Characterization of dimers of soot and non-soot particles formed by charged coagulation

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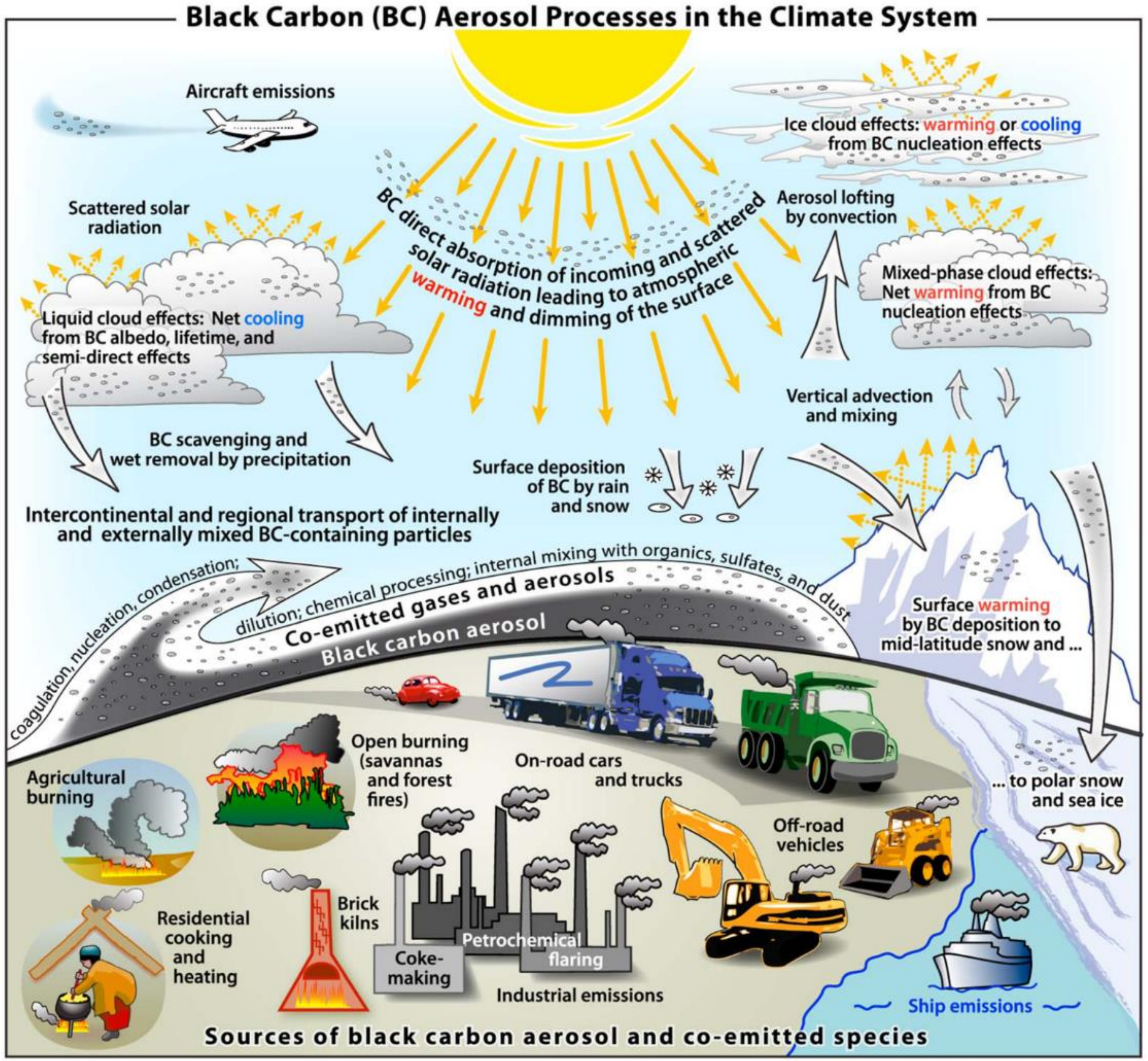


Bond et al., 2013

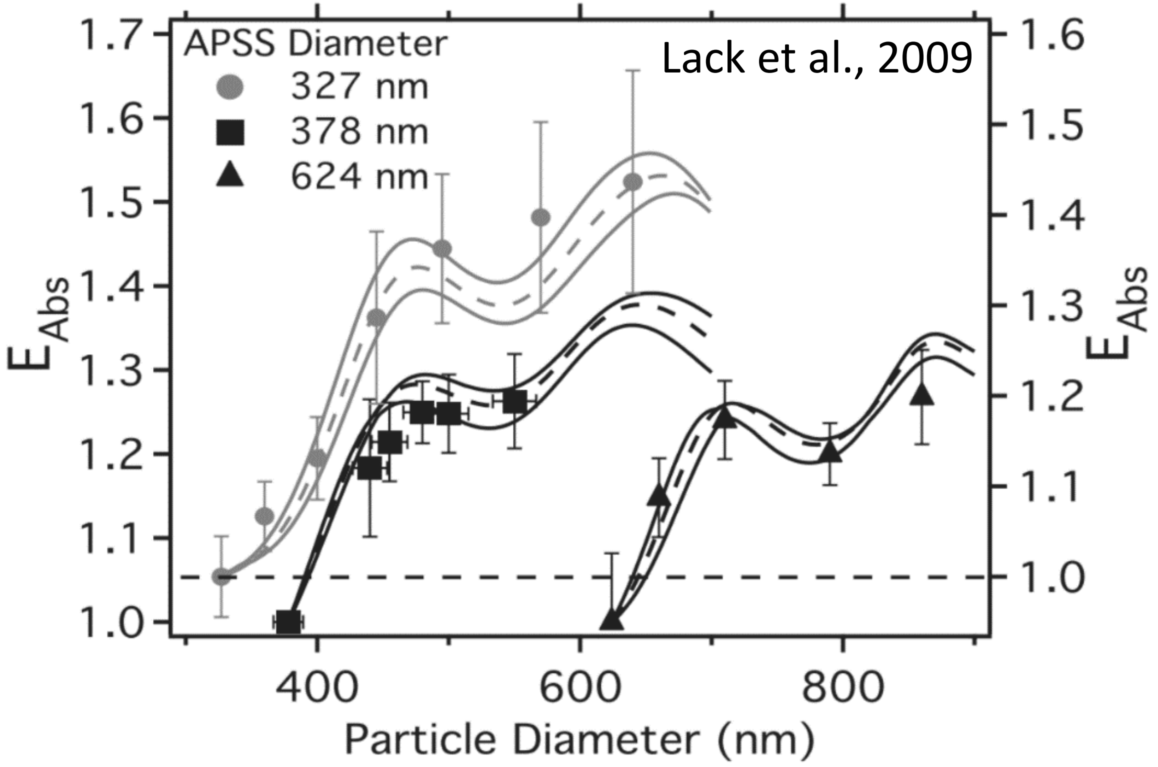
Here Soot = Black Carbon (BC)

Focus of experiments in our lab

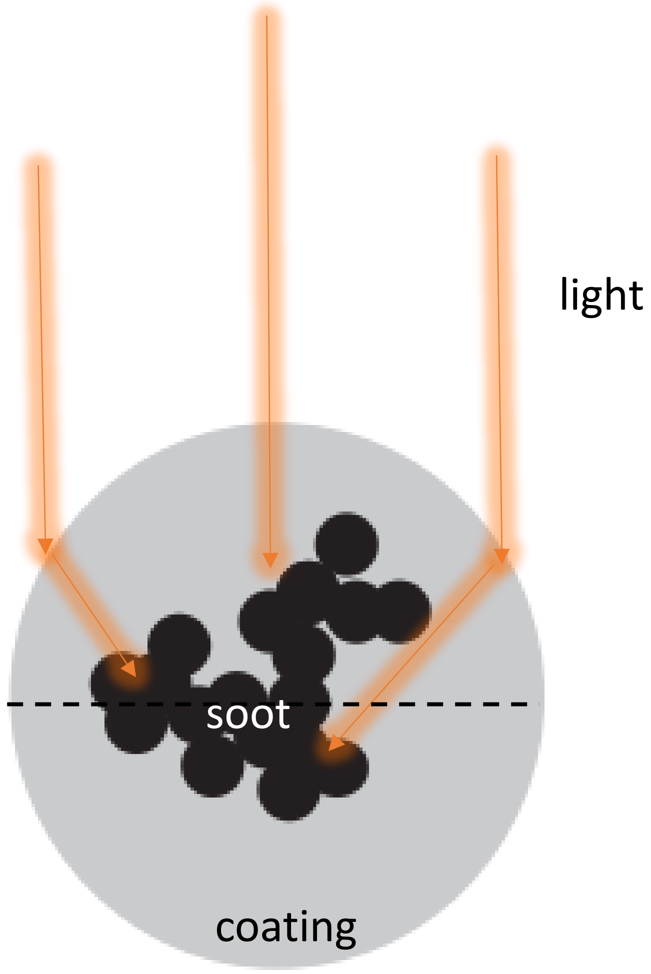
1. Ice nucleation on BC particles under cirrus cloud conditions
2. Optical properties (i.e., absorption) of BC containing particles

- *Internal and external mixtures of BC containing particles*

Absorption enhancement (lensing)



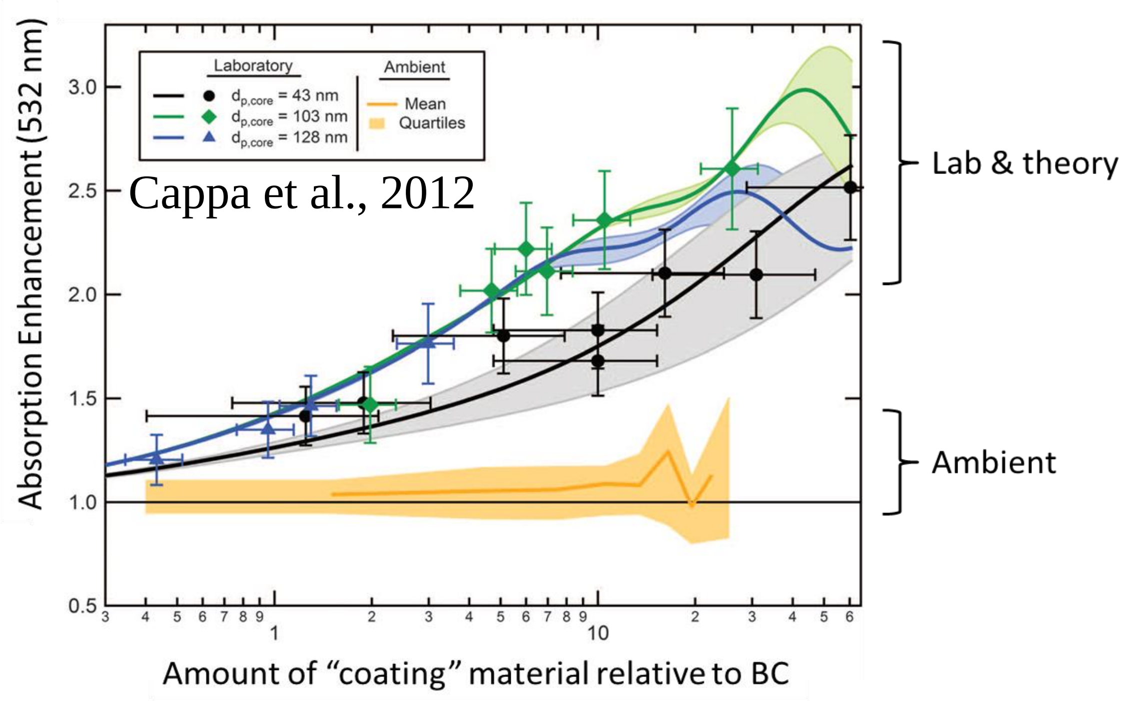
Dioctyl sebecate (DOS) coated absorbing PSL particles well characterized by core-shell Mie theory



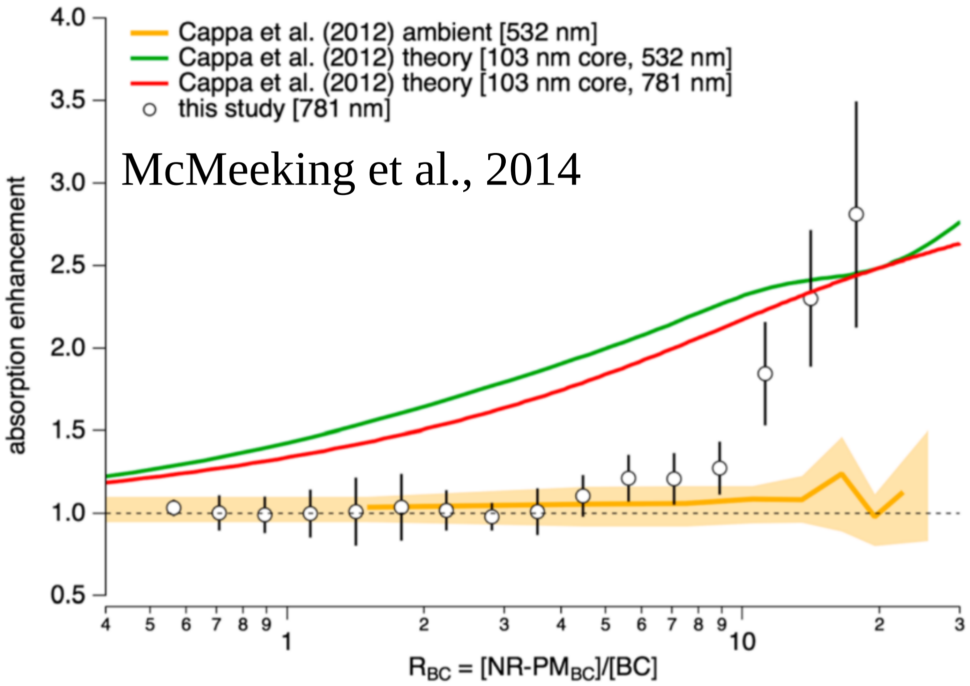
Non-soot material on soot particles increase optical cross sections

Discrepancy between ambient/source and core-shell results

Ambient vs Laboratory

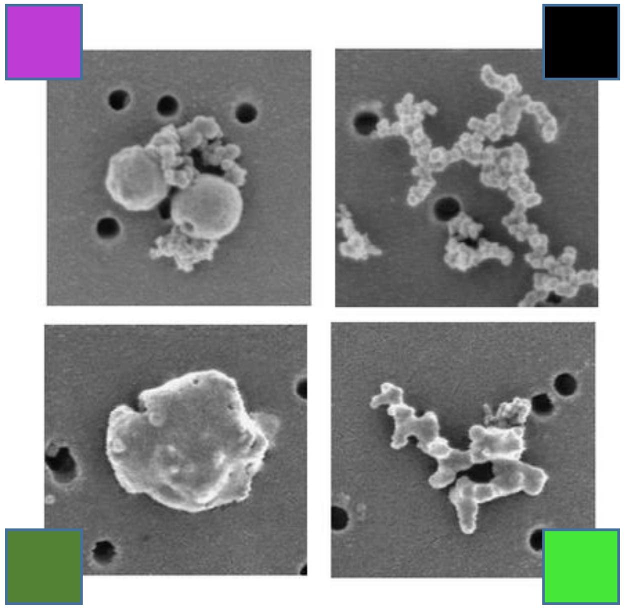


Biomass burning source

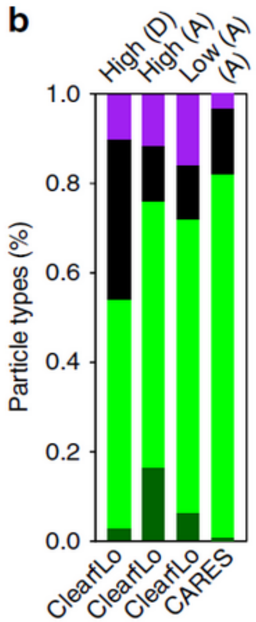


- Gap between well-characterized laboratory and field (ambient and biomass burning) measurements, either due to particle variability or population mixing

Particle morphology versus population mixing

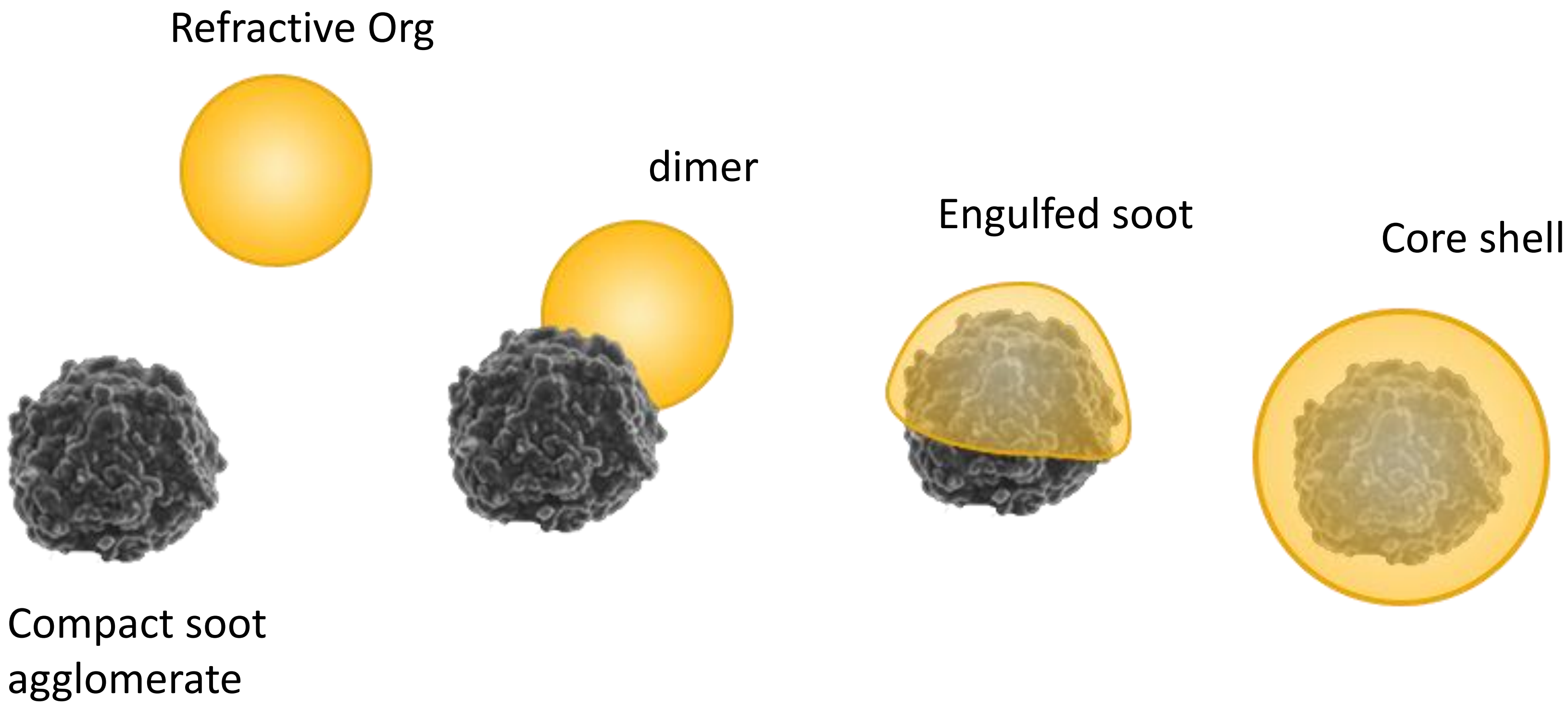


Liu et al., 2015 - Mich. Tech. Univ.



- Core-Shell structure has an increased optical cross section which enhances the absorption of light
- Core-shell is the most common representation in models
- Other types of internal and external mixtures are prevalent in the atmosphere
- What are the relative effects of morphology versus population mixing?

Objective – study coagulated BC particle types



- Bare soot, thinly coated and thickly coated were extensively studied in previous campaigns
- Data is scarce for the dimer structure of coagulated particles (complex experiments)

Characteristic times of bipolar coagulation

Total concentration	Tau
1e4	52h
1e5	5.5h
1e6	30min
1e7	3min

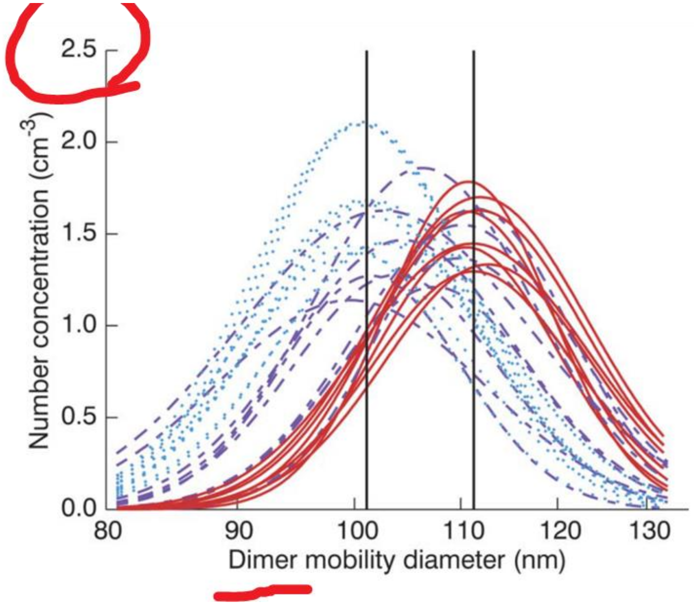
Kim et al. 2005

Known challenges…

1. Coagulation – slow process

2. Low number concentrations of dimers generated, near detection limit of optical measurements

For mono $>1*10^5$ cm^{-3}

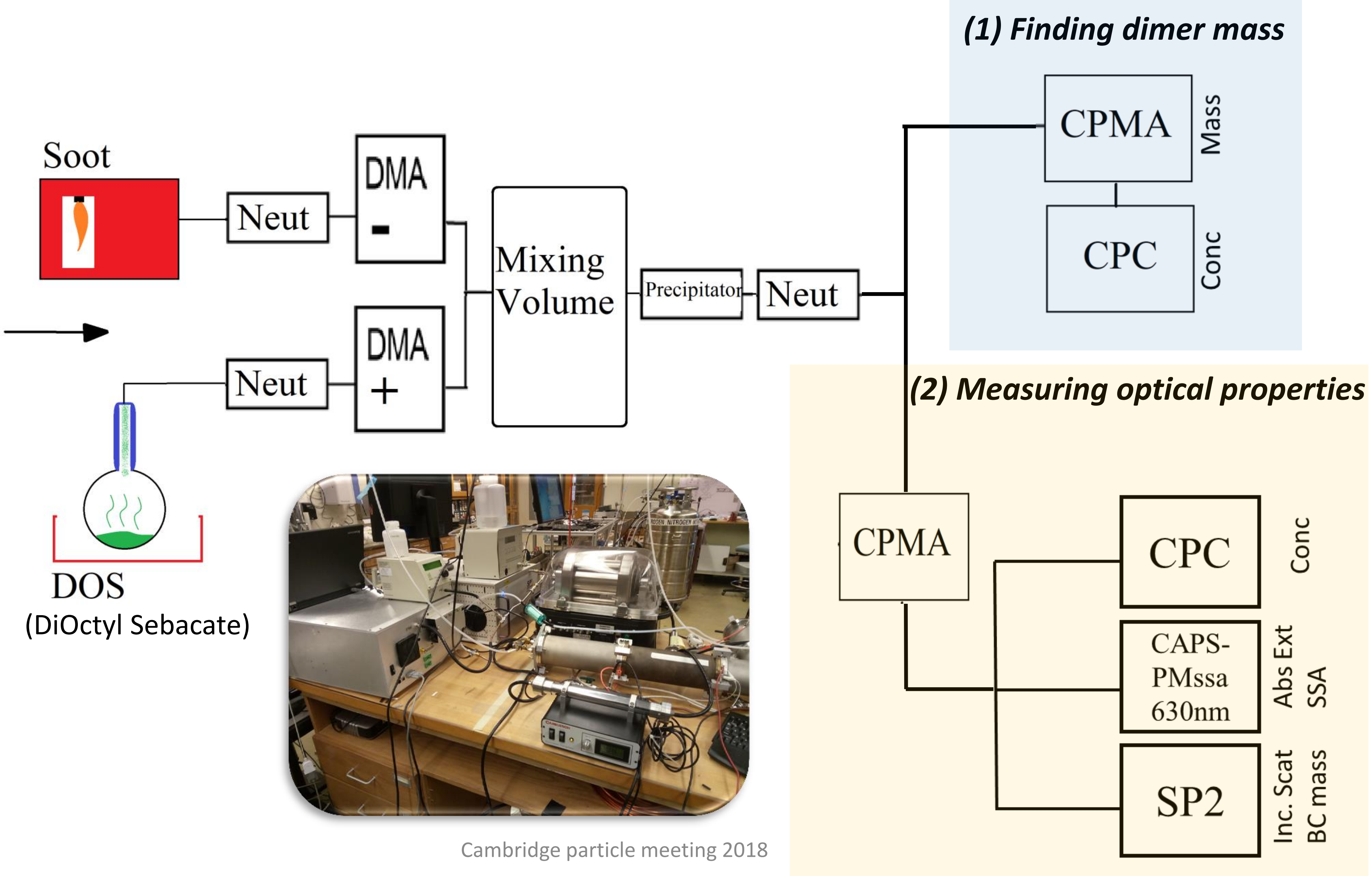


Petters & Rothfuss (2016)

Charge-enhanced coagulation
Process steps

1. Generation of two **polydispersed** particle distributions
2. **Size** select monodisperse particle distributions **with opposite charges**
3. Neutralization of the charge by **Coagulation**
4. Removal of all remaining charged particles i.e. **separation of the dimer**
5. **Recharging** the neutral particles to detect the neutral dimer with a CPMA (Centrifugal Particle Mass Analyser, Cambustion Ltd.)
6. Measuring **optical properties** of dimer particles!

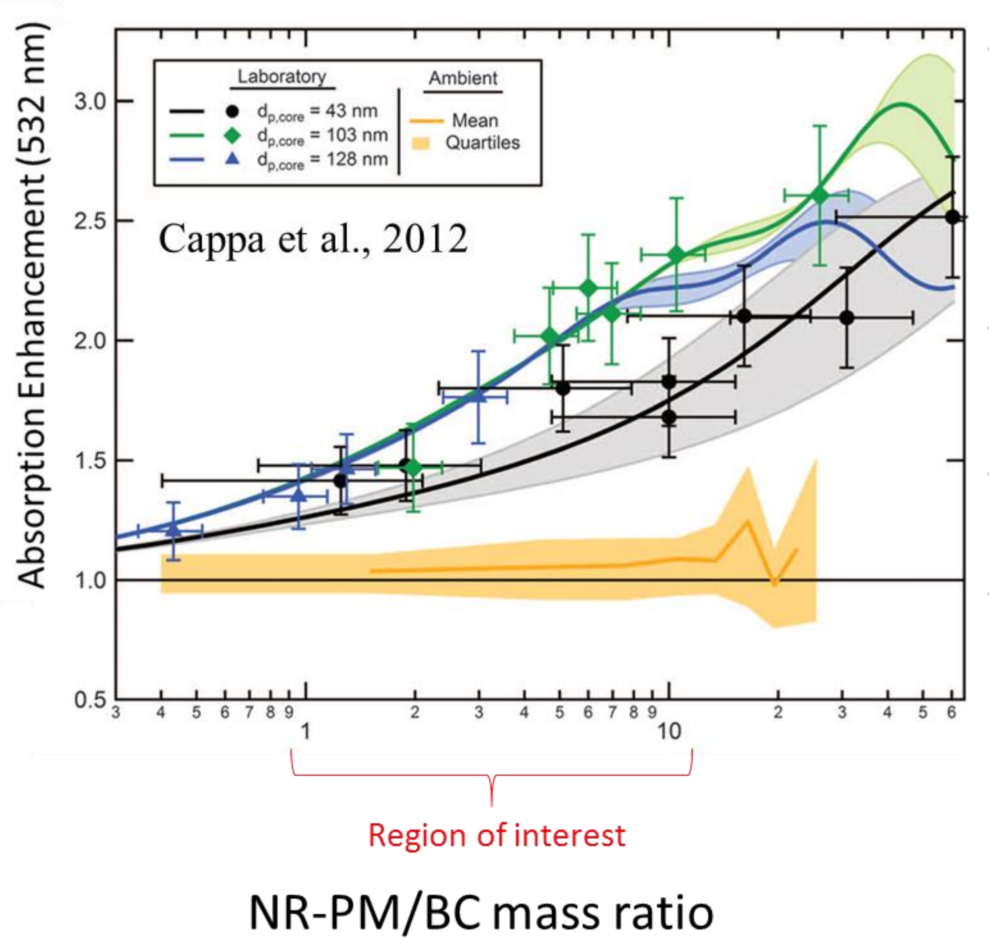
Experimental setup



Non-soot particle generation

Experimental plan

- DOS-DOS (liquid-liquid) experiments
  - Optimize experiments
  - Assess methodologies
- DOS-Soot experiments over "Region of Interest" in NR-PM/BC ratio and E_{abs} space
- Study more interesting mixtures, including ammonium sulfate-soot and secondary organic aerosol (SOA)-soot



Identification of mass-distribution peaks

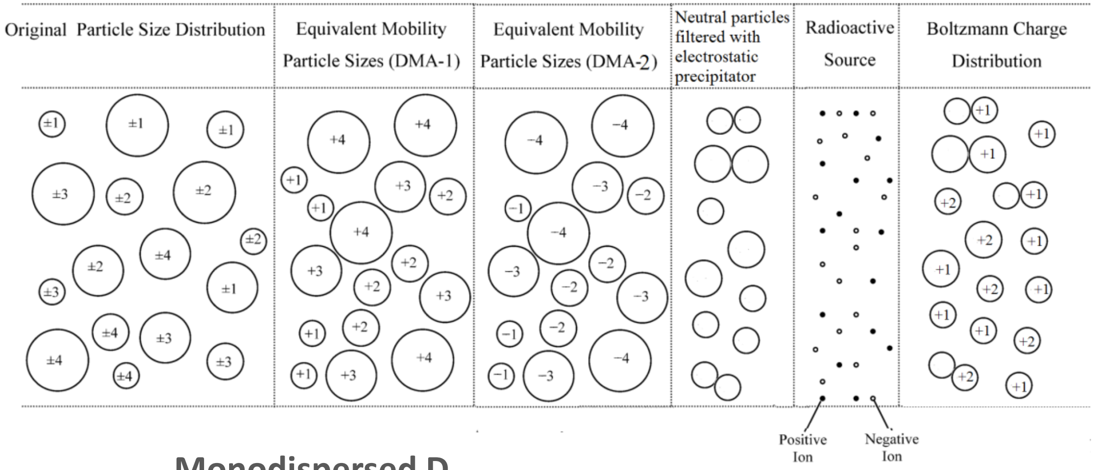
(re-neutralized positive monodispersed particles)

Mixing time

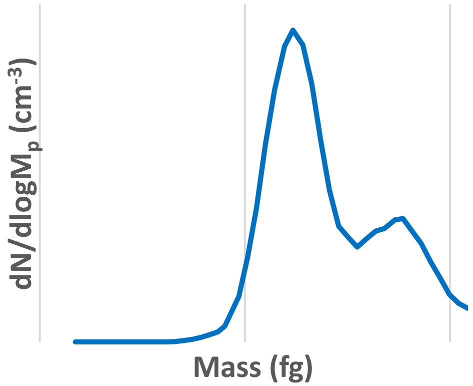
DOS-DOS	0 min	11 min	3.5 h
$\frac{C_{dimer}}{C_{monomer}}$	0.05 %	2.6 %	3 %

(for identical conditions)

	DOS-DOS	Soot-DOS	Soot-A.S
$\frac{C_{dimer}}{C_{monomer}}$	4 %	6 %	3.5%



Monodispersed D_m
(Mass scan)



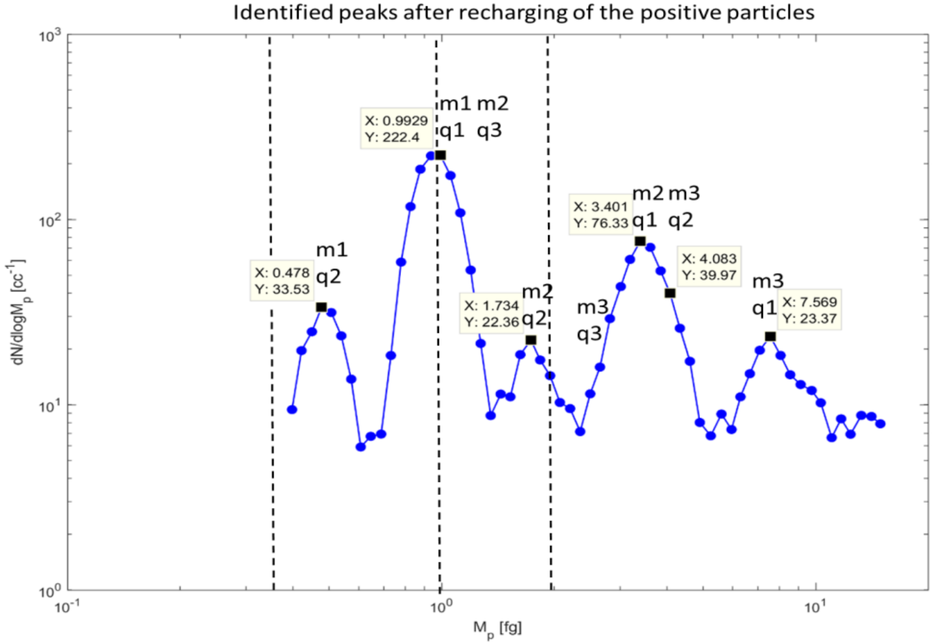
Recharging

Monodispersed mobility diameter (Mass scan)

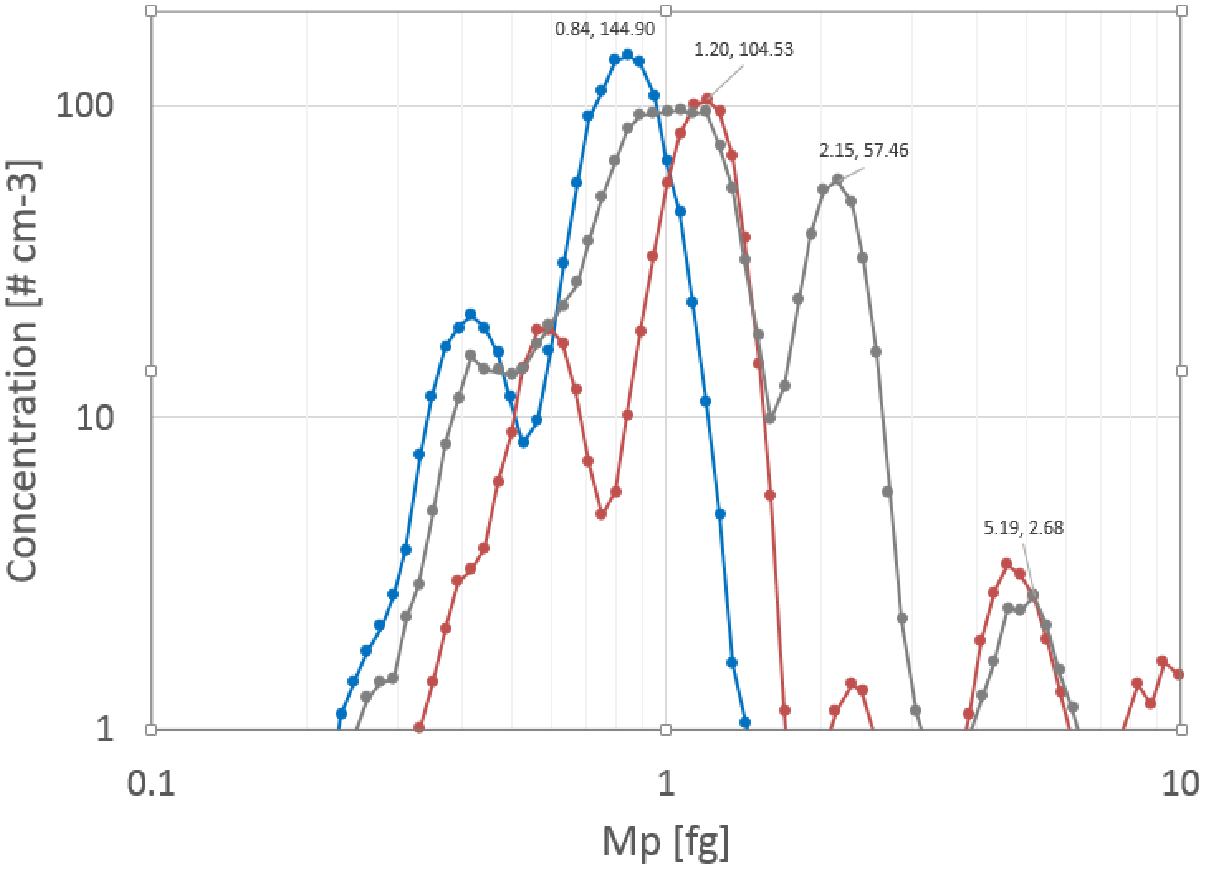
m1/(q=1)=1

m2/(q=2)=1.7

m3/(q=3)≈2.6



DOS-DOS liquid coalescence – optimized conditions

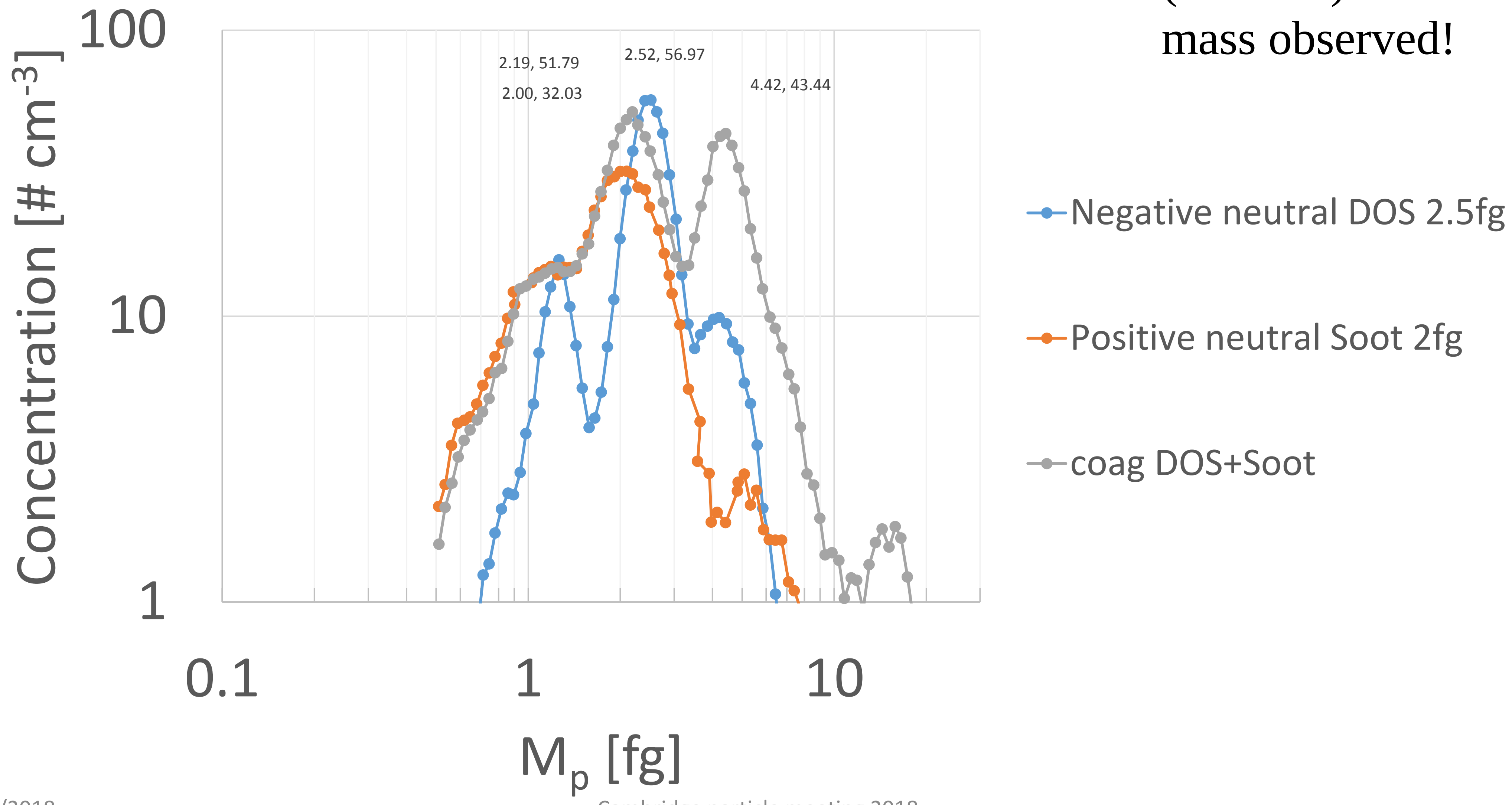


(-) particles
(+) particles
(0) neutral coagulated

- Clear confirmation of coalescence of two particles at higher mass (grey)

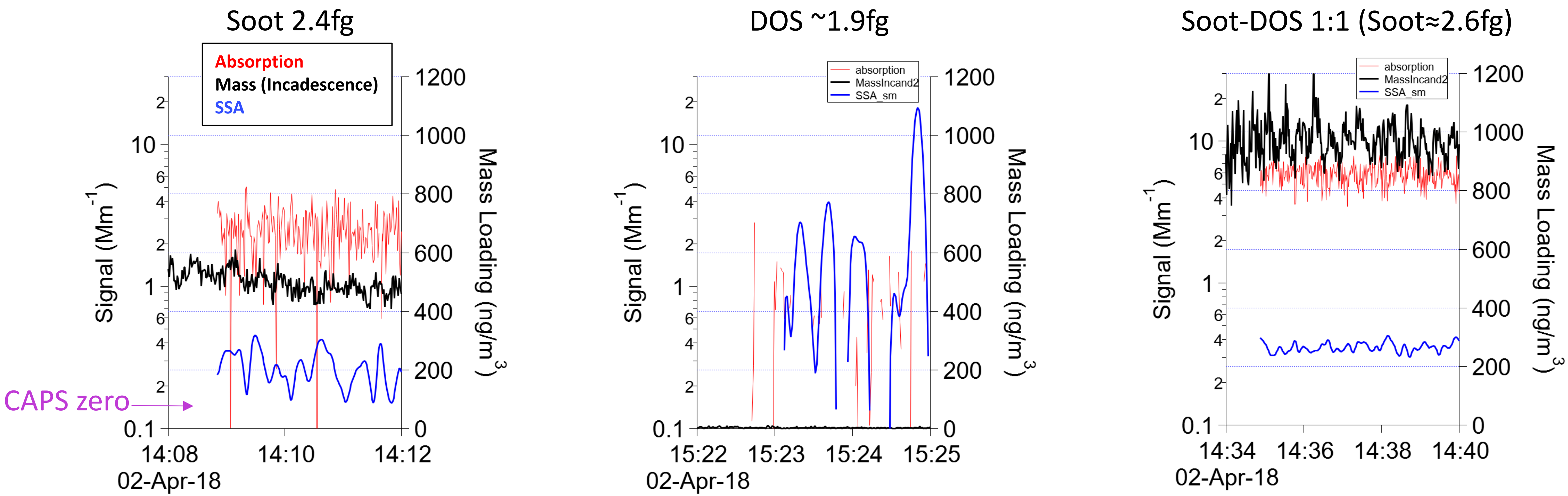
- Preliminary experiments show a clear peak for coalesced negatively charged 0.9 fg DOS with positively charged 1.2 fg DOS particles at a mass of 2.15 fg

Soot-DOS coagulation



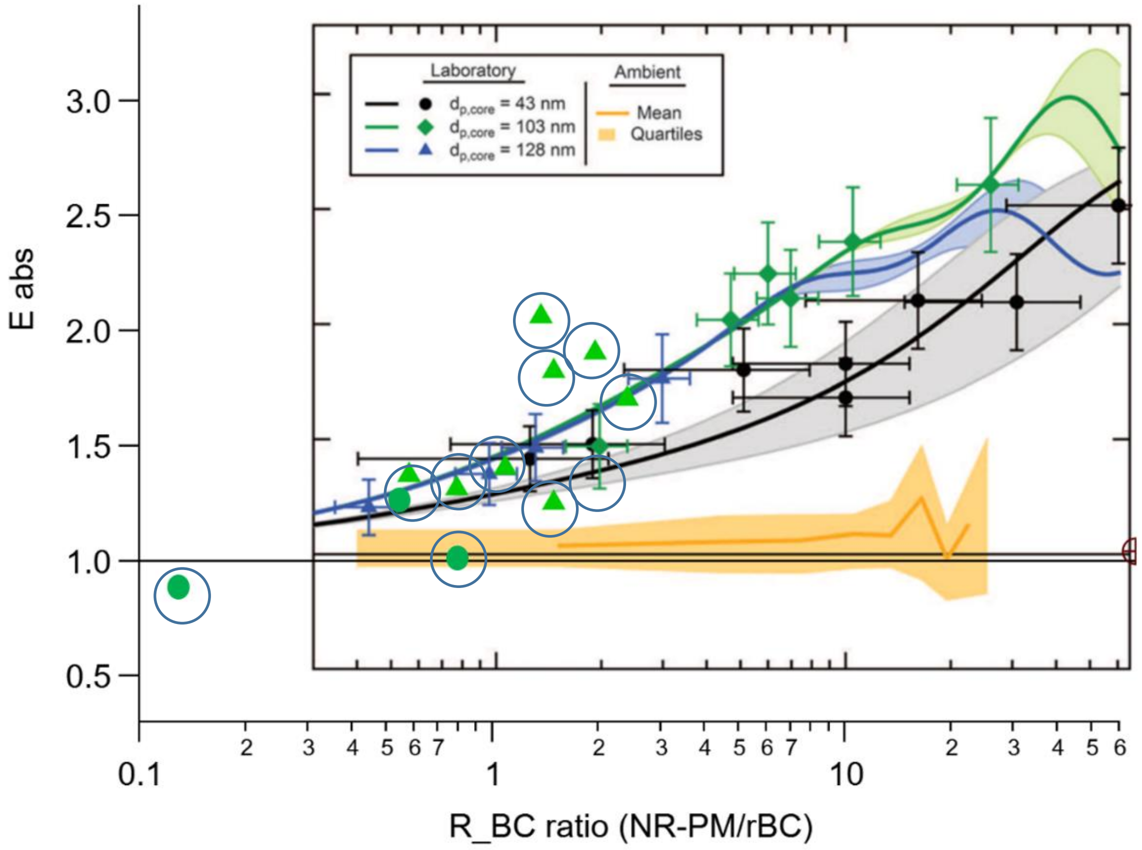
- Clear coagulated (doubled) neutral mass observed!

Optical detection (DOS-Soot) using CAPS PMssa

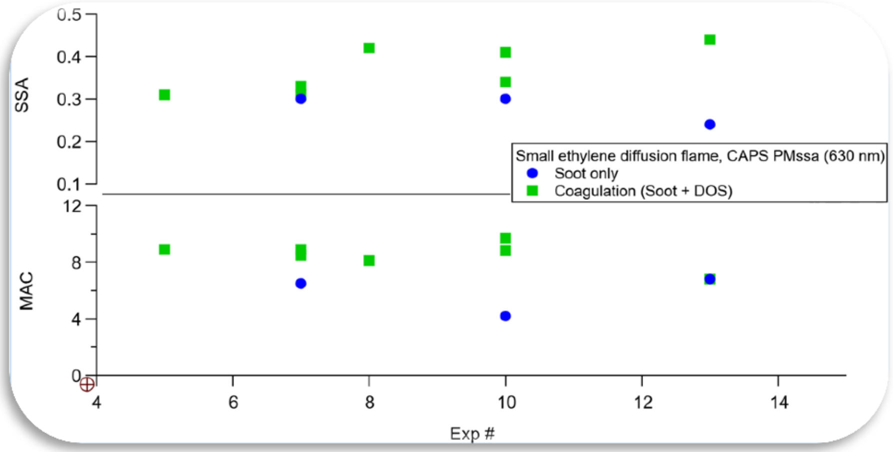


- During baseline measurements, CAPS signal is zero
- 20% Absorption enhancement (Eabs) was calculated for the dimer
- Single Scattering Albedo is higher than for pure soot and lower than for pure DOS

Preliminary results



- Most data follow Mie theory with some exceptions



- SSA of coagulated dimers are similar ot higher than pure soot particles, as expected
- MAC of coagulated dimers and pure soot particles are similar but in some cases are higher, which was unexpected
- Observation: DOS likely “wetting” soot particles on experimental time-frame

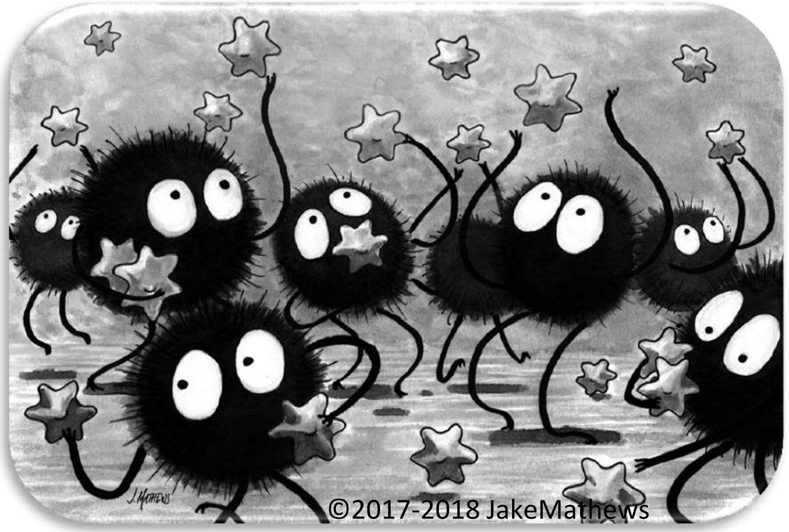
Summary

- Rarely studied coagulation of monomers was achieved and reproduced in a laboratory setup
- The process was optimized to allow shorter coagulation time
- Several types of monomers were coagulated (DOS-DOS, Soot-DOS, Soot-A.S, Soot-A.N, Soot-SOA)
- The process was optimized to allow optical detection (CAPS-PMssa, SP2)
- Preliminary results of Eabs, MAC and SSA for uniform distribution of Soot-DOS dimers are reported

Future work

- Higher Rbc ratios
- viscous aerosols (e.g. SOA) to reduce core shell structures
- Include coagulated dimers into population mixing studies
- Study humidity influence on coagulation efficiency

Questions?



Acknowledgements

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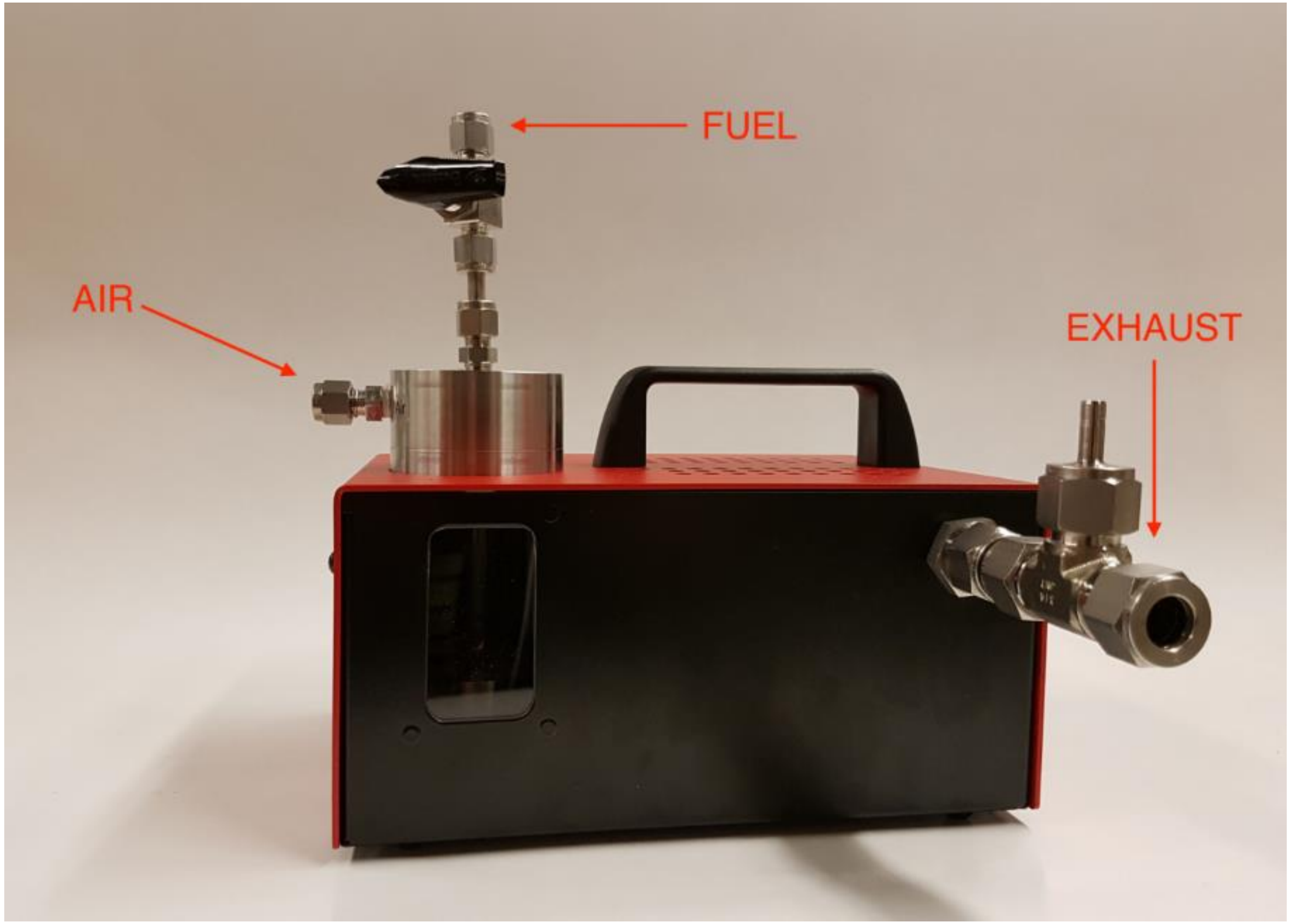
Extra Slides

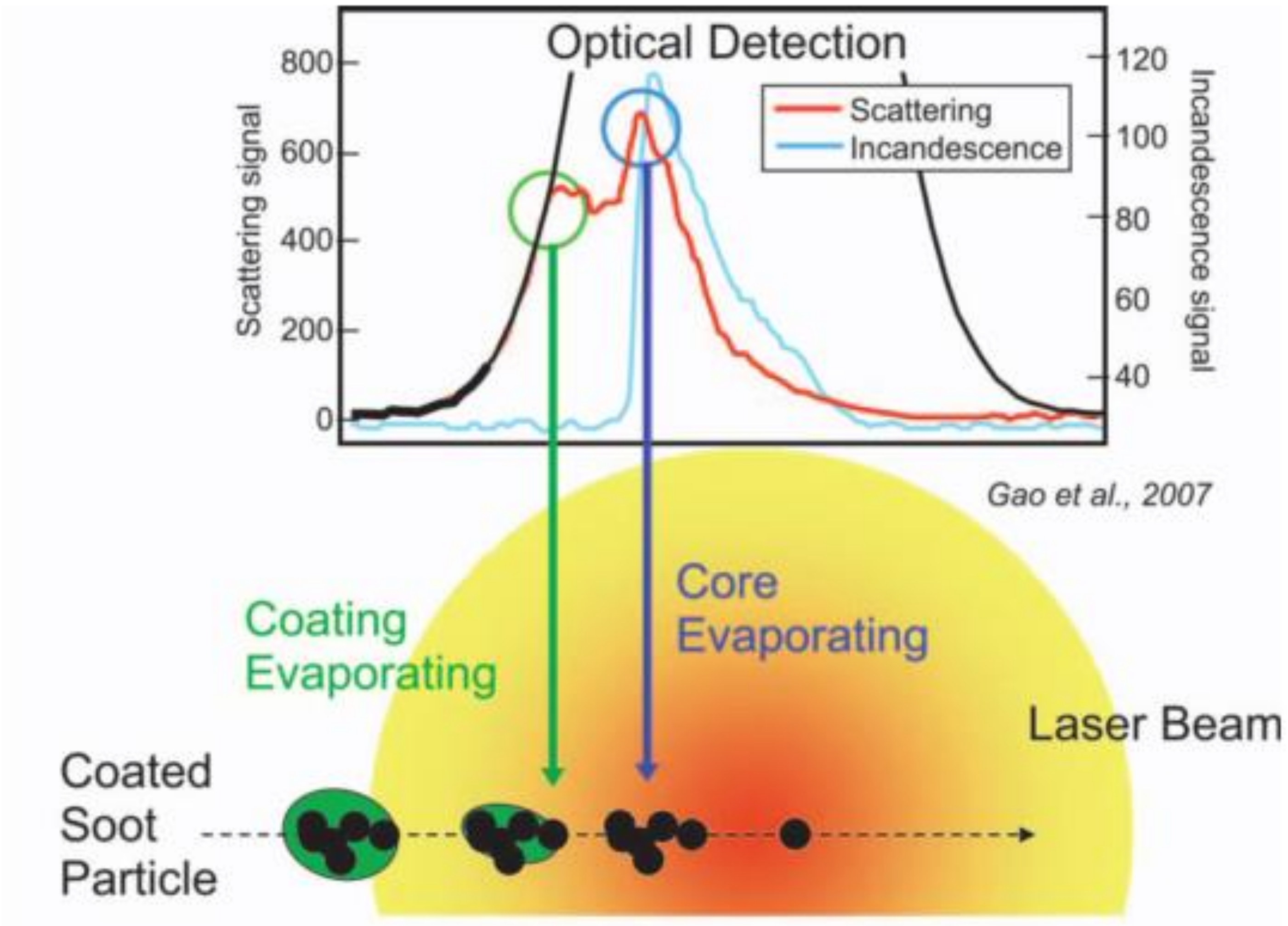
Inverted Burner Soot Generator

Argonaut Scientific Corporation
11119 – 50th Ave, Edmonton,
Alberta, Canada



Soot Photometer (SP2)

- Organics scatter light, black carbon incandesce
- Determination of soot core mass for coated particles by temporal separation between scattering and incandescence signals

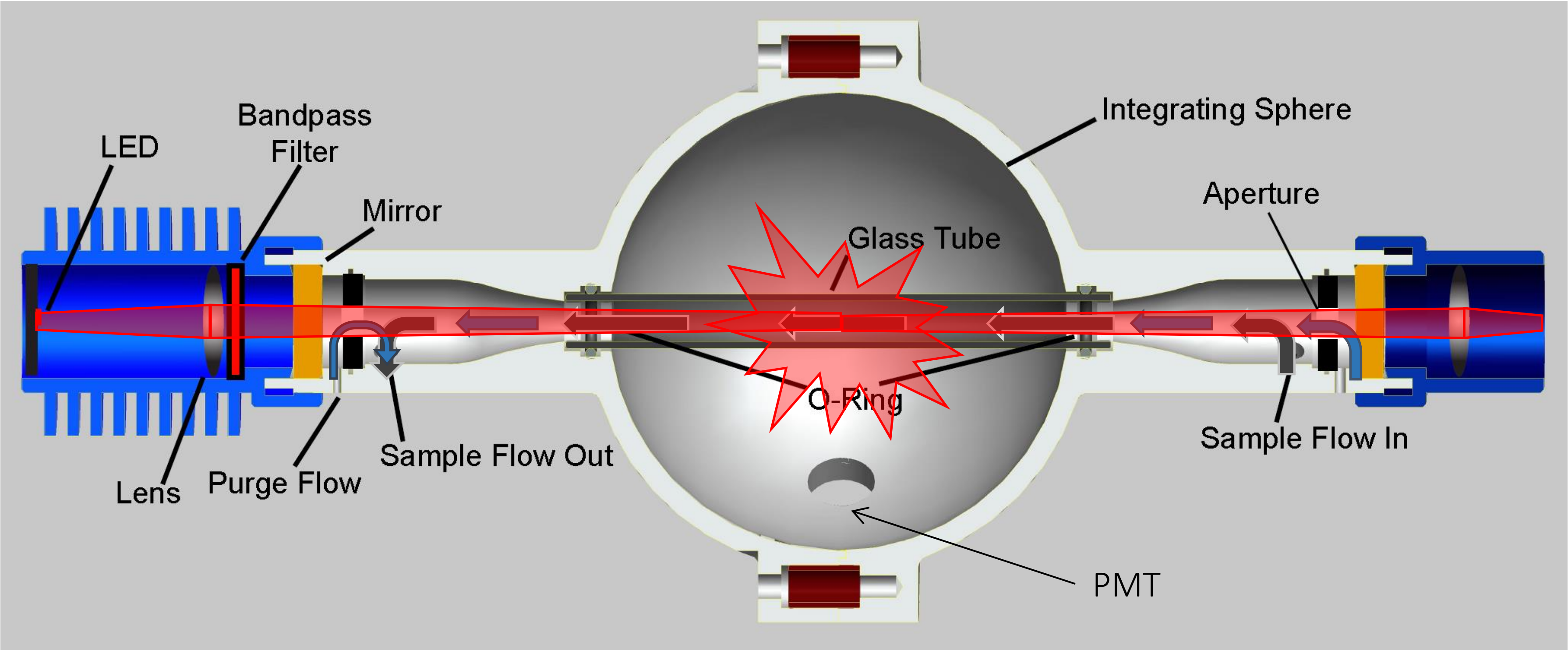


Onasch et al., 2012

CAPS PM$_{ssa}$ Monitor

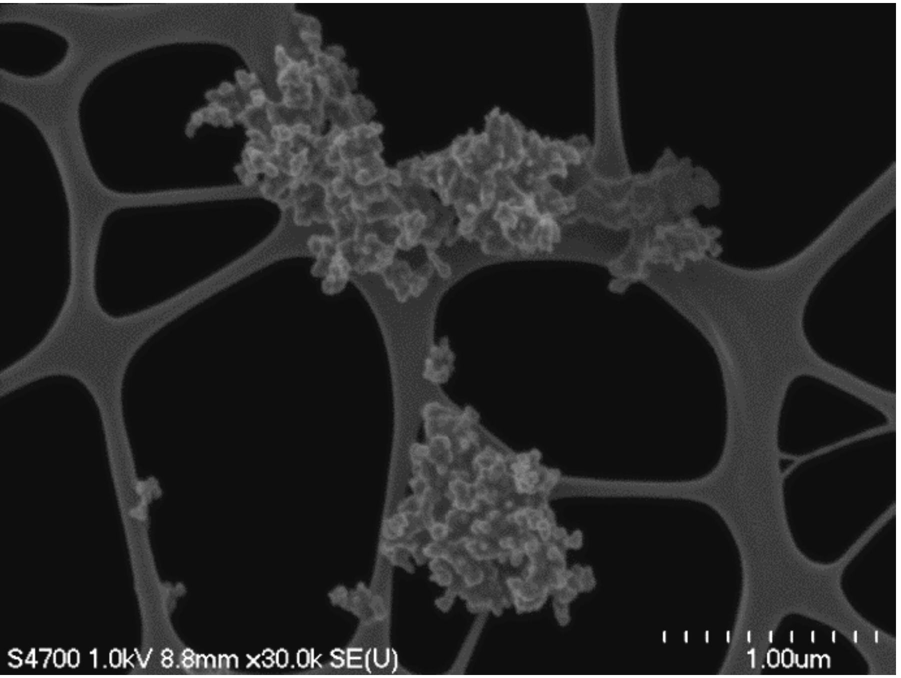
Scattering and Extinction

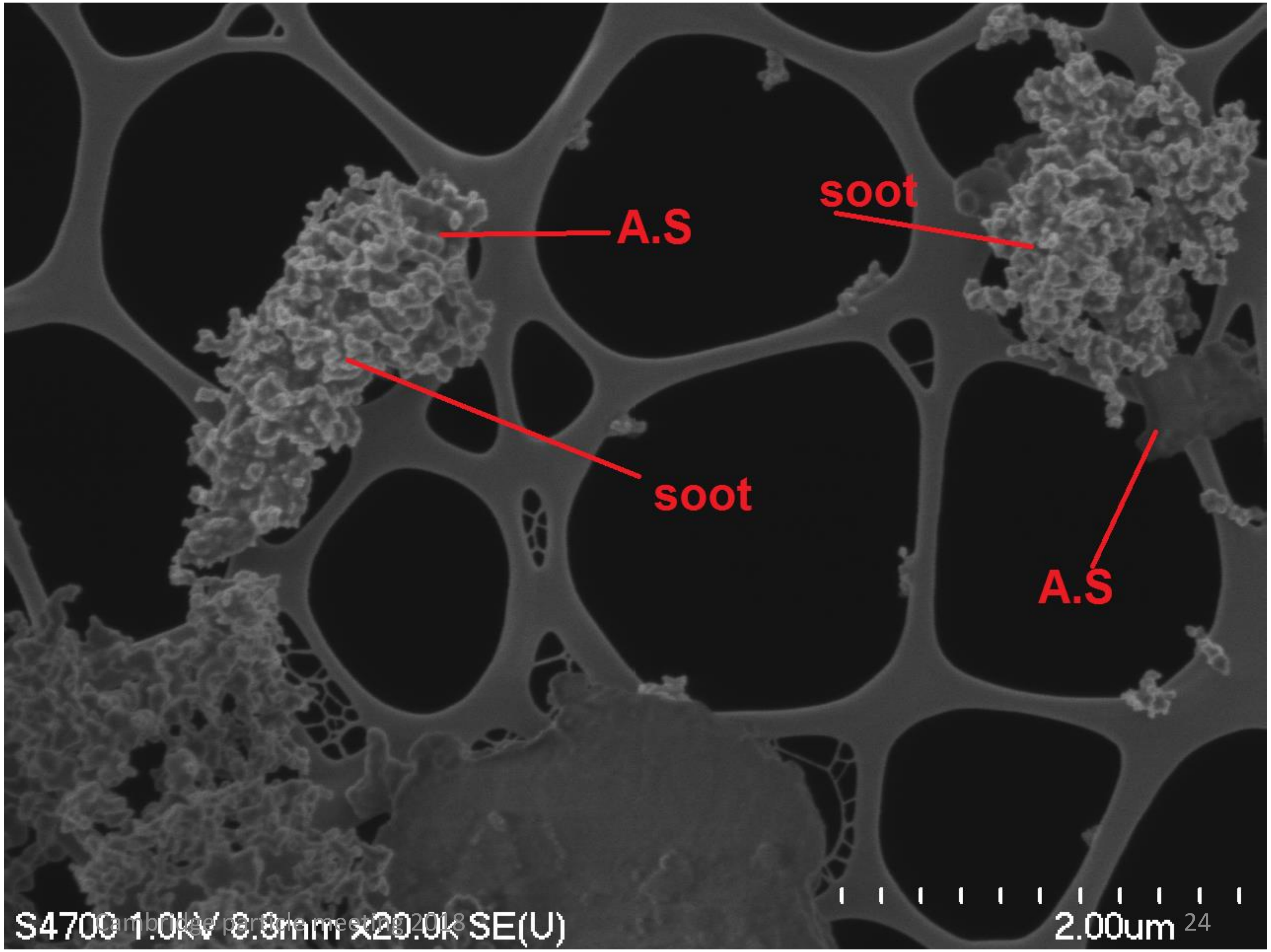
- Extinction – Cavity Attenuated Phase Shift Technique
- Scattering – Inverse Integrating Nephelometer
  Integrating Sphere with Lambertian Surface
  Minimal Bias w.r.t. Scattering Angle



Aerosol Sci. and Technol. 49:267-272 (2015)

Ammonium sulphate + soot dimers
SEM sample collection





Dioctyl sebacate oil