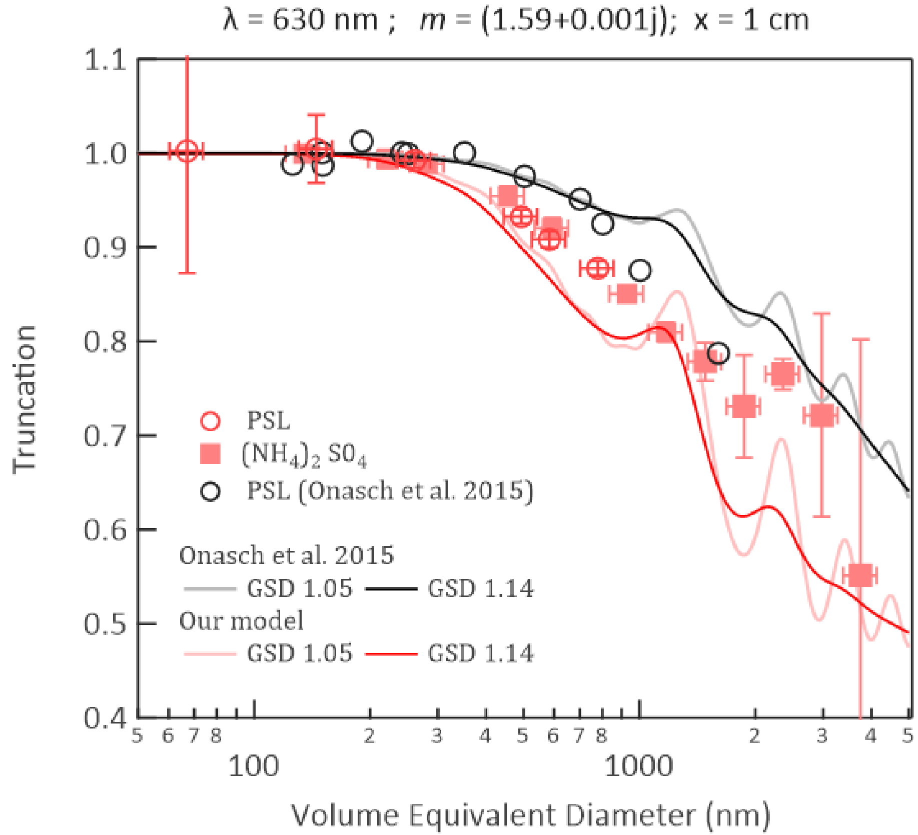


Onasch, T. B., Massoli, P., Kebabian, P. L., Hills, F. B., Bacon, F. W., & Freedman, A. (2015). Single scattering albedo monitor for airborne particulates. *Aerosol Science and Technology*, *49*(4), 267–279. https://doi.org/10.1080/02786826.2015.1022248

CAPS PM_{SSA} truncation experiments

Particles size selected by Aerodynamic Aerosol Classifier
➔ truly monodisperse

Measured truncation defined as:
Ratio of measured scattering to extinction coefficients
(always normalized to truncation of Rayleigh scattering)



- Model accounting for glass tube reflection predicts greater truncation than measured for PSL spheres and ammonium sulphate
- Model neglecting glass tube reflection predicts less truncation than measured… geometry?

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Further work

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- More measurements to constrain the models
  - Measurements with different scatterers **and absorbers**
    - *Nigrosin, fullerene soot, aquadag… a black carbon standard?*
- Investigate CAPS PM_{SSA} wavelength dependencies
  - 450 nm
  - 630 nm
  - 780 nm
- Towards accurate in situ absorption measurements
  - Improve CAPS PM_{SSA} measurement
    - Monitor and control *RH, T, Q*
    - Improved scattering and extinction measurements
    - Improved models
  - Comparison between filter-based (e.g. MAAP) and photoacoustic (e.g. PAX) methods

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