SP-AMS: $C_{>5}^{+}$ and soot nanostructrure

Vilhelm B. Malmborg, **Axel C. Eriksson**, Sandra Török, Yilong Zhang, Kirsten Kling, Edward C. Fortner, Louise Gren, Sanghoon Kook, Timothy B. Onasch, Per-Erik Bengtsson , Joakim Pagels

See also Malmborg et al 2018:

https://www.sciencedirect.com/science/article/pii/S0008622318309886

Rationale for studying soot nanostructure

- Soot formation and oxidation (destruction)
- Reactivity (particle filter regeneration and biological)
- Source attribution
- Radiative forcing (colour)
- Ice nuclei??

Nanostructure metric:
Fringe (”aromatic island”)
length

Quantifying Soot Nanostructure with HR-TEM

- **HR-TEM analysis:**
  - Fringe length [nm]
  - Tortuosity $\frac{Fringe\ length}{Endpoint\ distance}$

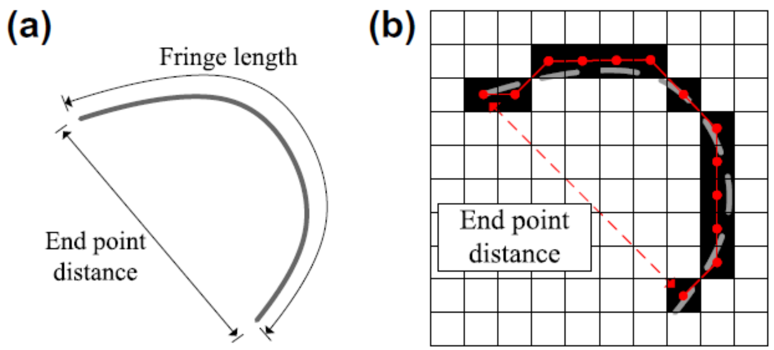
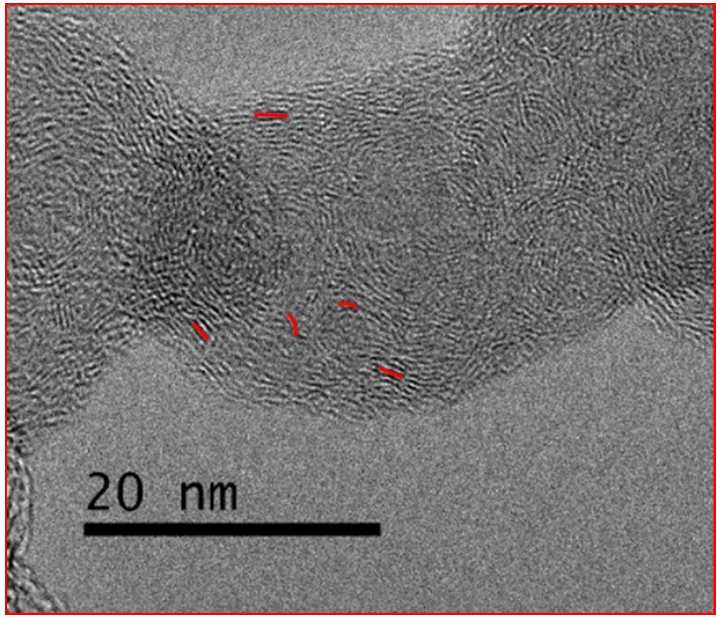


Fig. 10. Calculation of fringe length and tortuosity: (a) scheme in the continuous domain, and (b) approximation in the digitized image domain.

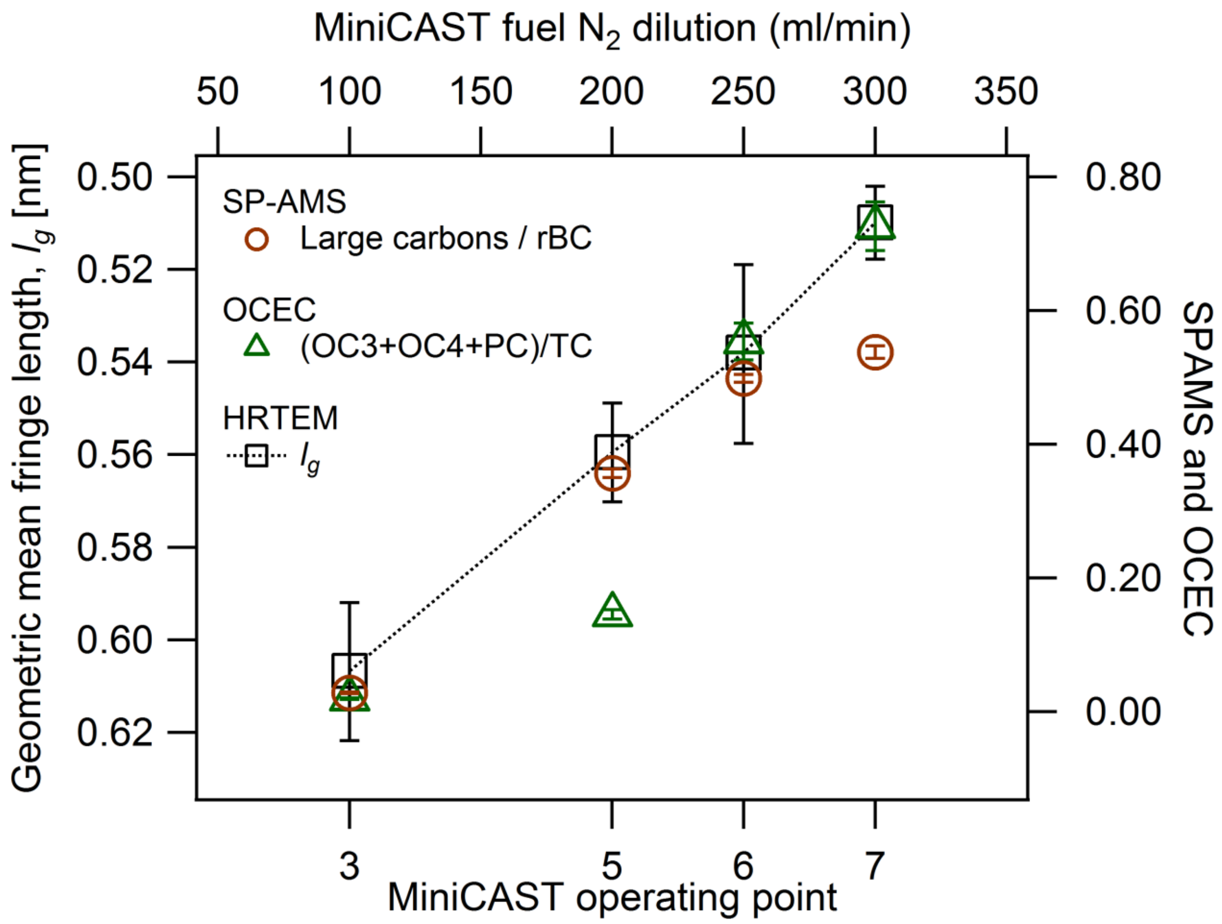
Yehliu et al. / Combustion and Flame 158 (2011) 1837–1851



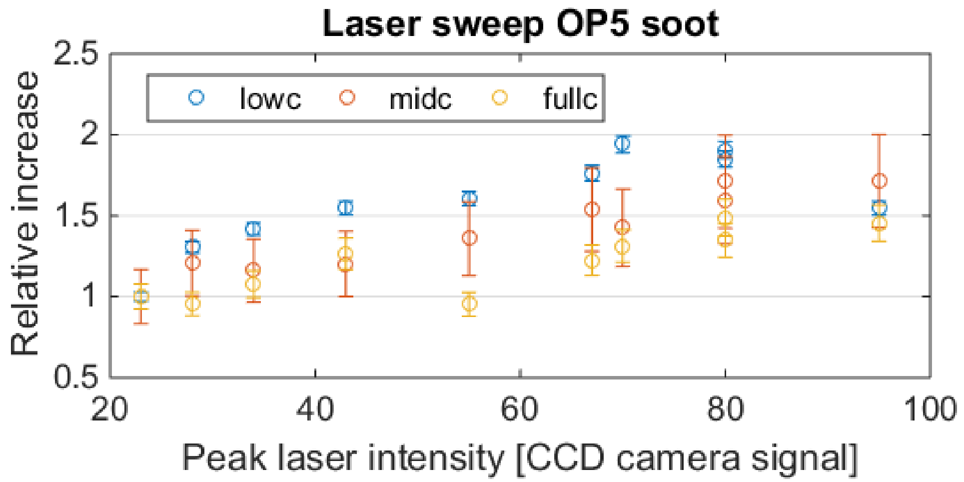
Can we measure soot nanostructure?

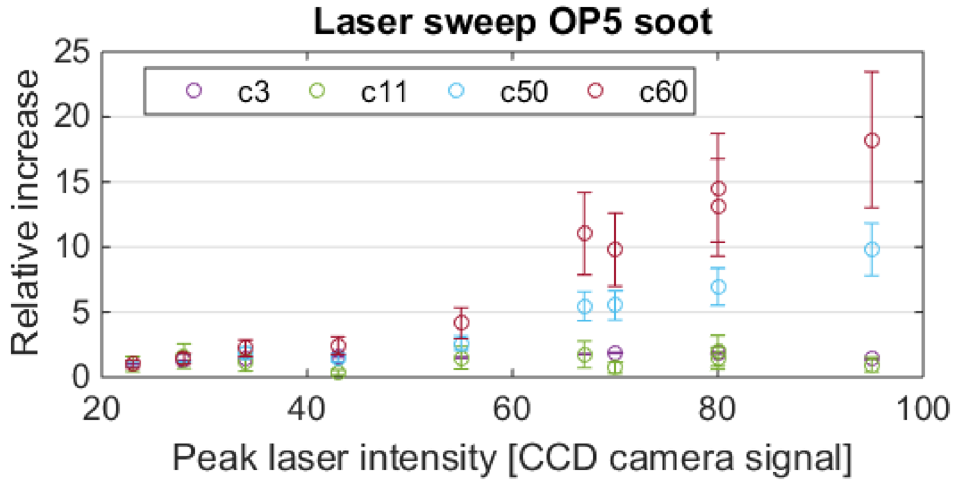
- Difficult to reproduce large carbon fragments in previous studies
- LU instrument: decent reproducibility over time
- LU instrument has relatively low laser power (low Refl. Mirror) -> low heating rate
- Still RIE 0.2-0.3 (last IE from RB I have is 550 ions/pg @ AB 7.E5 Hz)
- Mid-carbons and small fullerene carbons scales ~ linear with low C in laser power drops
- Large fullerenes increase exponentially with laser power -> secondary ionisation mechanism (Onasch et al. 2015) -> avoid high laser powers and large fullerenes

Comparison with TEM and OCEC



Effects of laser power on carbon ions



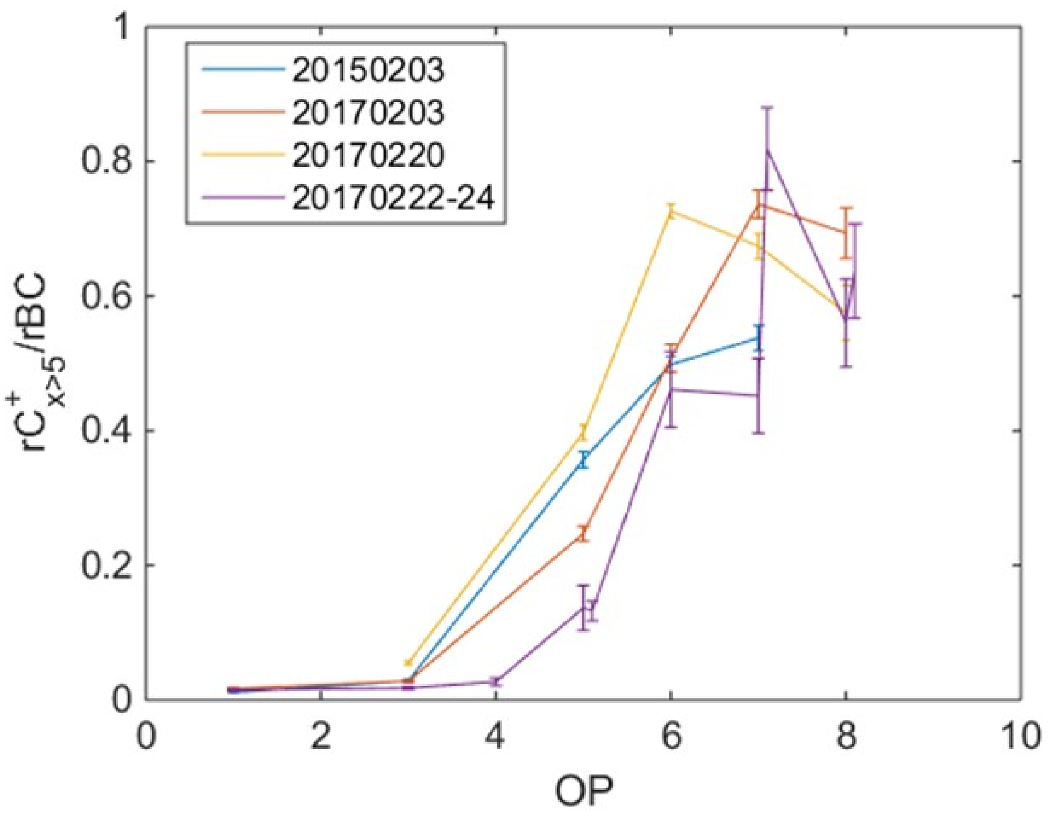


- Large fullerenes (>~C50) shows exponential increase with laser power
  - Avoid too high laser powers -> Thermal vaporisation of large fullC

Repeatability over time mini-CAST soot

Mature

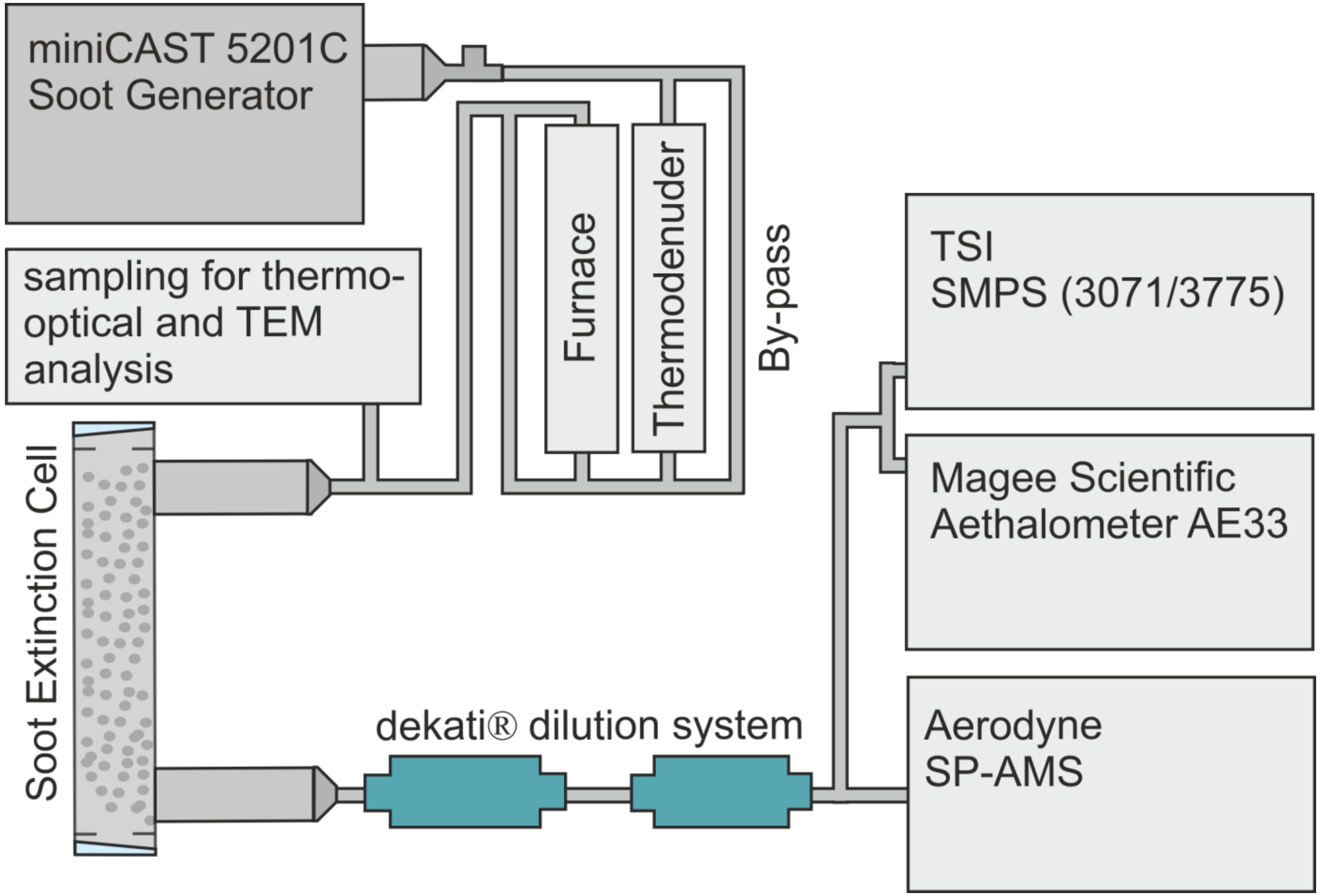
Immature



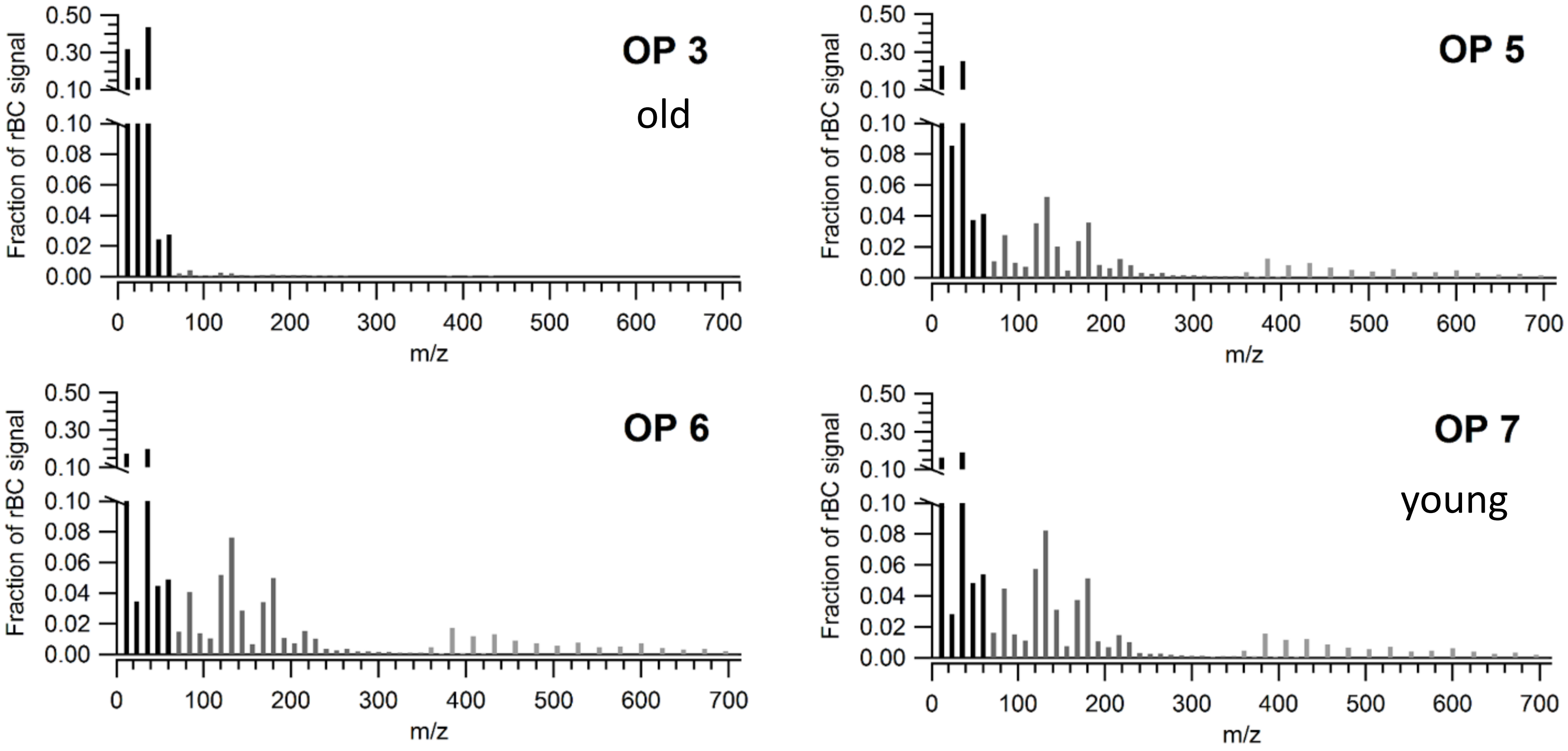
Set-up

Sandra Török et al. (2018): Investigation of the absorption Ångström exponent and its relation to physicochemical properties for mini-CAST soot,

Aerosol Science and Technology, DOI: 10.1080/02786826.2018.1457767

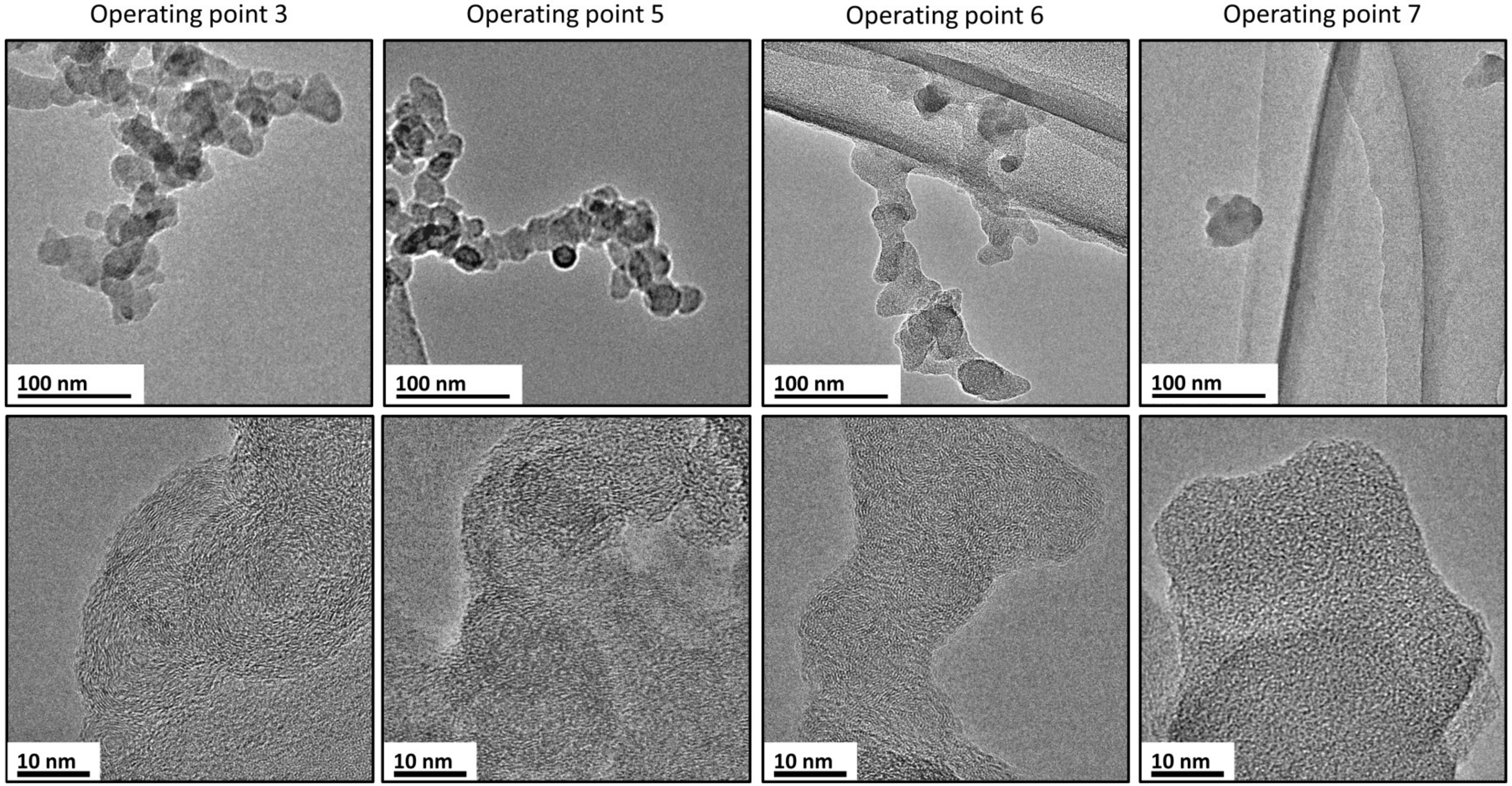


CAST operating point (OP) simulates soot marturity. (For some reason it starts at 7 and goes backwards)

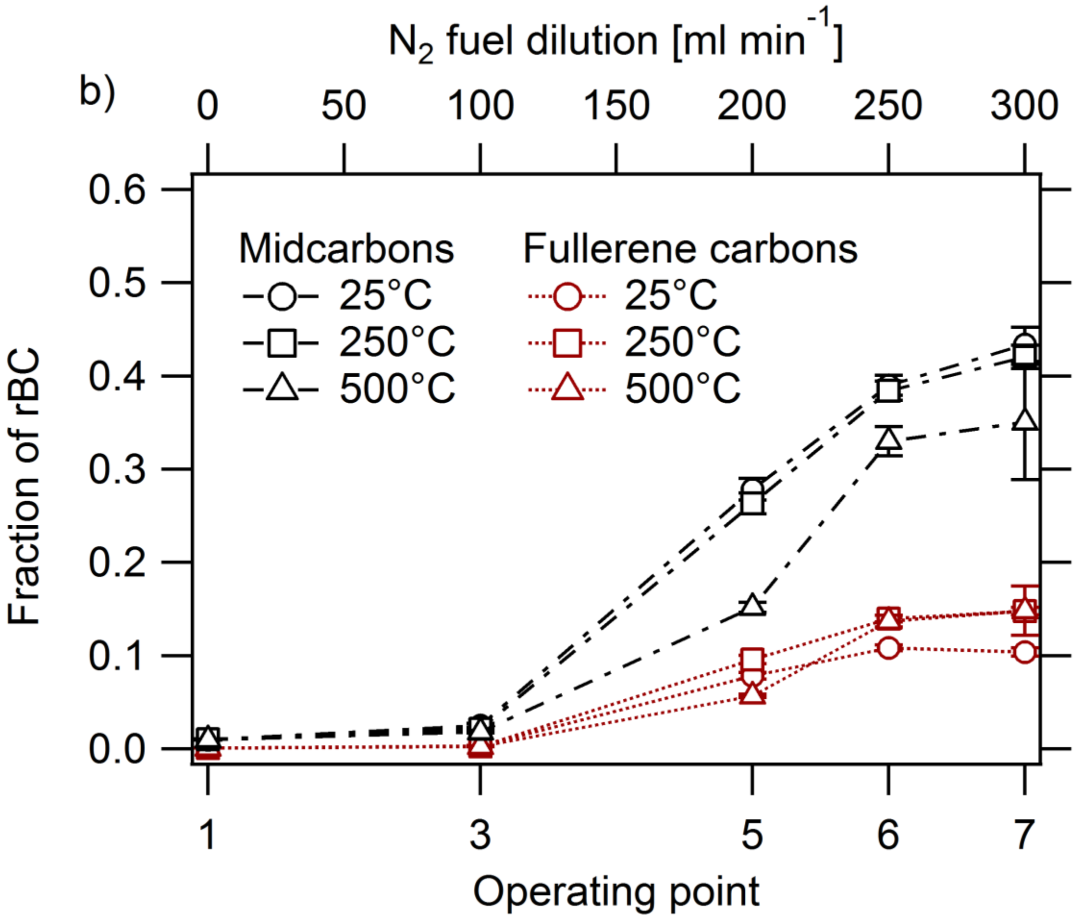


HR-TEM (Transmission Electron Microscopy)

Nanostructure evolves:

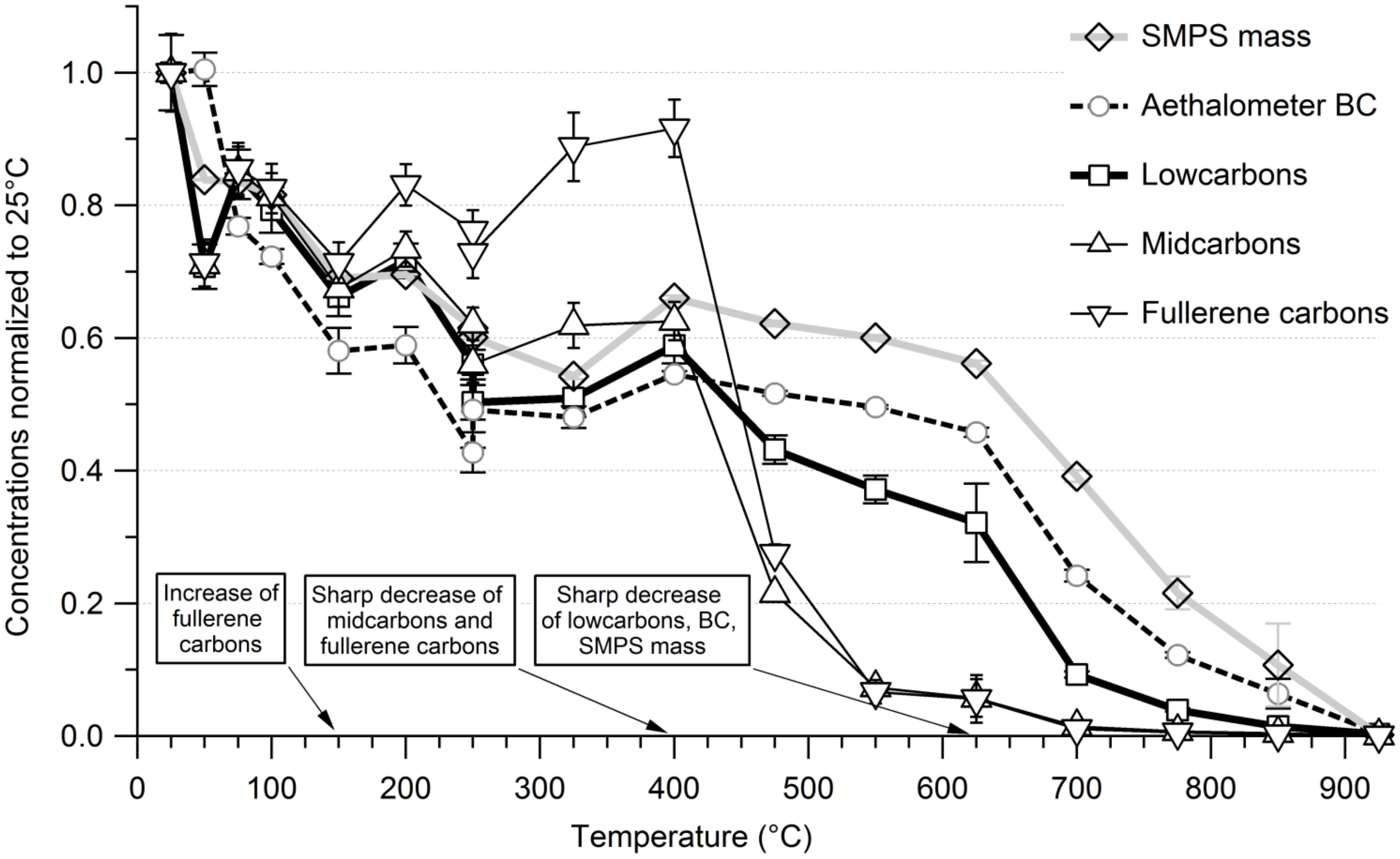


Heating changes the average soot nanostructure

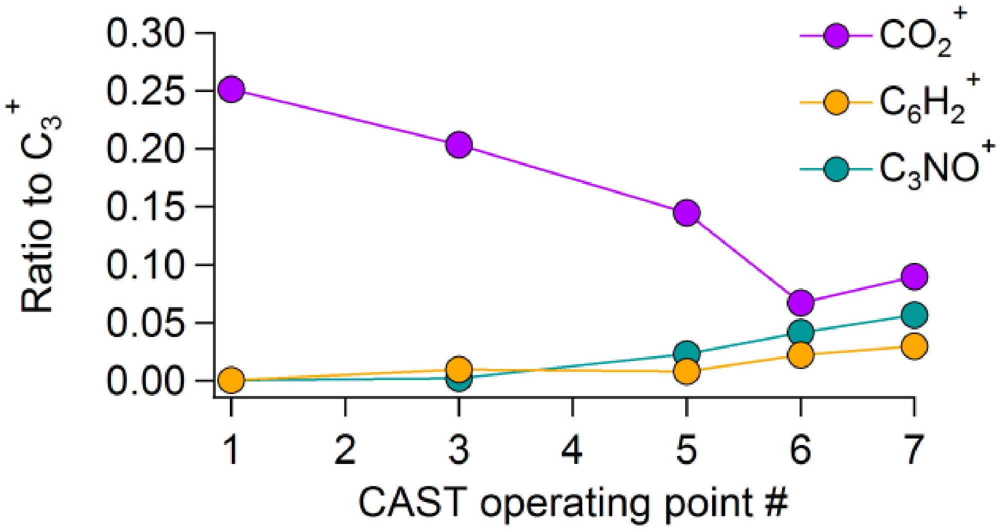


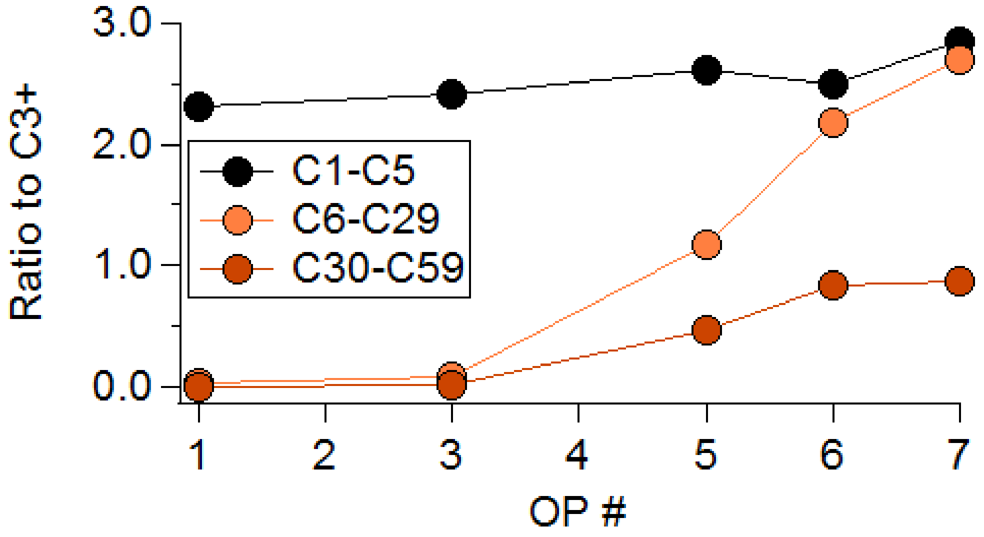
Heating changes the average soot nanostructure

evaporation and oxidation experiment at op 6 (young soot)

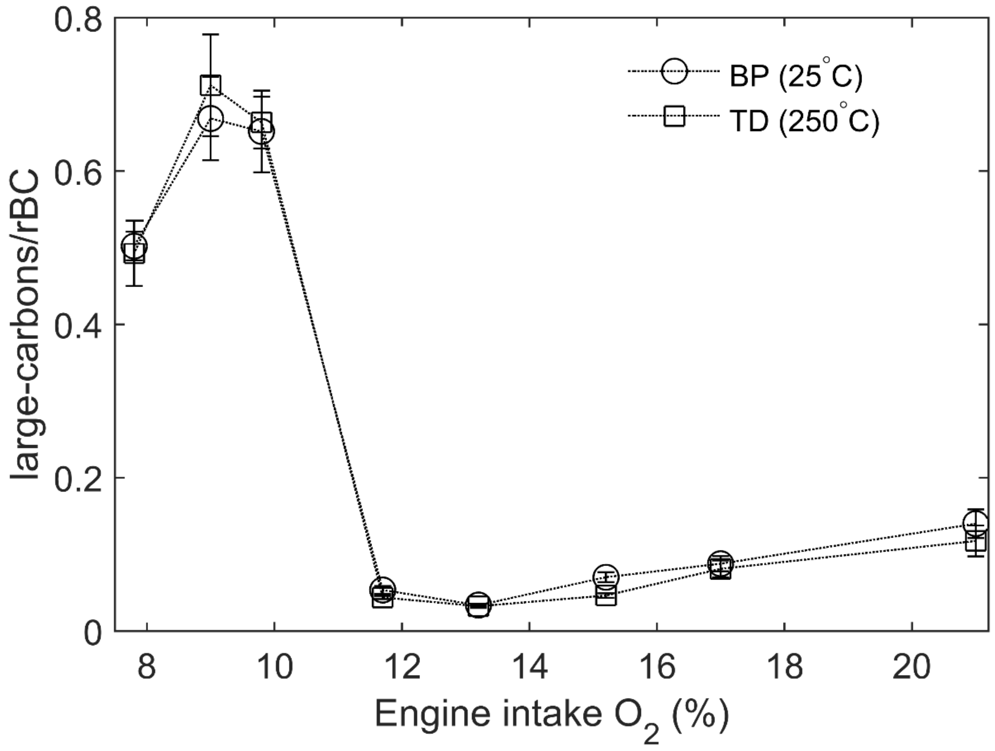


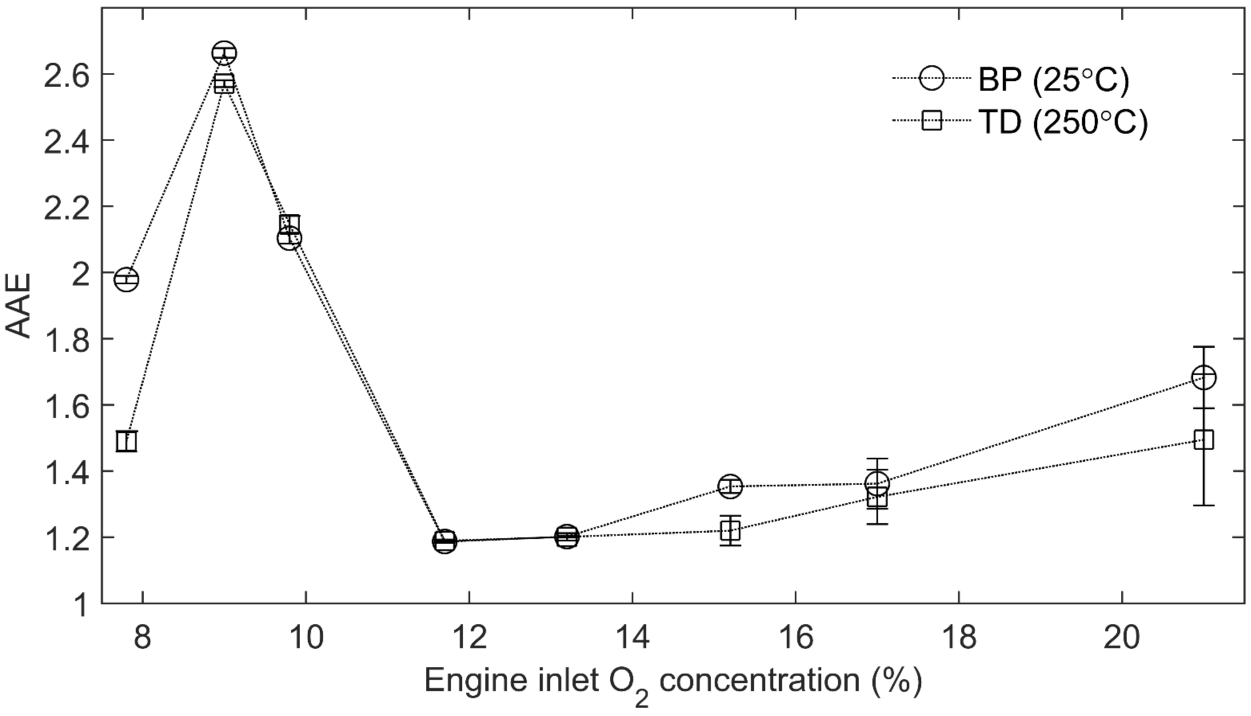
Connection to heteroatoms





Engine: effects of ”exhaust gas recirculation” (EGR) in biodesel combustion





Towards reproducibility and quantification

- Laser (overlap with particle beam, intensity)
- Ion source tuning
- Dual vs single vaporizers (+cylinder around ionization region)
- Nebulizable reference needed!

Summary

- Fringe length was inversely correlated ($r=-0.97$, $p=0.028$) to large carbon ions in SP-AMS mass spectra, which shows that we can use SP-AMS to measure carbon nanostructure.
- (lowcarbon ions ($C \leq 5+$) were correlated to elemental carbon ($r=0.98$, $p<10-8$))

- A warning: repodicibility between instruments is poor.

ESSAY

The importance of stupidity in scientific research

Martin A. Schwartz

Journal of Cell Science 2008 121: 1771 doi: 10.1242/jcs.033340

"Science makes me feel stupid too. It's just that I've gotten used to it. So used to it, in fact, that I actively seek out new opportunities to feel stupid. I wouldn't know what to do without that feeling."