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Broadband Light Scattering and Cavity Ring-Down Spectroscopy Measurements on Single Light-Absorbing Aerosol Particles

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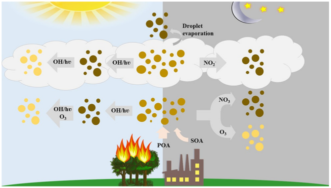
Project Goals

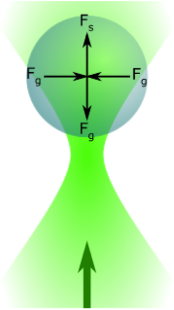
- Want to devise a method for determining the optical properties of absorbing aerosol
- Important for improving climate models as these commonly assume no absorption by aerosol
- Study single levitated particles to overcome averaging effects and maximise fundamental insight
- Use optical trapping to achieve levitation
- Combine cavity ring-down spectroscopy and broadband light scattering for measurements

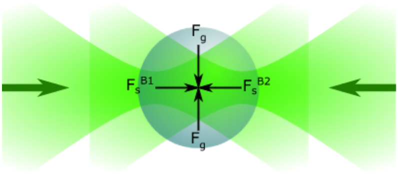


https://bc-policy-landscape.amap.no/areas-of-action/open-biomass-burning
Hems *et al.* *ACS Earth Space Chem.* **5**, 722–748 (2021)

Optical Trapping
Usual Methods

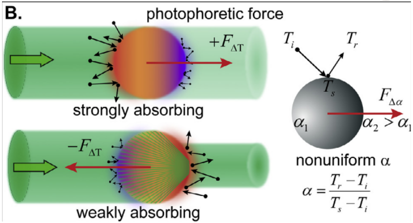
- Optical trapping uses optical forces to hold single particles in place
- Most optical traps consist of either one or two beams
- Good for long term observation of single particles

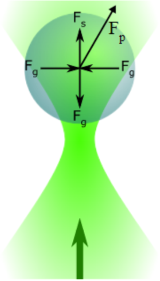




Trapping Absorbing Particles
Photophoresis

- Previously shown optical trapping configurations don't trap absorbing particles
- Photophoretic forces push the particle away from the highest intensity point
- The same intensity gradient responsible for trapping transparent particles stops trapping of absorbing ones



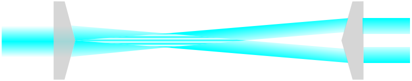


Gong *et al.* *JQSRT* **2018**

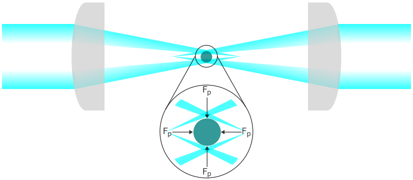
Trapping Absorbing Particles

Counterpropagating Hollow-Beam Trap

- Generate hollow beams using conical lenses



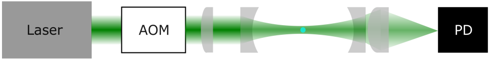
- Focusing two of these at slightly different point creates an intensity void
- When absorbing particle is illuminated, it heats unevenly giving photophoretic force

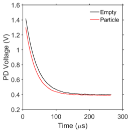


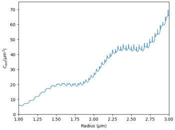
- Photophoretic forces push particle back into the void

Cavity Ring-Down Spectroscopy

The Basics





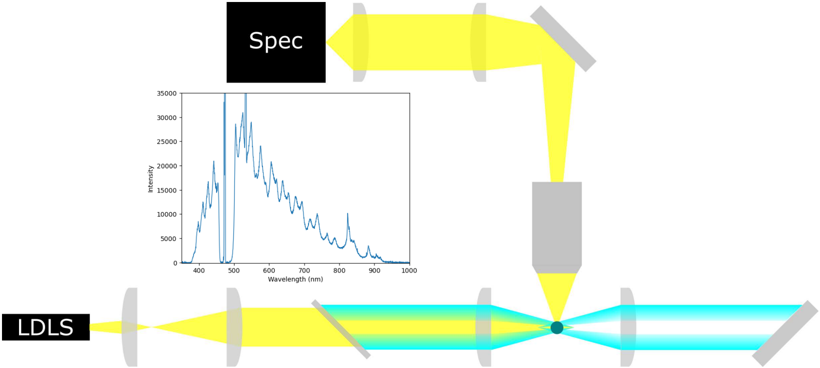


- CRDS provides sensitive measurements of particle extinction
- Watch decay in intensity of a laser pulse as it passes back and forth through cavity
- Decay time is related to extinction cross-section
- Needs a complementary method to disentangle various factor contributing to extinction

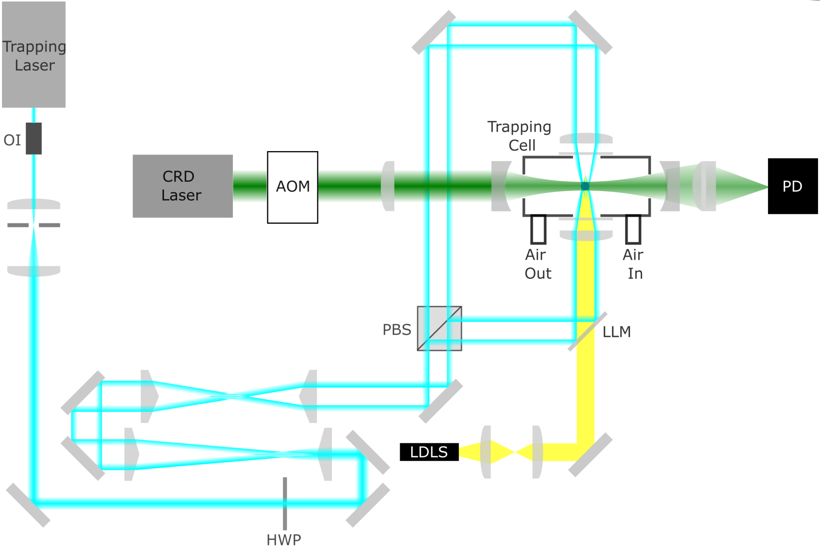
$$\sigma_{ext} = \frac{\pi L w_0^2}{2c}\left(\frac{1}{\tau} - \frac{1}{\tau_0}\right)$$

Broadband Light Scattering

- Broadband light scattering involves illuminating a particle with a spectrum of wavelengths and measuring the variation in scattered intensity as a function of wavelength
- Fitting of spectrum allows for determination of particle size and wavelength-dependent refractive index



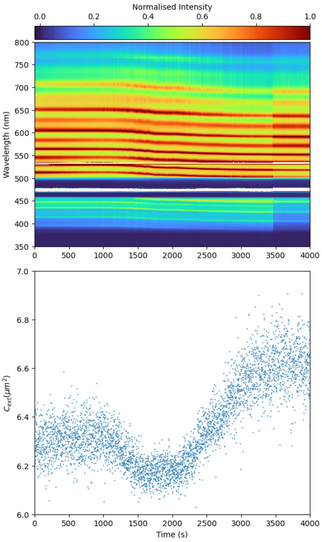
Setup


Trapping Laser
OI
CRD Laser
AOM
Trapping Cell
PD
Air Out
Air In
PBS
LLM
LDLS
HWP

Results

Sucrose/water/nigrosin

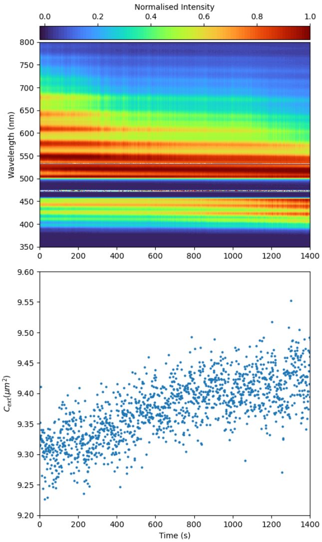
- Measured data for a hygroscopic system under changing RH conditions
- Used nigrosin because it is well-characterized, readily available dye
- See nice transition as RH changes from high to low



Results

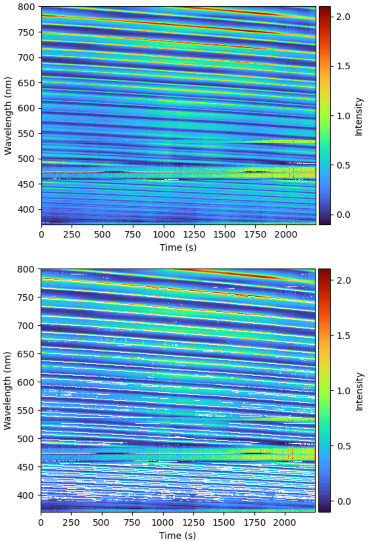
PEG-1500/nigrosin

- Also measured a volatile system with PEG-1500 and nigrosin
- Size change is small due to low vapour pressure
- Could imply significant heating



Analysis
Peak Extraction

- In order to develop data analysis approach, took some data with a volatile, nonabsorbing organic (1,2,6-hexanetriol)
- Allows results to be compared against established approaches (MRFIT)
- Remove background and break spectra into groups of peaks
- Fit groups to find peak positions
- Results a little noisy, need to optimise algorithm



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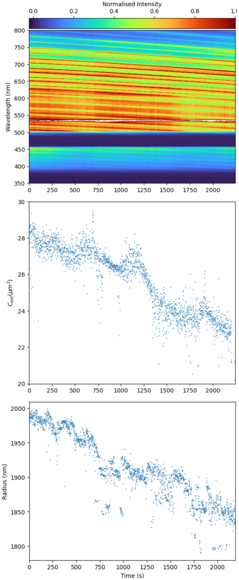

Conclusions and Future Work

- Built and optimised an instrument for measuring the optical properties of single absorbing aerosol particles
- Levitation of single particles is achieved using counterpropagating hollow beams
- Particles are interrogated using broadband light scattering and cavity ring-down spectroscopy
- Been able to make measurements on both volatile (PEG-1500) and involatile (aqueous sucrose) systems
- Fine tuning of data analysis continues using data from nonabsorbing particles