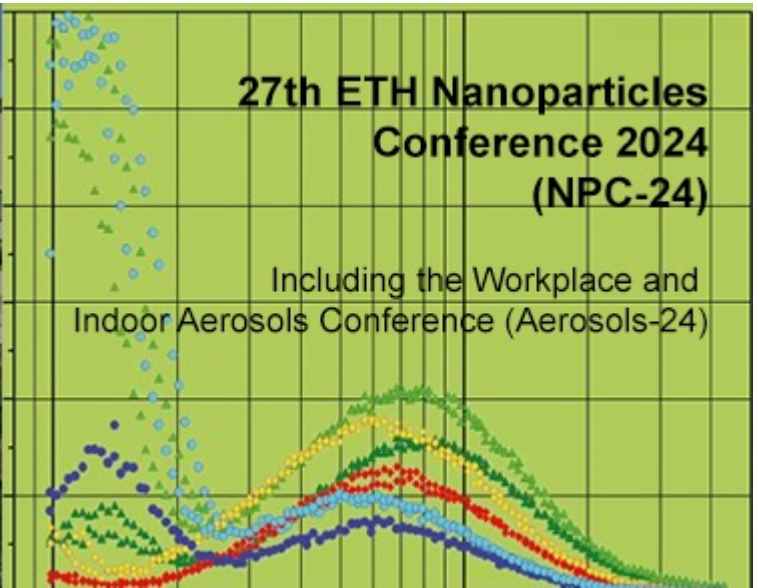
The effect of hydrogen addition on combustion generated nanoparticles from premixed ethylene flames



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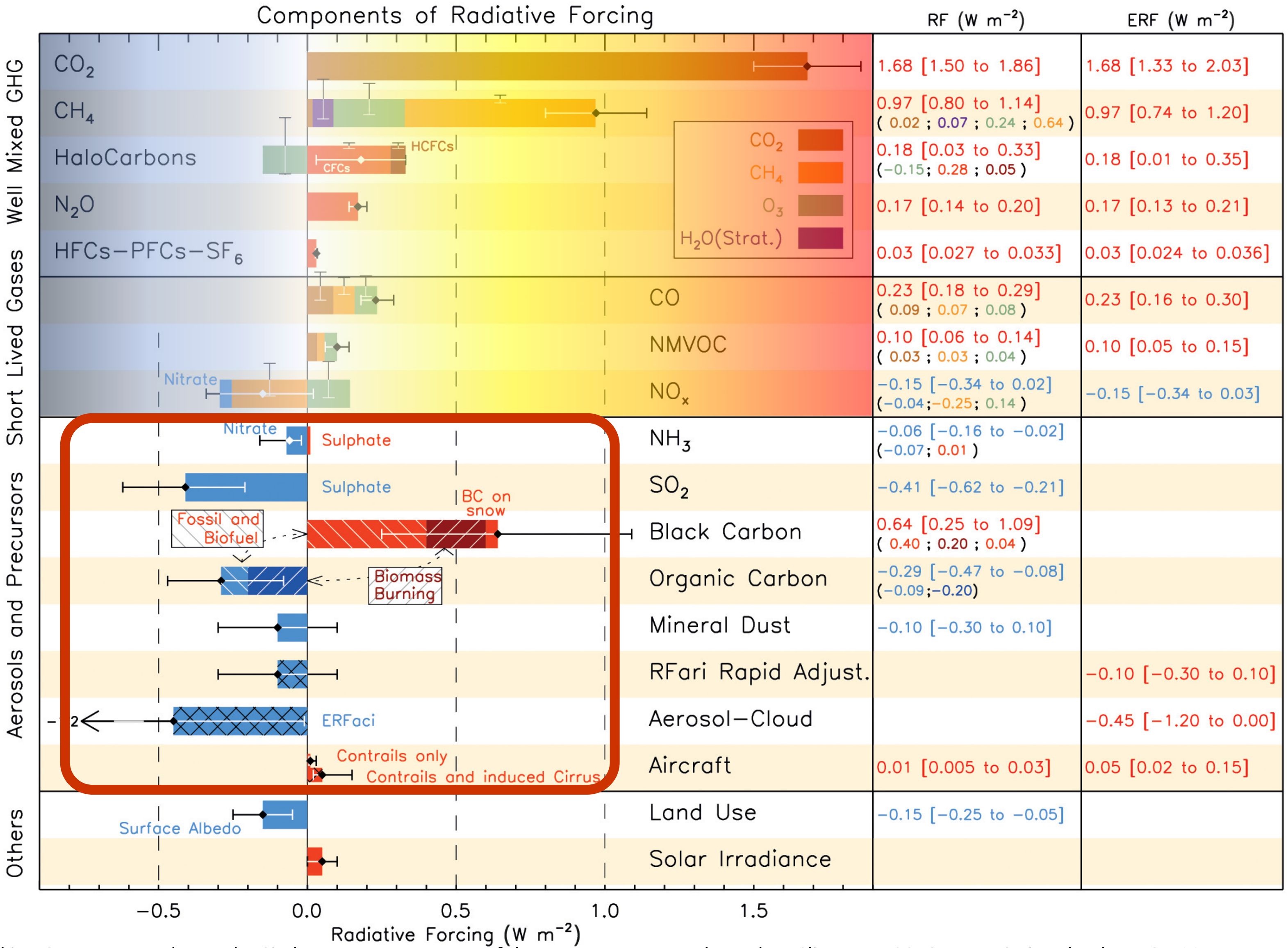
Date: 12-06-2024

Motivation

- Environmental impact of aerosols
- Black carbon

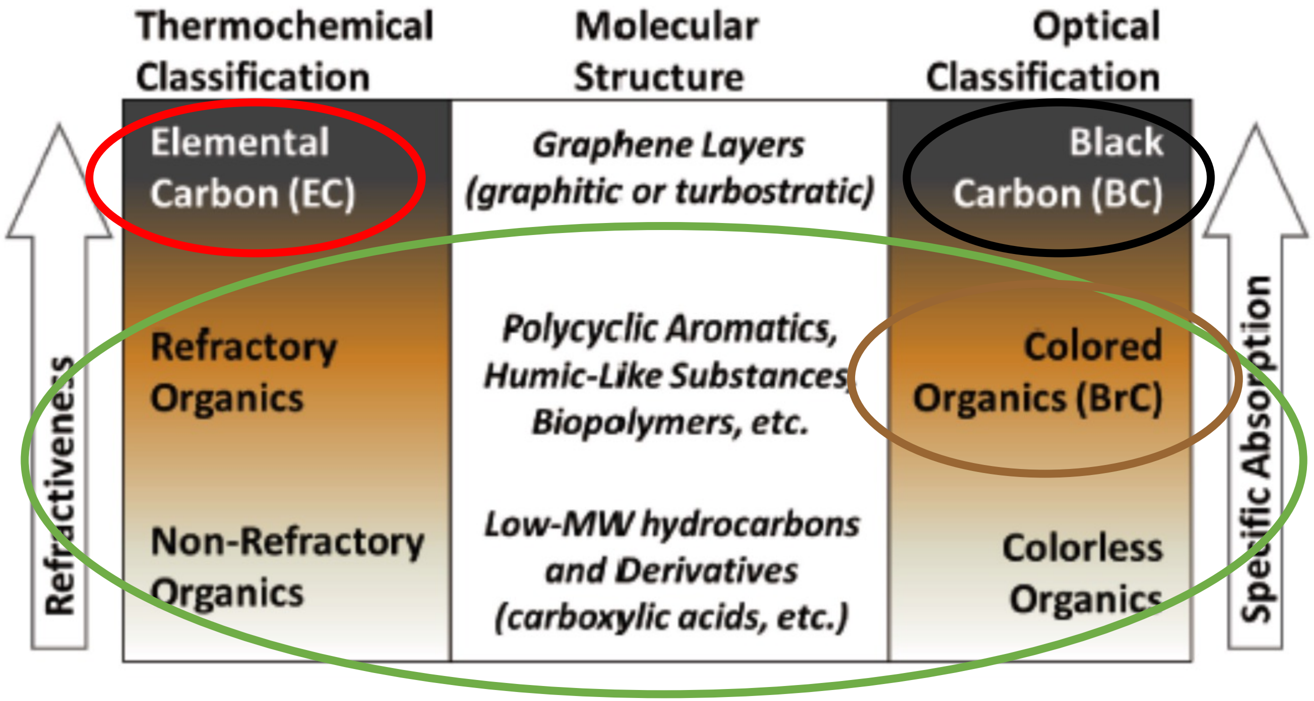
- Very potent climate forcer
- Very short lived

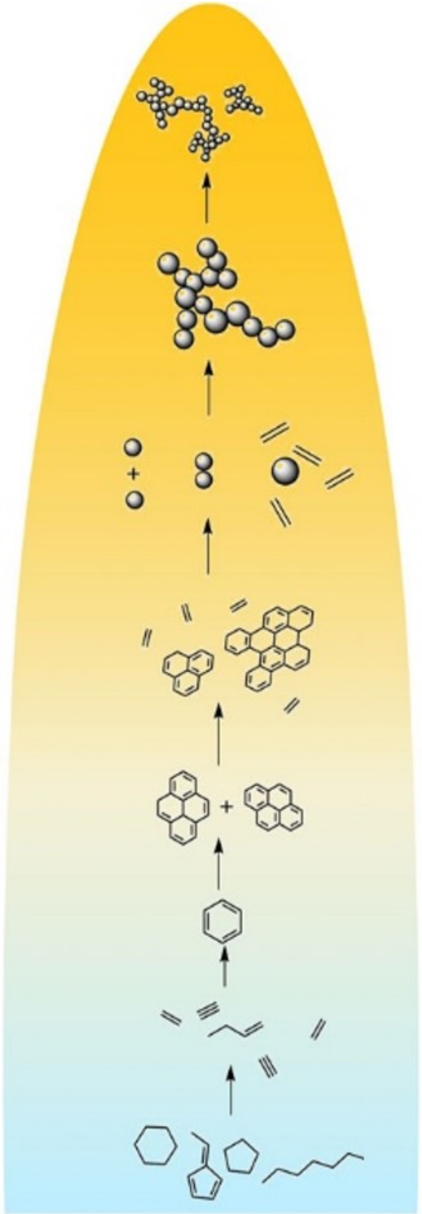


IPCC, 2023: *Climate Change 2023: Synthesis Report.* Contribution of Working Groups I, II and III to the Sixth Assessment Report of the Intergovernmental Panel on Climate. IPCC, Geneva, Switzerland, pp. 35-115

Different types of carbon aerosols

- Black Carbon
- Organic Carbon
- Brown Carbon
- Elemental Carbon
- Total Carbon
- Comparison is hard



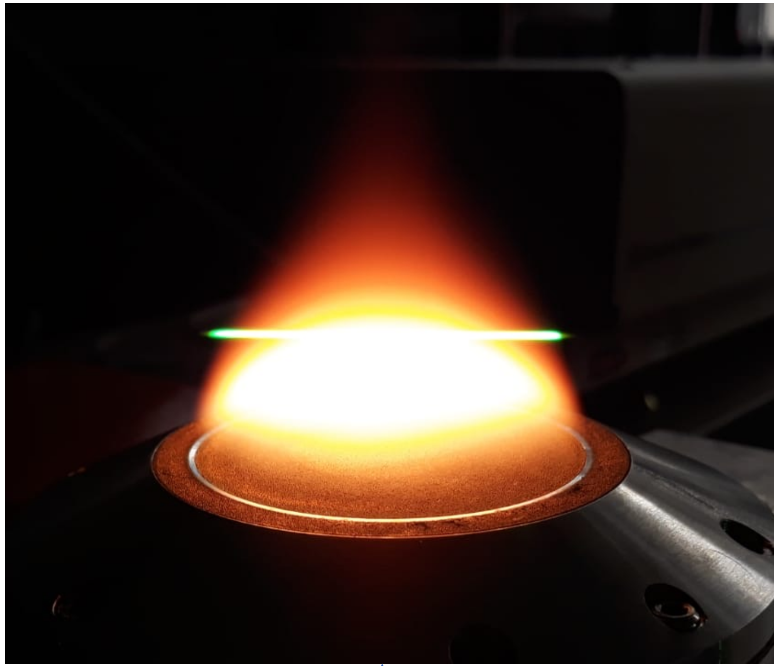


- Formation of soot
- Might co-fueling be a (temporary) solution?

Pöschl, U. (2005): Atmospheric Aerosols: Composition, Transformation, Climate and Health Effects, Atmospheric Chemistry, 44, 7520–7540
Xi, J et al (2021).: A Review of Recent Research Results on Soot: The Formation of a Kind of Carbon-Based Material in Flames, Frontiers in Materials, 8, 695485

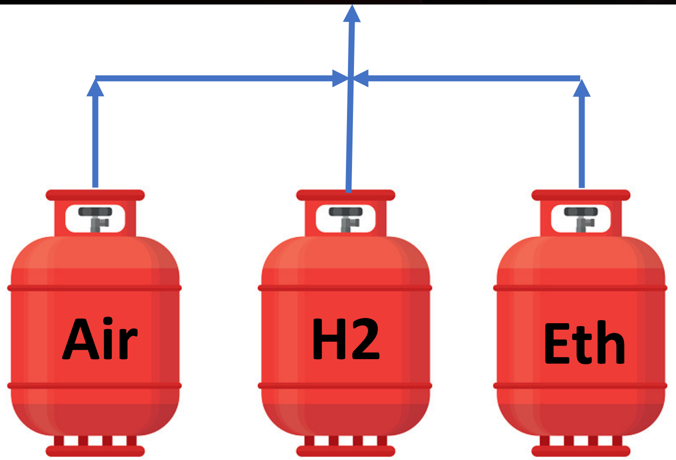
Flames of interest

- Fuel rich (φ = 2.3)
- Premixed laminar flames
- Fuels: Ethylene (C_2H_4) and Hydrogen
- Hydrogen mole fraction α ranges from 0 – 0.5
- Fuel exit velocity ranging from 6 – 10 cm/s to parameterise flame temperature



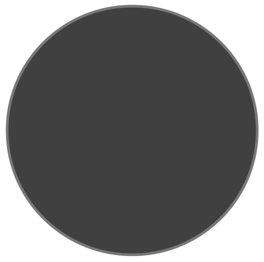
- *Do the optical, physical and thermochemical properties of the aerosol change as a function of hydrogen addition and/or flame temperature?*

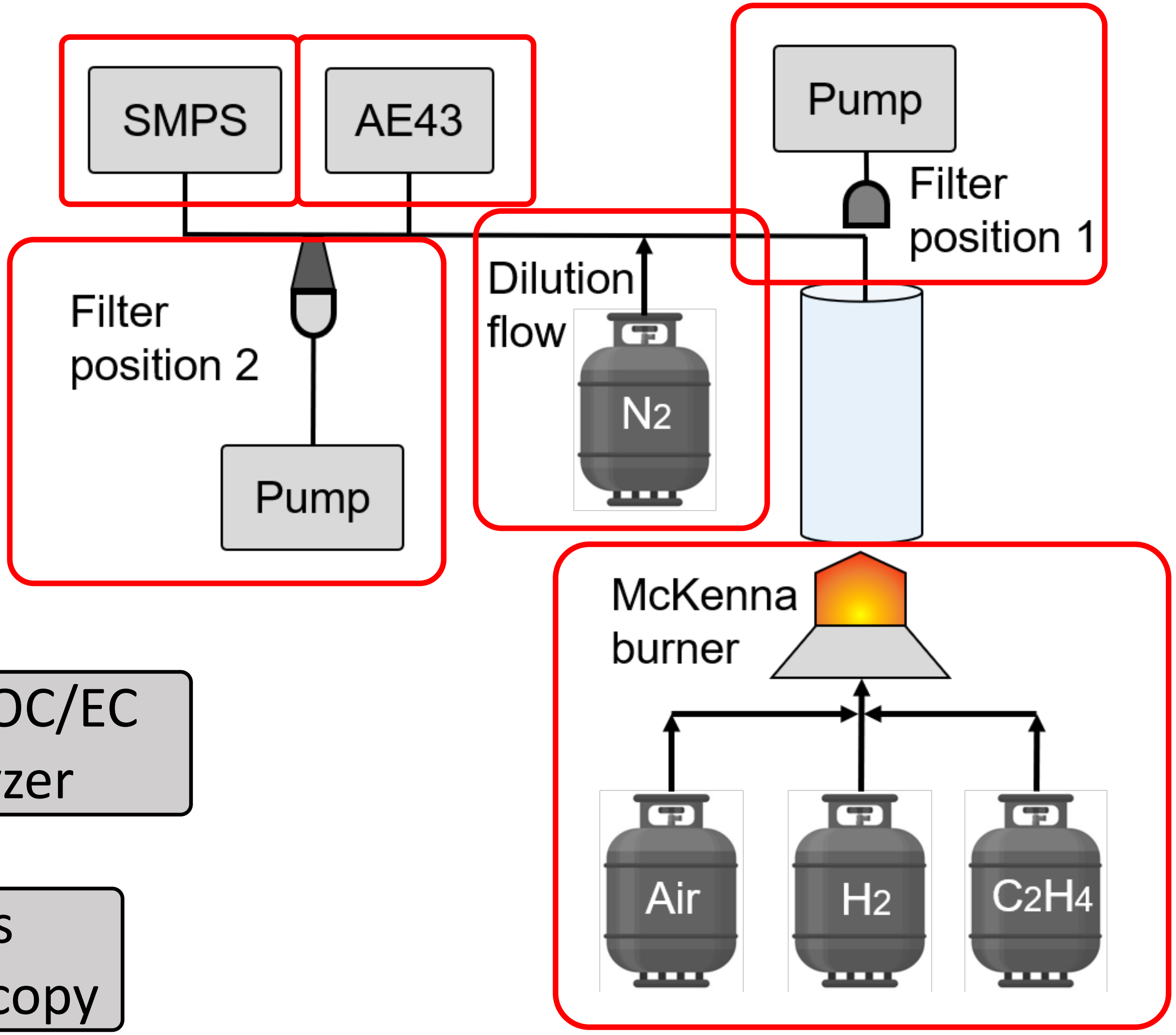
$$\phi(1-\alpha)\,C_2H_4 + \phi\alpha H_2 + (3-2.5\alpha)(O_2 + 3.78\,N_2)$$



Experimental setup

- Flame setup
- Dilution with Nitrogen
- Particle size distribution
- Optical absorption spectrum
- Filter sampler
- Mass of OC and EC
- Optical properties of OC





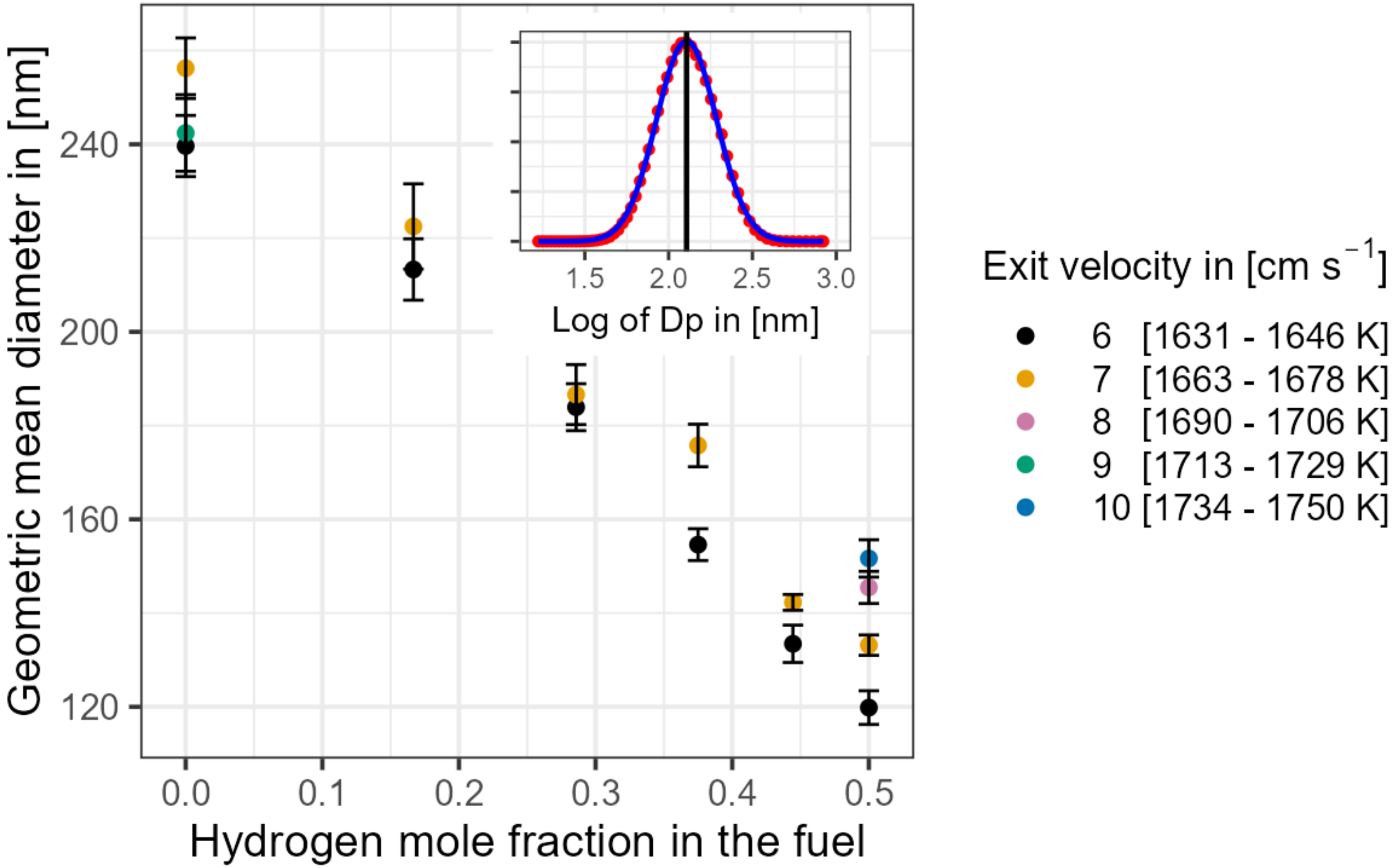
Sunset OC/EC analyzer

UV-vis Spectroscopy

Results: Count particle distribution

- Lognormal distribution
- No bimodal distribution

- Smaller particles for high hydrogen content
- Exit velocity is less impactful

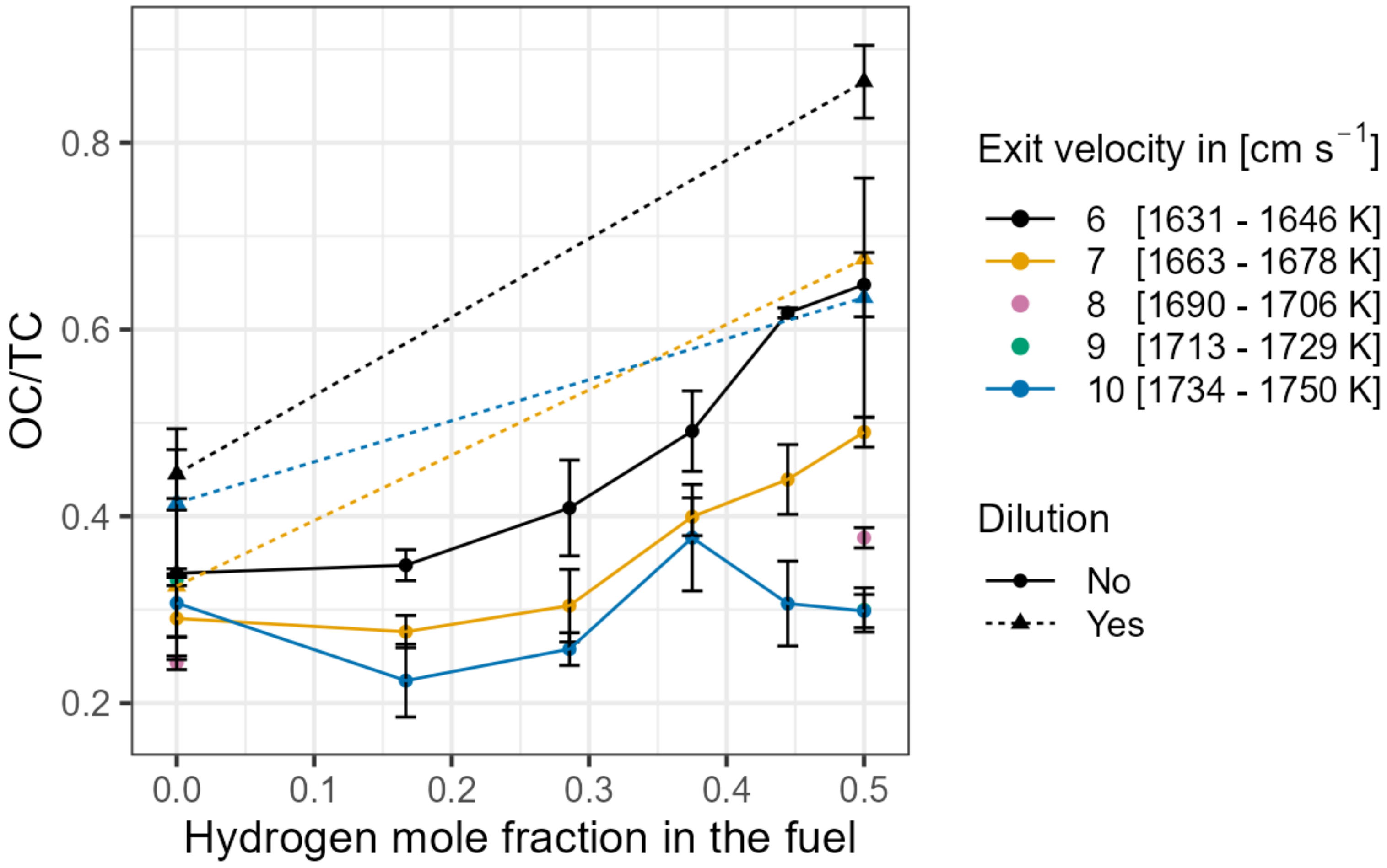


Results: Thermochemical properties

- Example of filters
- Sunset OC/EC analyser

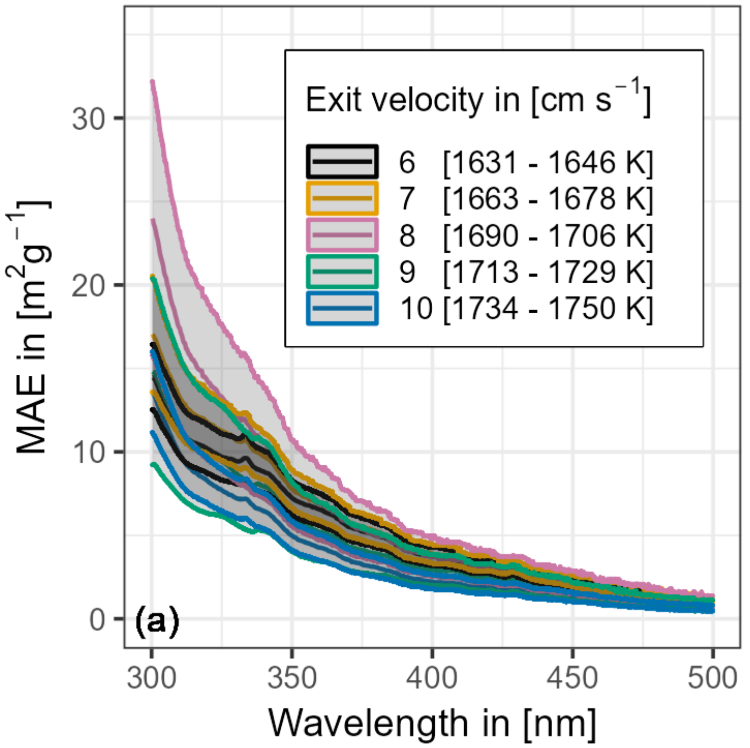
- EC dominant aerosol for low hydrogen content and or high exit velocities
- For high H2 content **combined** with low exit velocity, OC is dominant

- Dilution increases OC/TC



Results: Optical properties of OC

- Mass Absorption Efficiency
- Methanol dissolved, not PM
- No absorption after 500 nm
- Material doesn't change with flame parameters



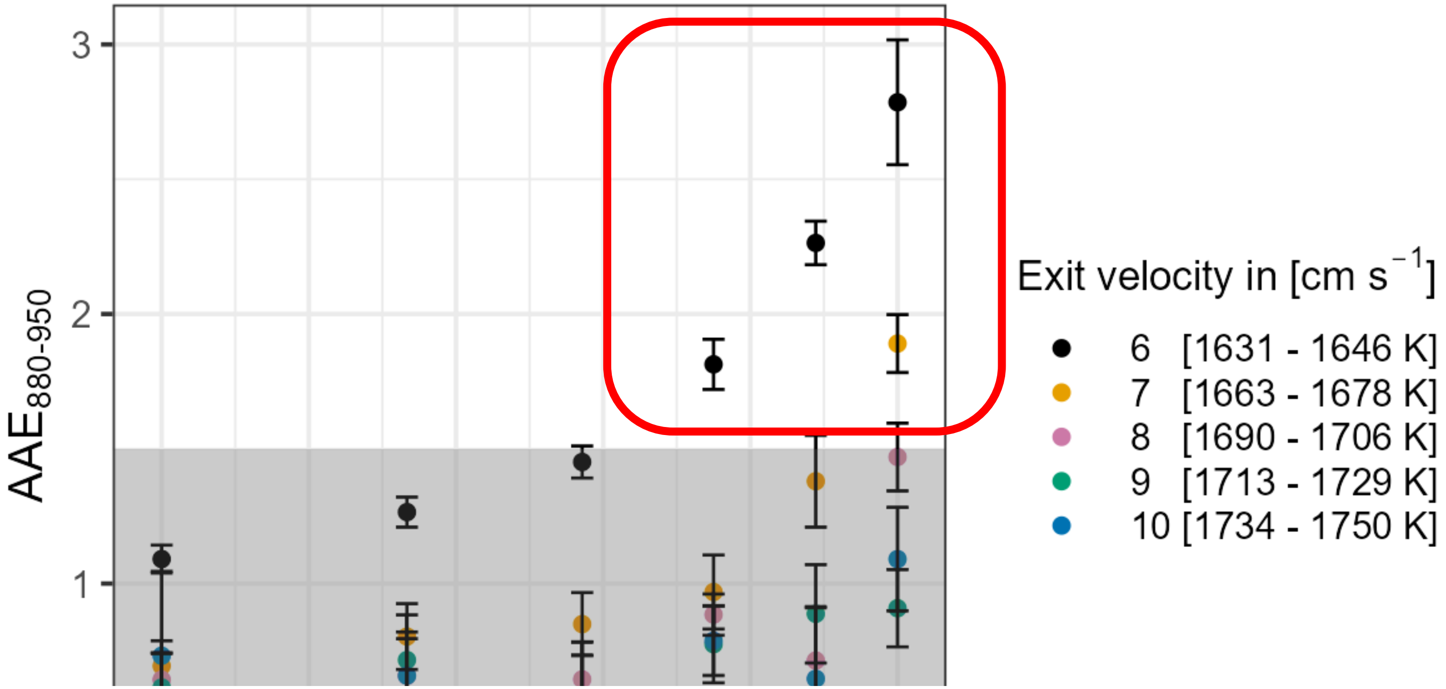
- Angström Absorption Exponent
- Same conclusion

$$MAE_{OC,\lambda} = \frac{b_{abs,dis,\lambda}}{c_{OC}}$$

$$C_{abs}(\lambda) = C_{abs}(\lambda_0) * \left(\frac{\lambda}{\lambda_0}\right)^{-AAE}$$

Results: Optical properties EC

- AAE 880-950
- Grey area denoted common BC values
- BC seems possible, except for these flame conditions
- Compare with MAC_{EC}
- MAC_{EC} of pure BC is 4.68 m2/g



Exit velocity in $[cm\ s^{-1}]$	Hydrogen mole fraction	EC MAC 880 in $[m^2 g^{-1}]$
6	0	11.5 ± 0.5
6	0.5	5.9 ± 0.5
7	0	7.7
7	0.5	9.1
10	0	9.8 ± 0.8
10	0.5	11.8 ± 0.6

- We conclude: Our EC does NOT resemble pure BC

$$MAC_{EC,\lambda=880} = \frac{b_{abs,\lambda=880}}{c_{EC}}$$

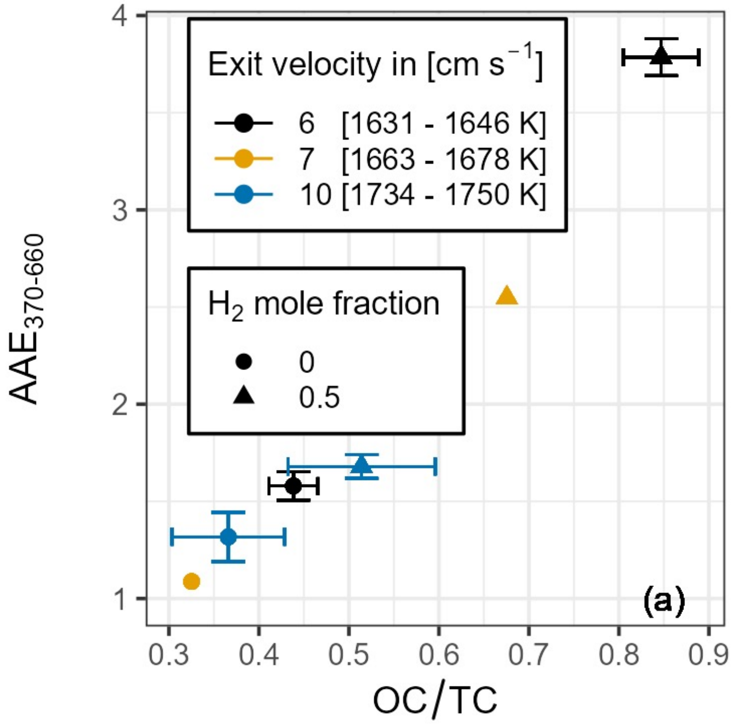
Bond, T. C. (2013).: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research: Atmospheres, 118, 5380–5552

Discussion

- Relate optical properties to OC/TC
- Very clear relation between AAE and OC/TC

- Relate MAC_{TC} to OC/TC
- Increased MAC_{TC} at first
- Later on, MAC_{TC} decreases

- Lensing effect could be an explanation

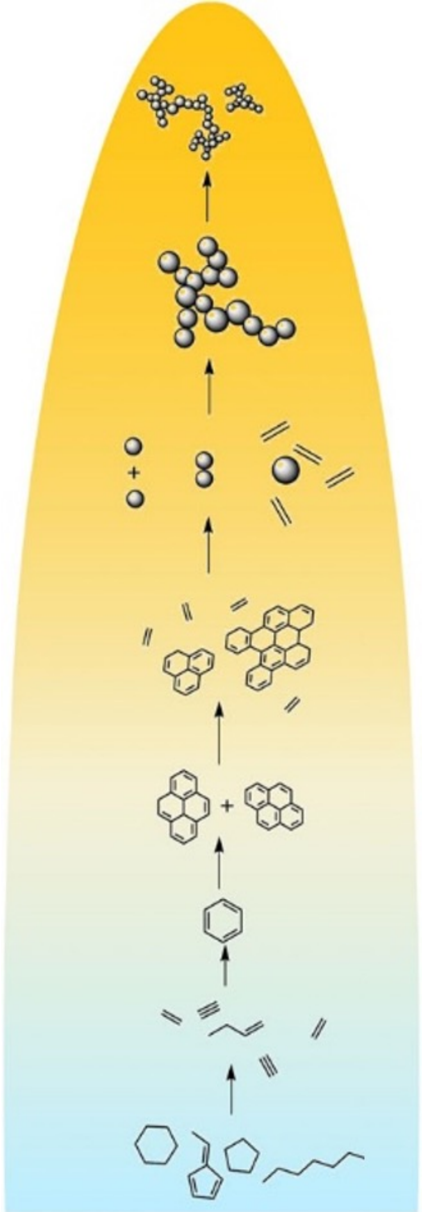


Discussion points

- Hydrogen addition decreases the particle size, but increases the volatility and absorption in shorter wavelenghts

- Enhanced absorption in shorter wavelengths due to coloured OC
- Our EC does not resemble pure BC
- Part of the OC that we found is a precursor to the EC
- The lensing effect could explain some of the increased light absorption efficiency

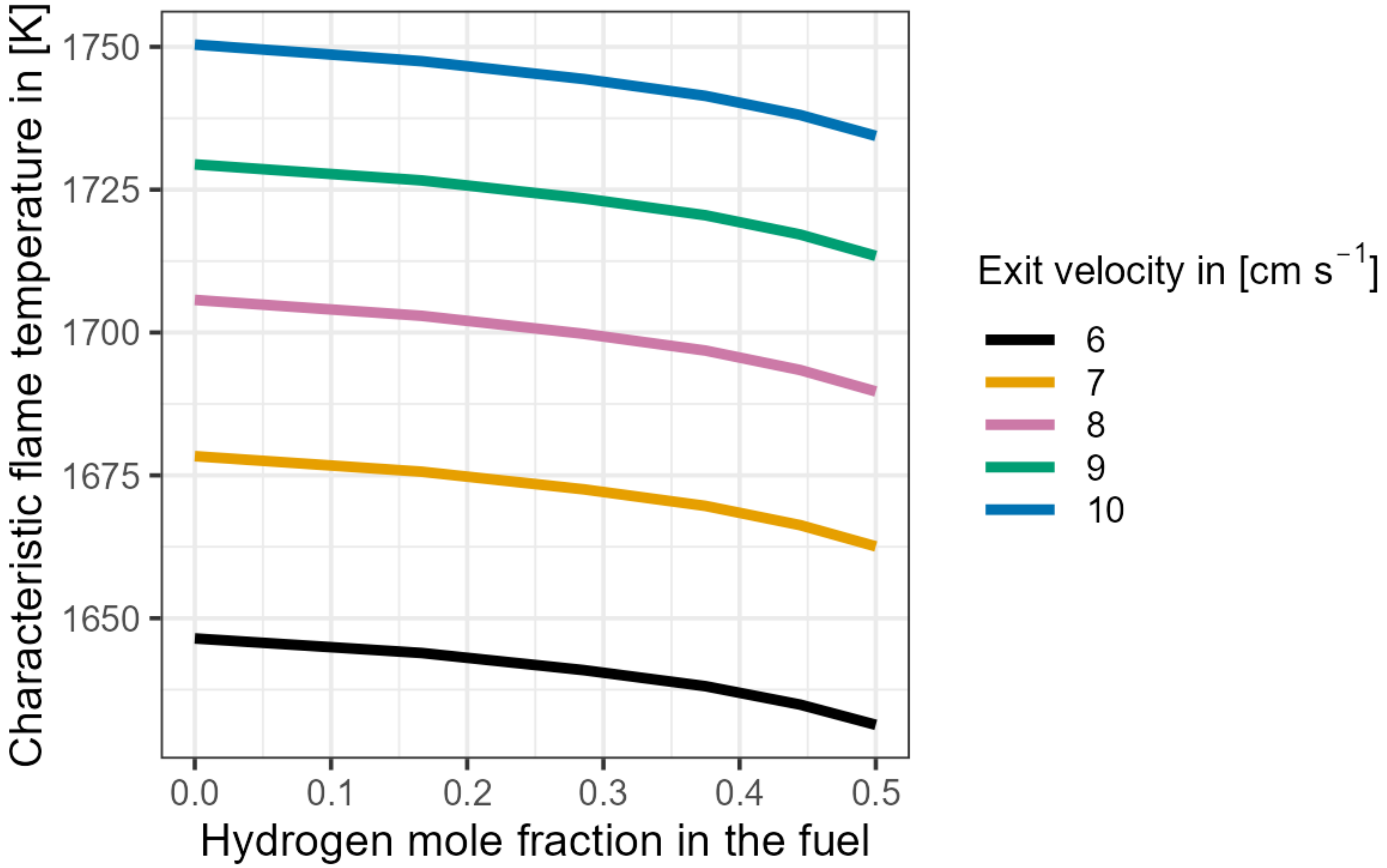
- Questions ?



Backup slides

Flame temperature

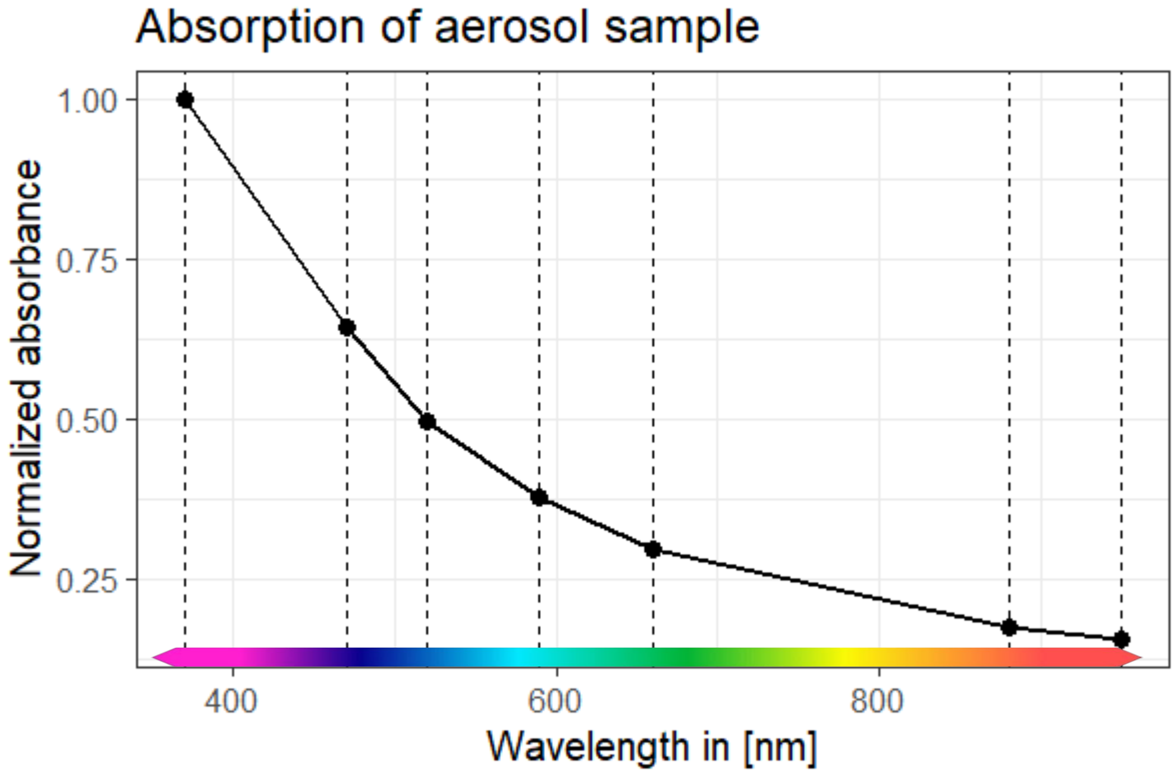
- San Diego mechanism
- Characteristic temperature taken at HAB = 1
- Hydrogen addition cools the flame
- Exit velocity has larger impact



Optical properties

- Ex-situ -> Above the flame
- Wavelength depending absorption
- Absorption Angstrom exponent
- AAE of Black Carbon ≈ 1
- AAE of Brown Carbon > 1

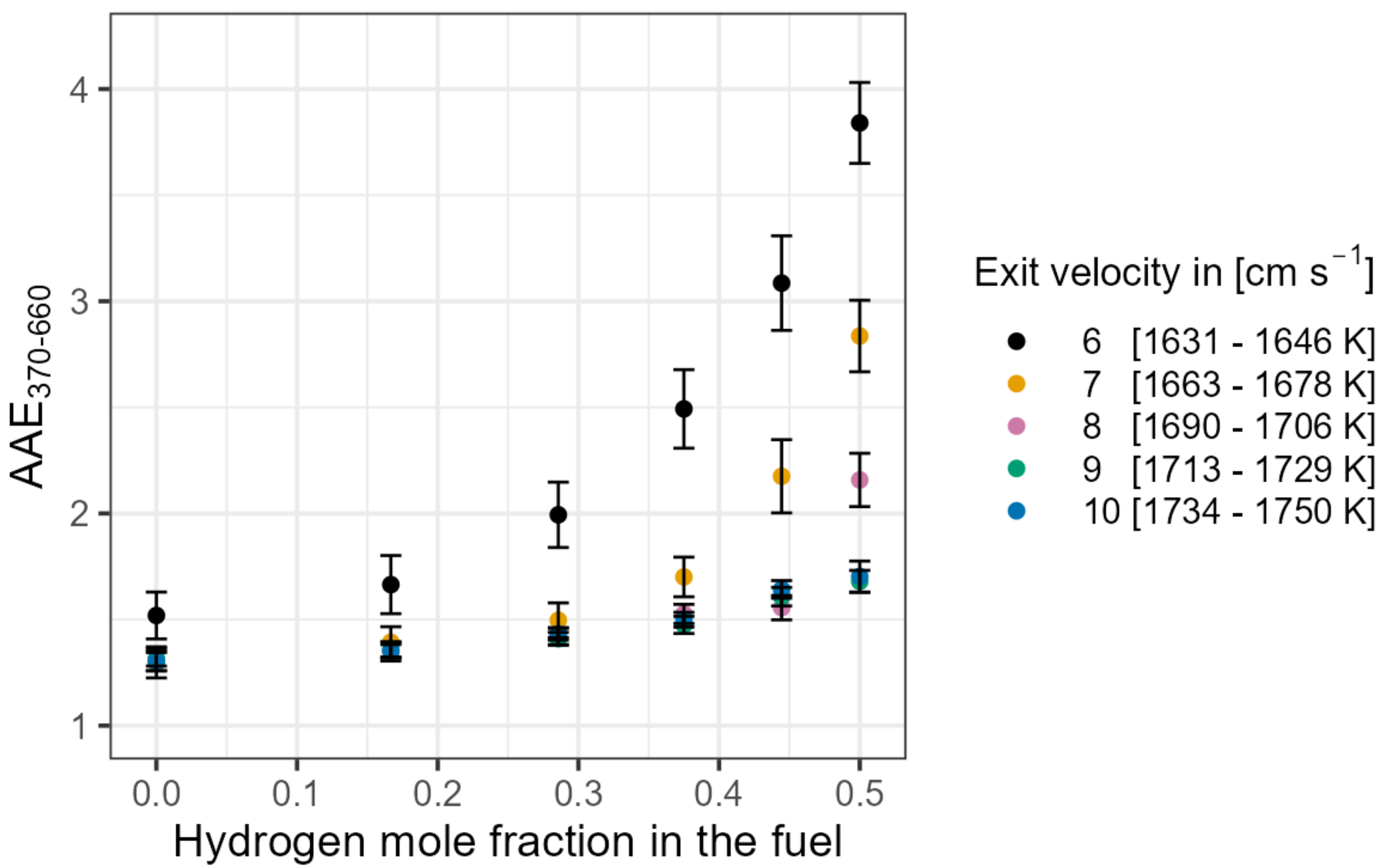
$$C_{abs}(\lambda) = C_{abs}(\lambda_0) \; * \; \left(\frac{\lambda}{\lambda_0}\right)^{-AAE}$$



BC + BrC **BC + BrC** **BC + BrC?** **BC** **BC**

Optical properties TC

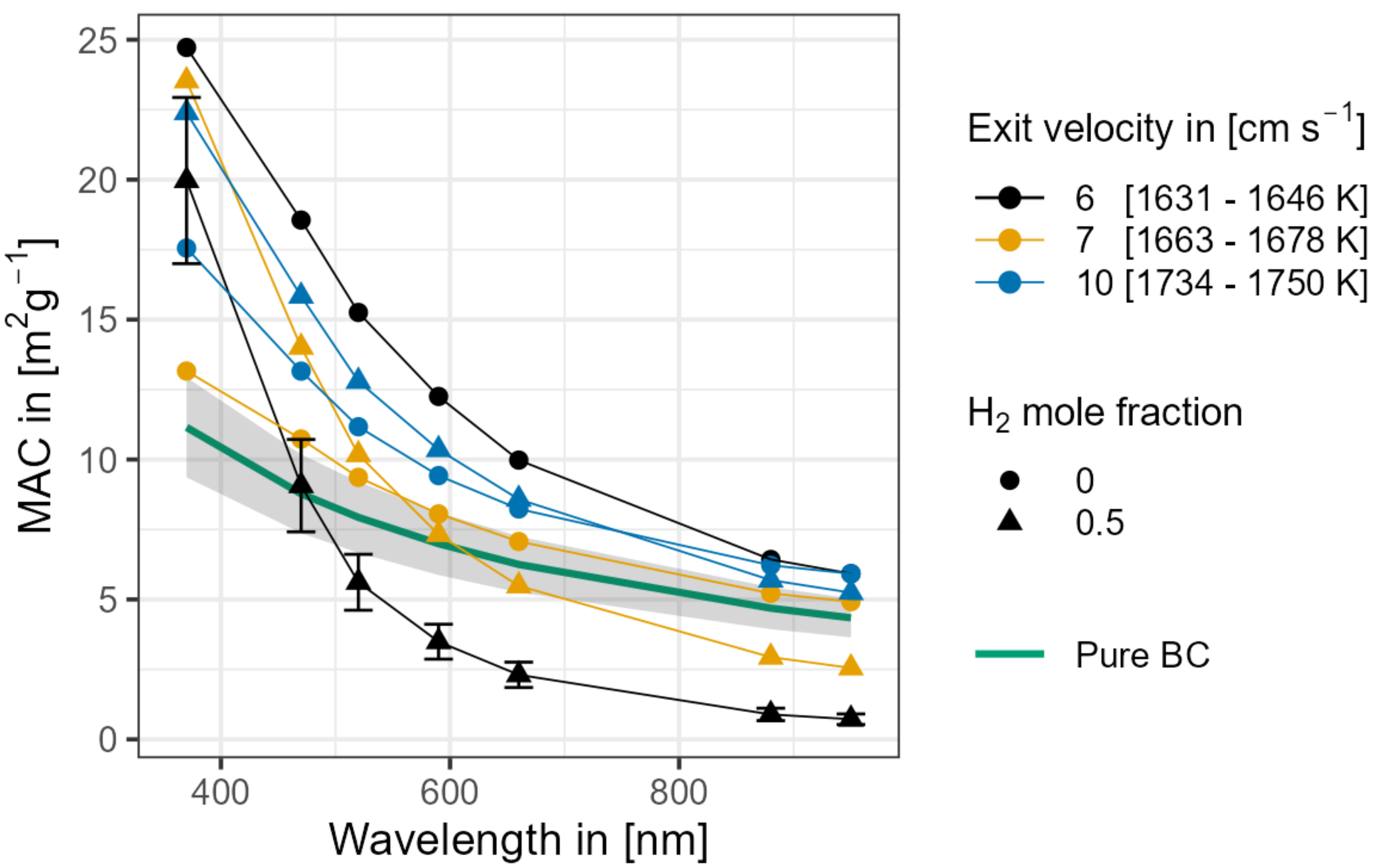
- AAE 370-660
- All above 1
- Resembles the OC/TC graph a lot
- For high hydrogen content **combined** with low exit velocity, OC is dominant



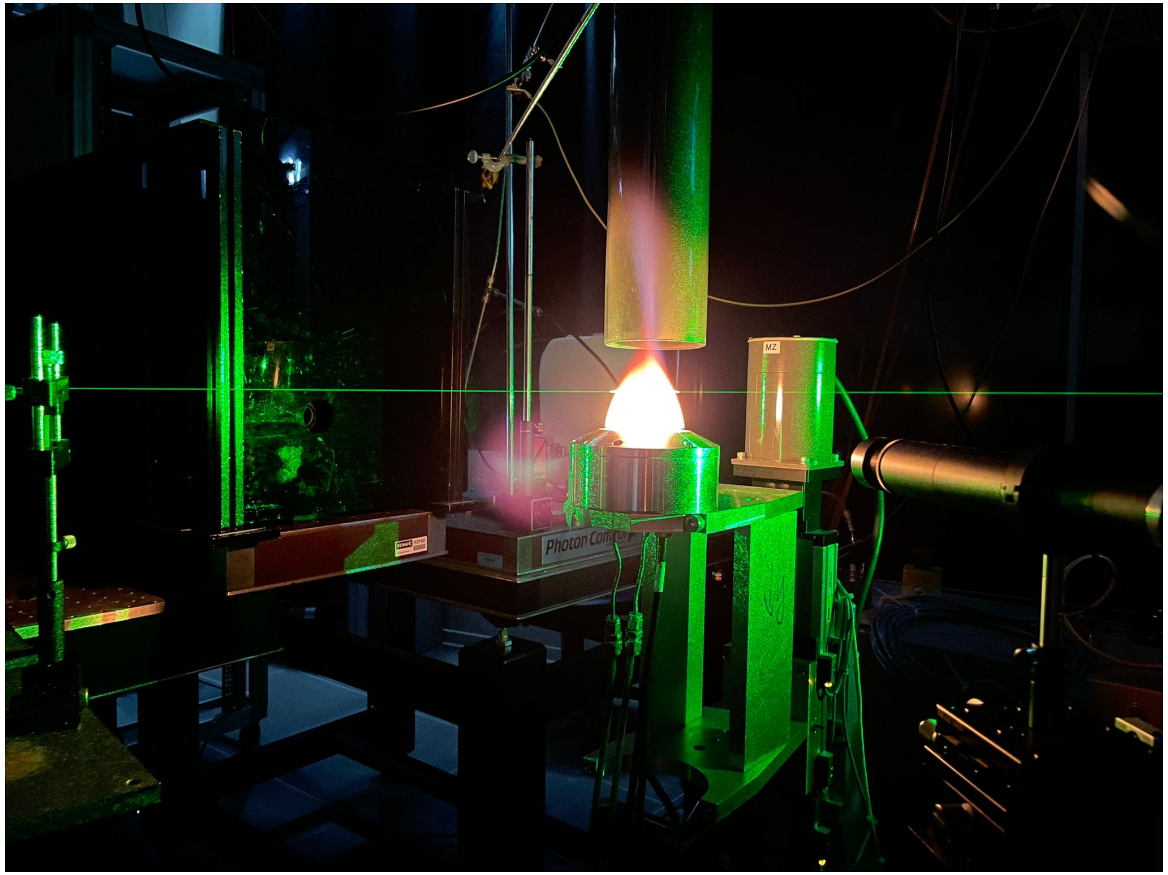
Results: Optical properties TC

- TC Mass Absorption Cross section
- Green line is pure BC
- OC increase absorption in the shorter wavelengths.
- Longer wavelengths seem ambiguous

$$MAC_{TC,\lambda} = \frac{b_{abs,\lambda}}{c_{TC}}$$

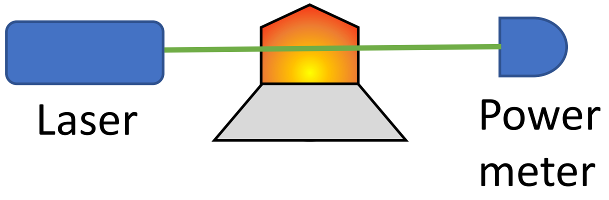


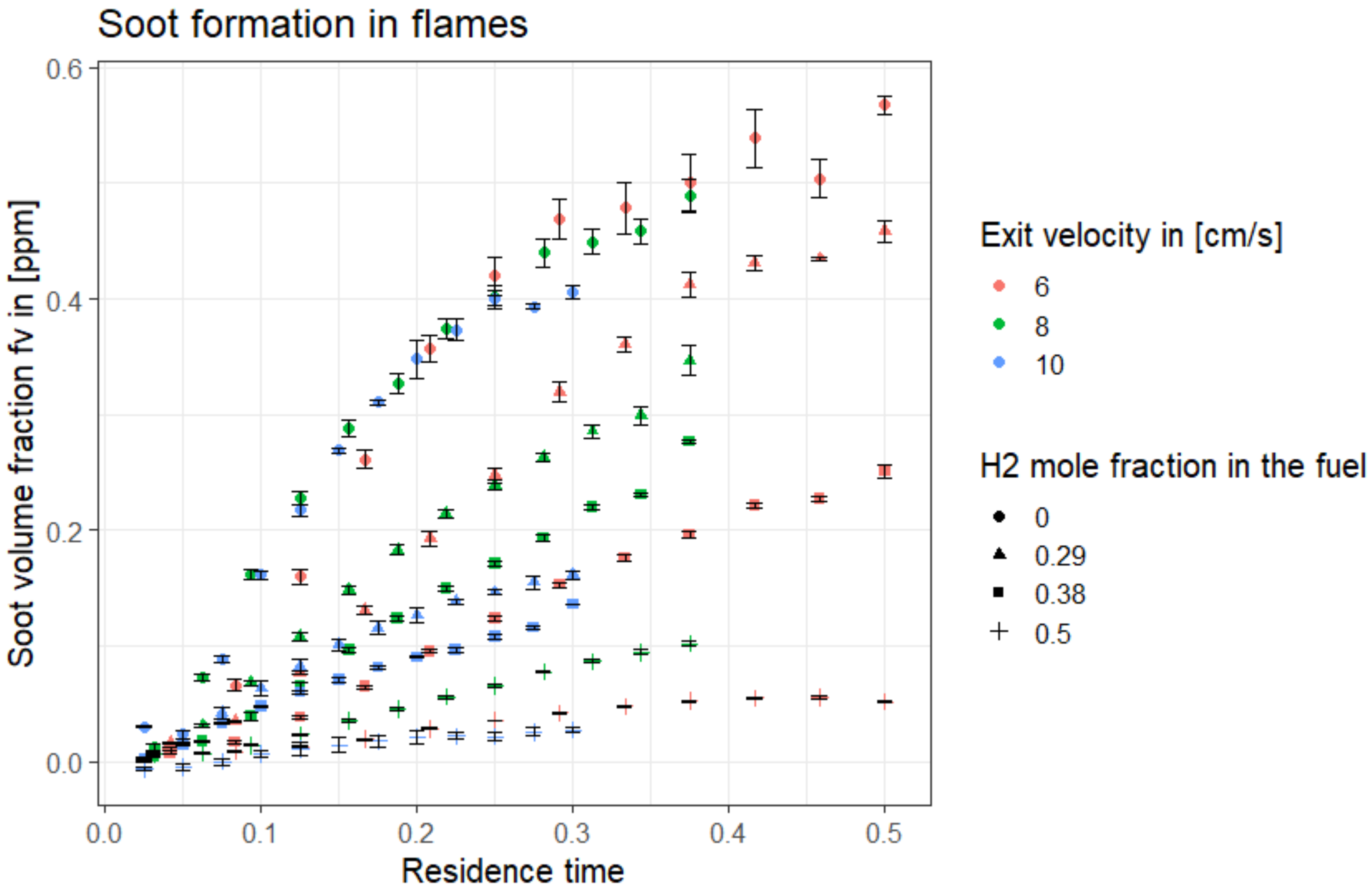
BC volume fraction by LLE



BC volume fraction by LLE

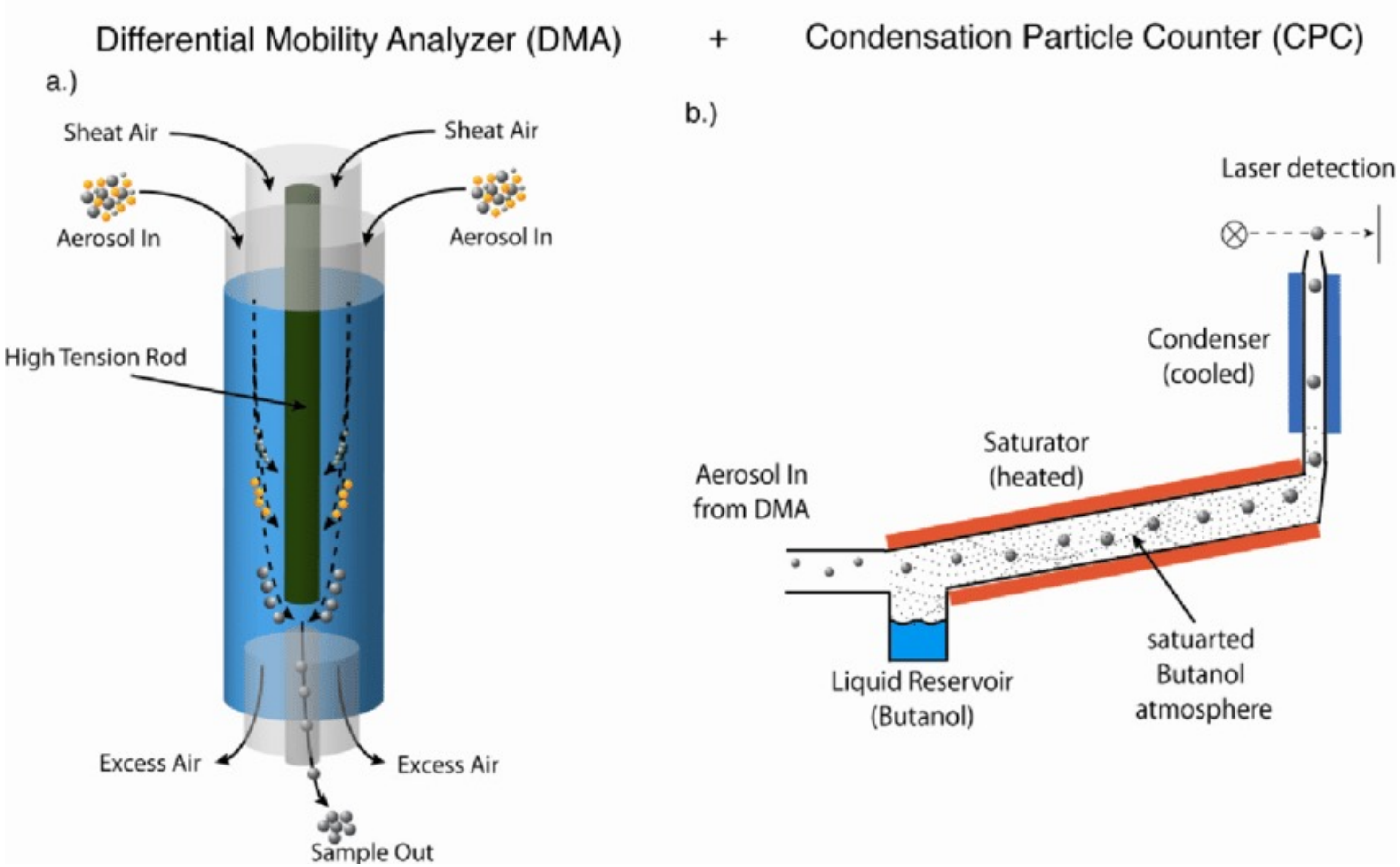
- Laser light extinction
- Measure decrease in laser intensity
- Differentiate HAB, calculate residence time
- BC volume fraction reduction by H2 addition





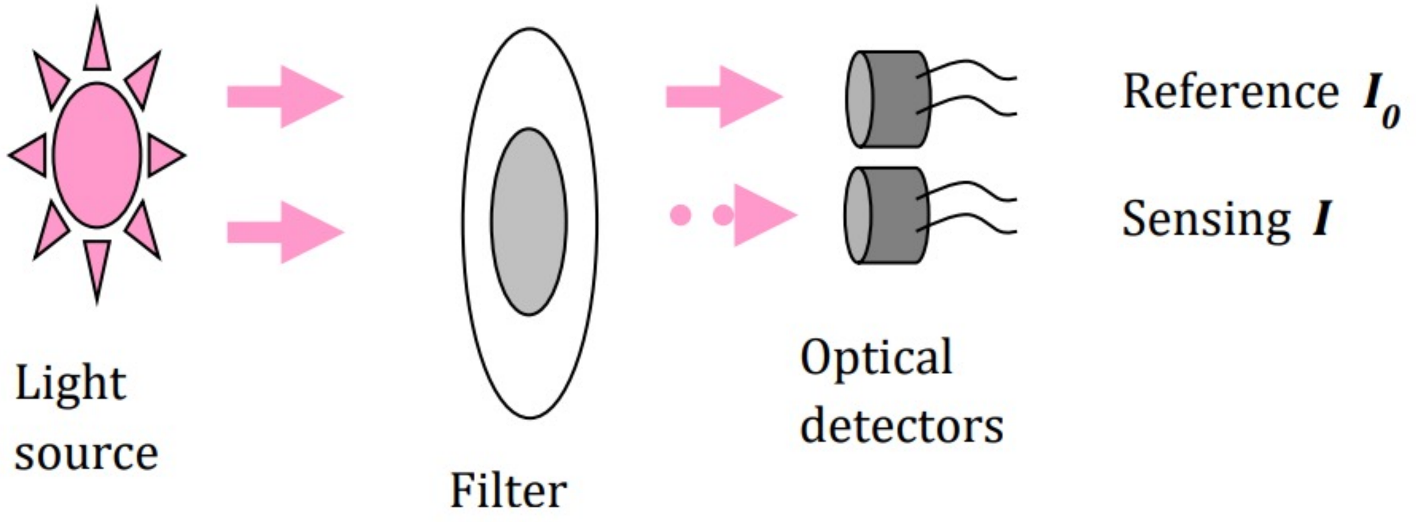
Working principle SMPS

- Particle sizer
- DMA
- CPC



Working principle Aethalometer

- Wavelength depending absorption
- Sample deposition
- Dual spot
- Attentuation on 7 wavelengths



Working principle Sunset OC/EC

- Sample filter
- Heat in helium environment for OC extraction
- Heat in helium + oxygen environment for EC extraction
- Measure CO2 from extracted carbon