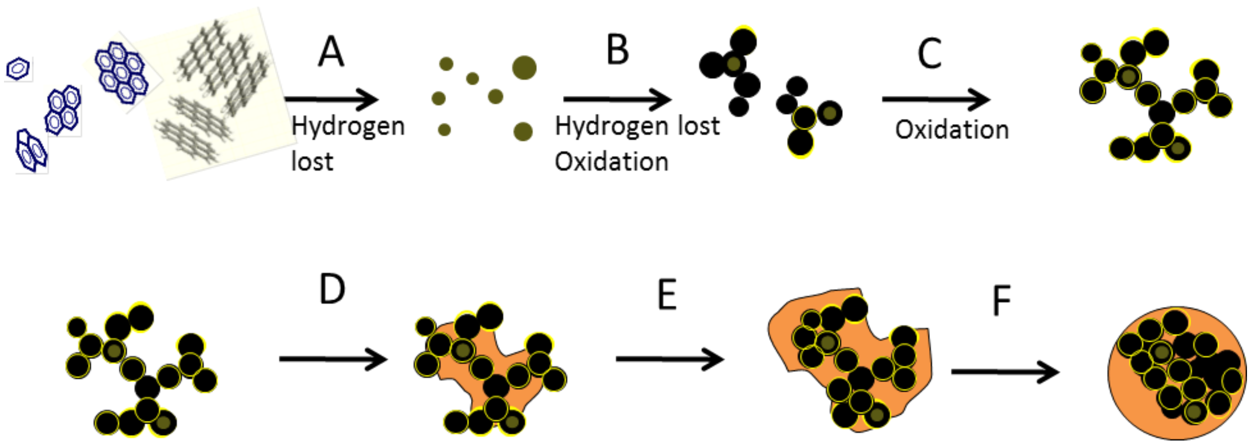
# Refractory constituents of soot probed with SP-AMS

Axel Eriksson et al.

Lund U

# Further reading

Phys-chem properties and atmospheric ageing of soot -growing up with an SP-AMS



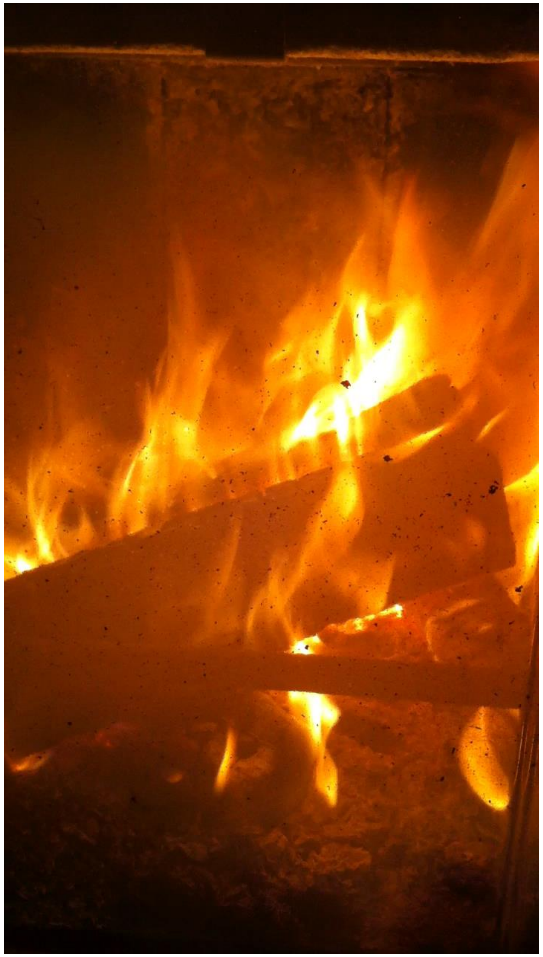

https://lucris.lub.lu.se/ws/files/6202971/5404278.pdf

# **Background**: soot formation and oxidation.

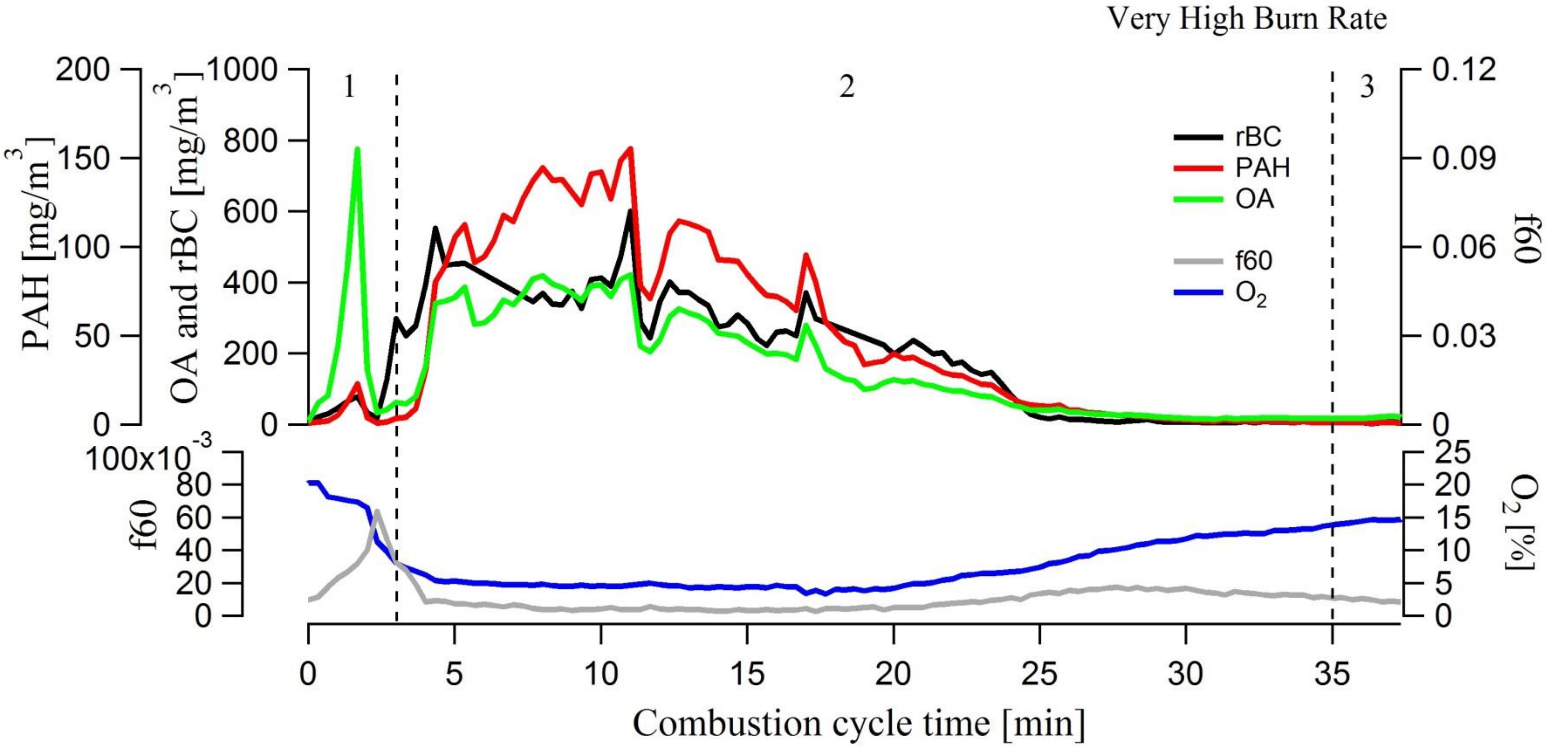
Wood smoke

Diesel exhaust

CAST burner soot



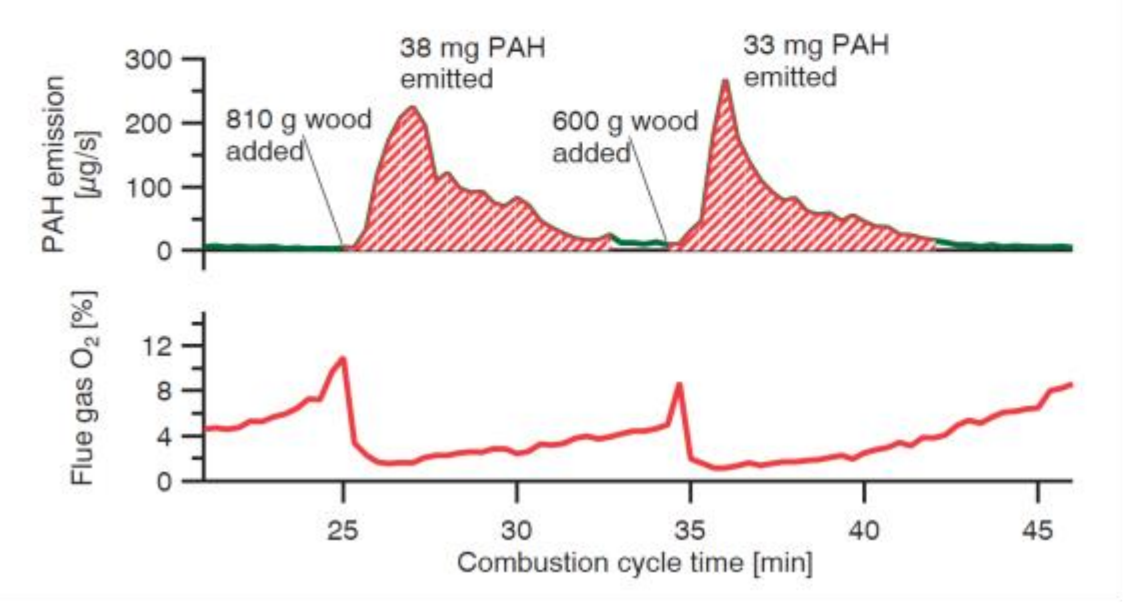
# The batch fired wood stove cycle:




Nielsen et al., submitted

# Wood stove emissions W-vaporizer

Adding more fuel mid-blaze:




(don’t do this at home)

# Wood stove emissions

# W-vaporizer vs W+L vaporizer

Large PAH (Eriksson et al, EST 2014) are not refractory

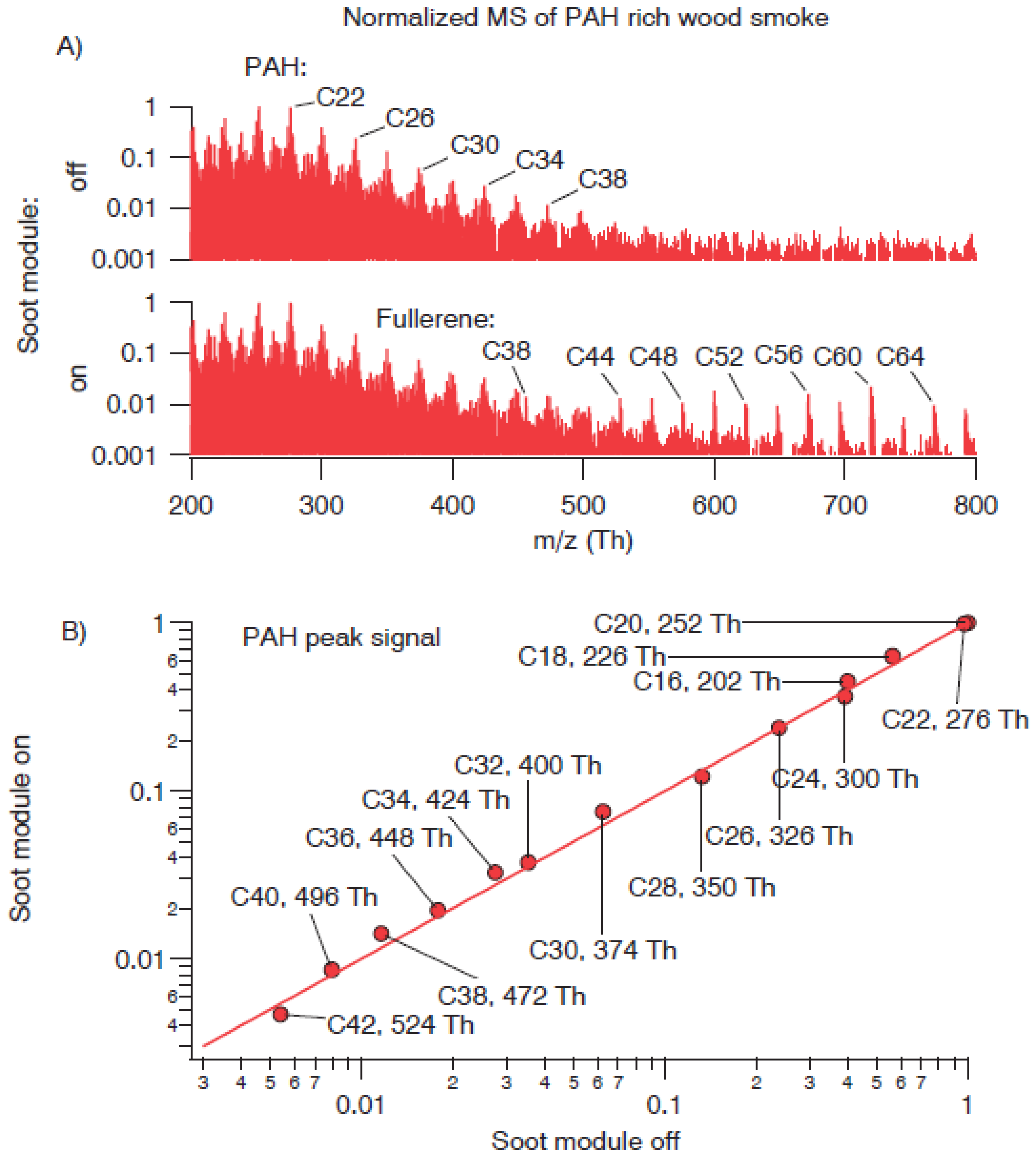
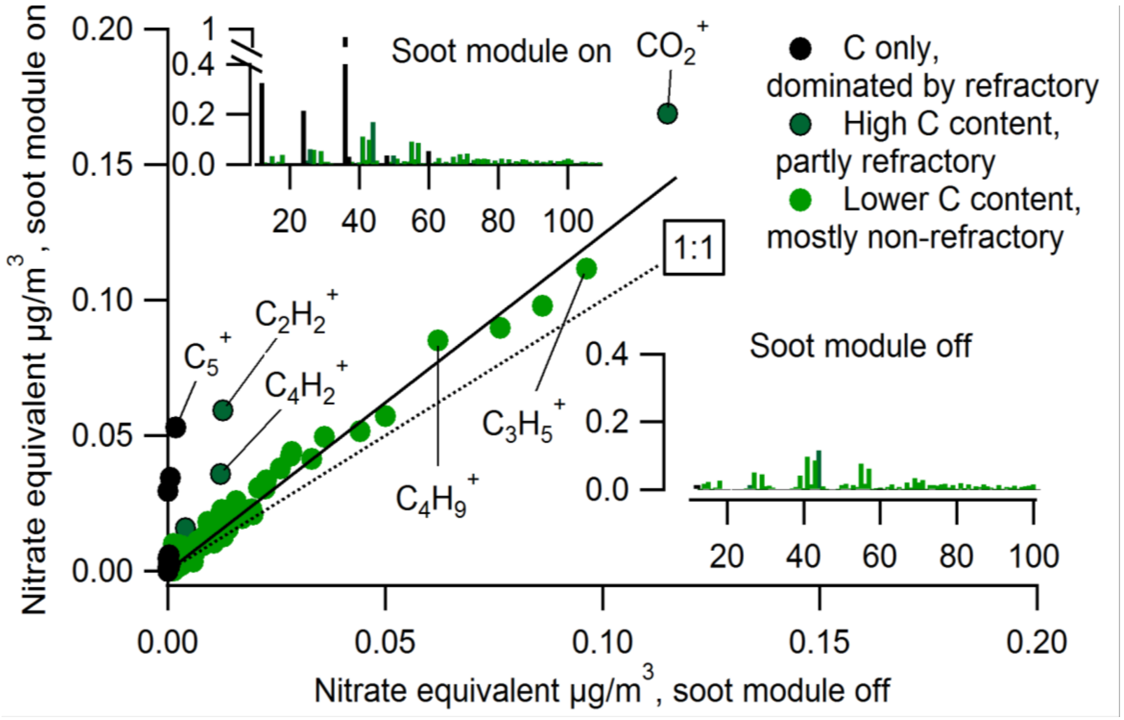



**Figure 6.** PAH-rich wood smoke measured with a dual-vaporizer Soot-Particle Aerosol Mass Spectrometer. A) Normalized mass spectra (m/z 200-800 Th) recorded with the soot module off (top) and on (bottom). B) scatter plot of normalized peak maximum intensities with and without the soot module engaged. Details in text.

# Signals from refractory wood smoke components



# The diesel engine cycle:

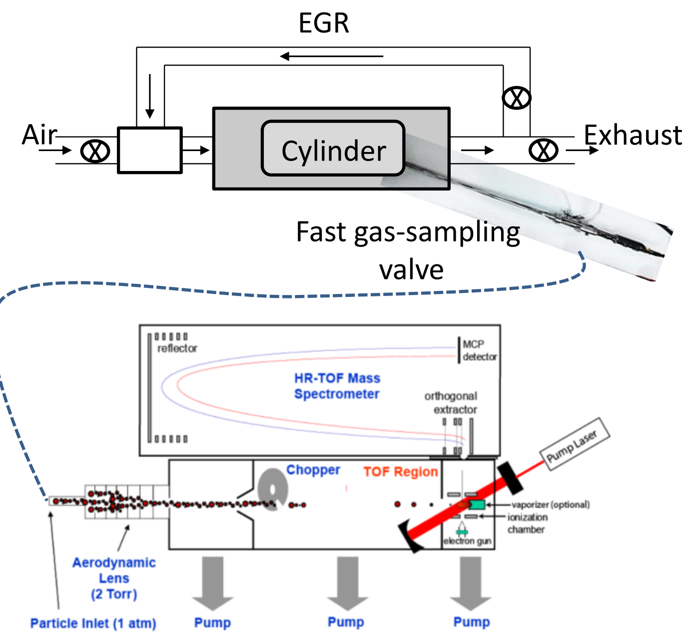
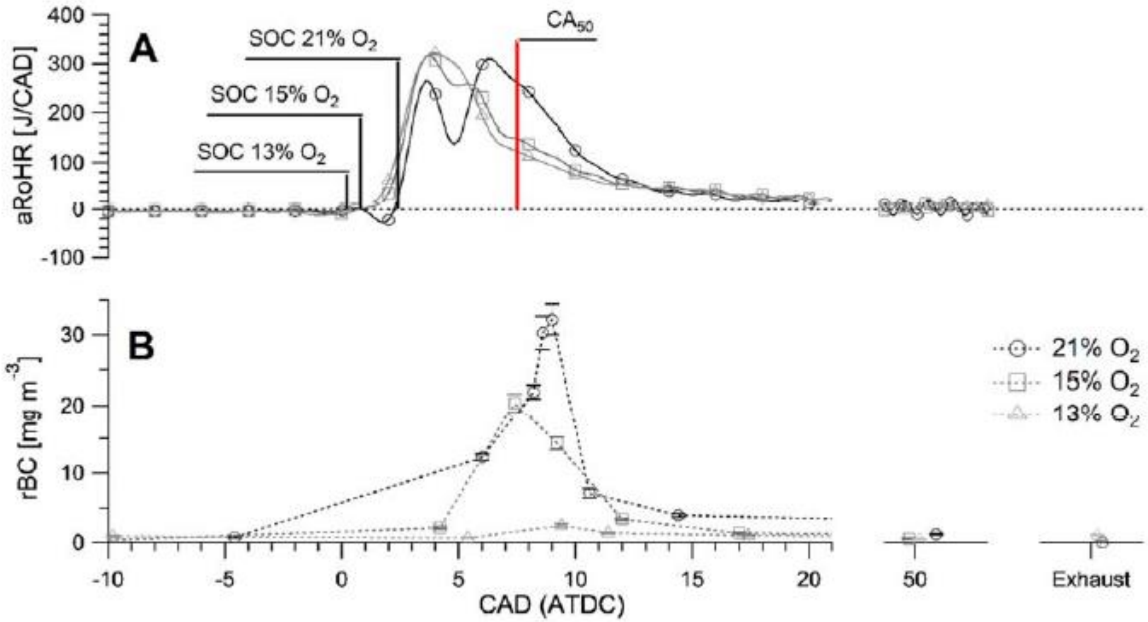
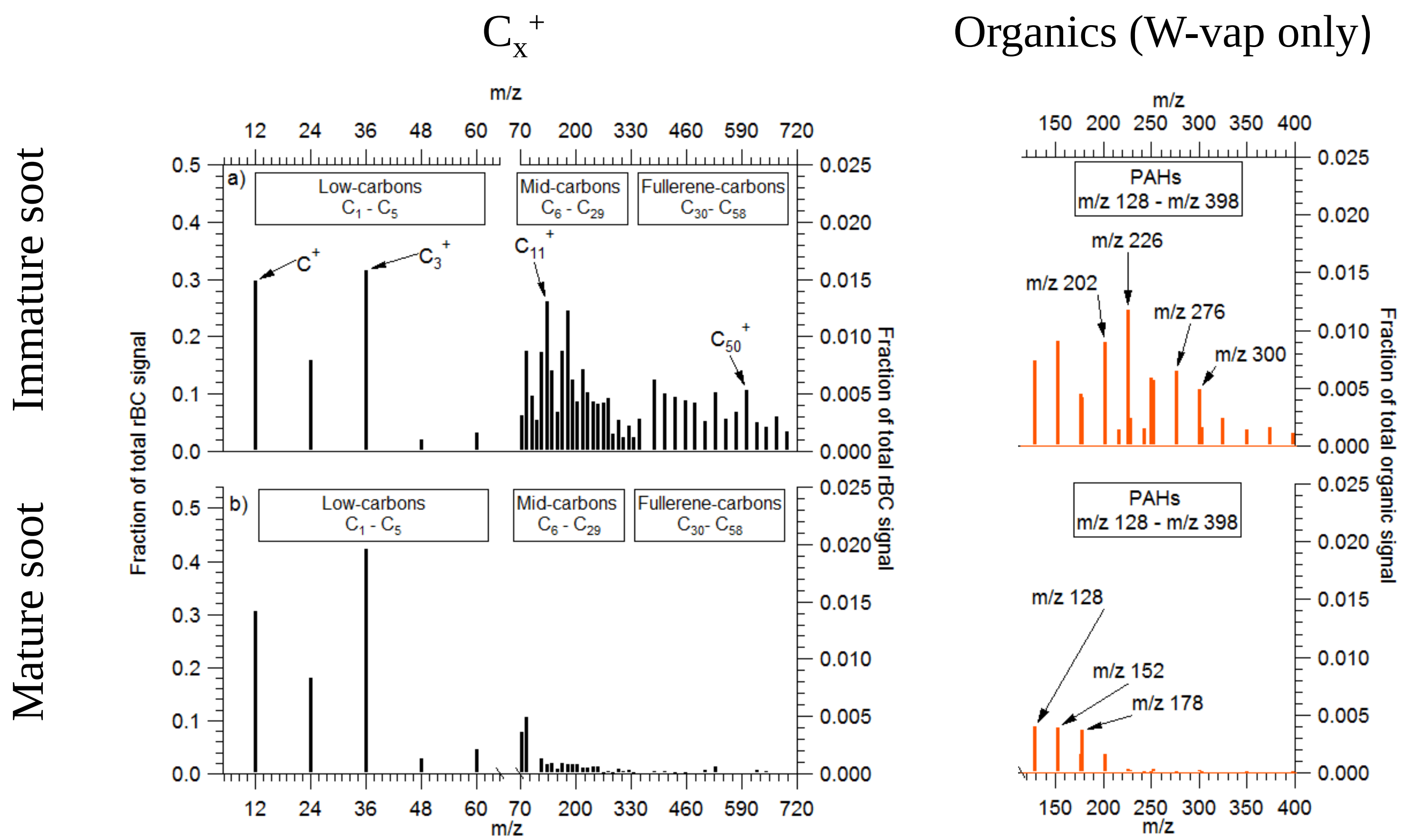



**Figure 1:** Set-up showing engine with EGR, the fast gas-sampling valve for particle sampling and the SP-AMS for particle characterization.



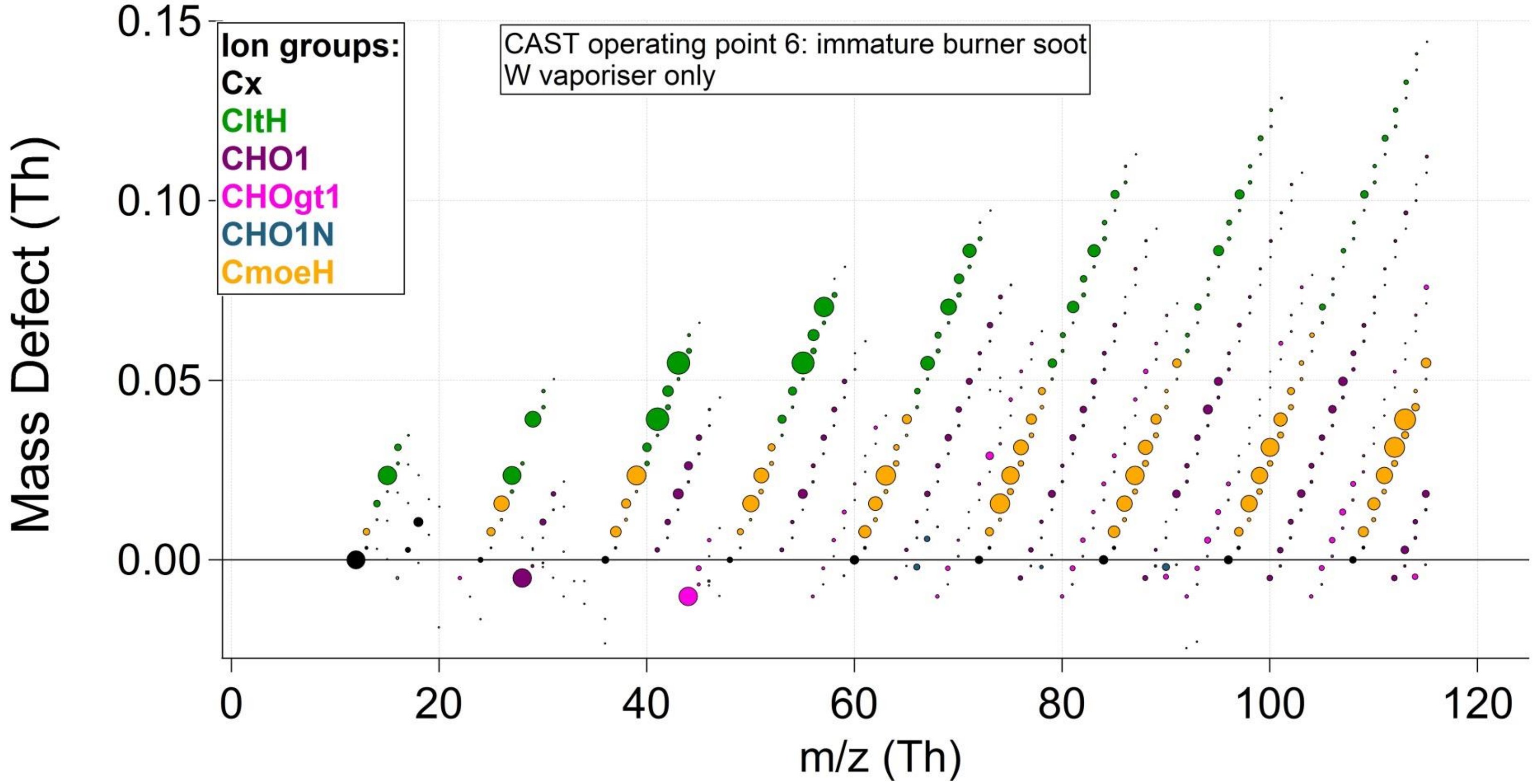

Malmborg et al., submitted

# Early and late cycle diesel soot

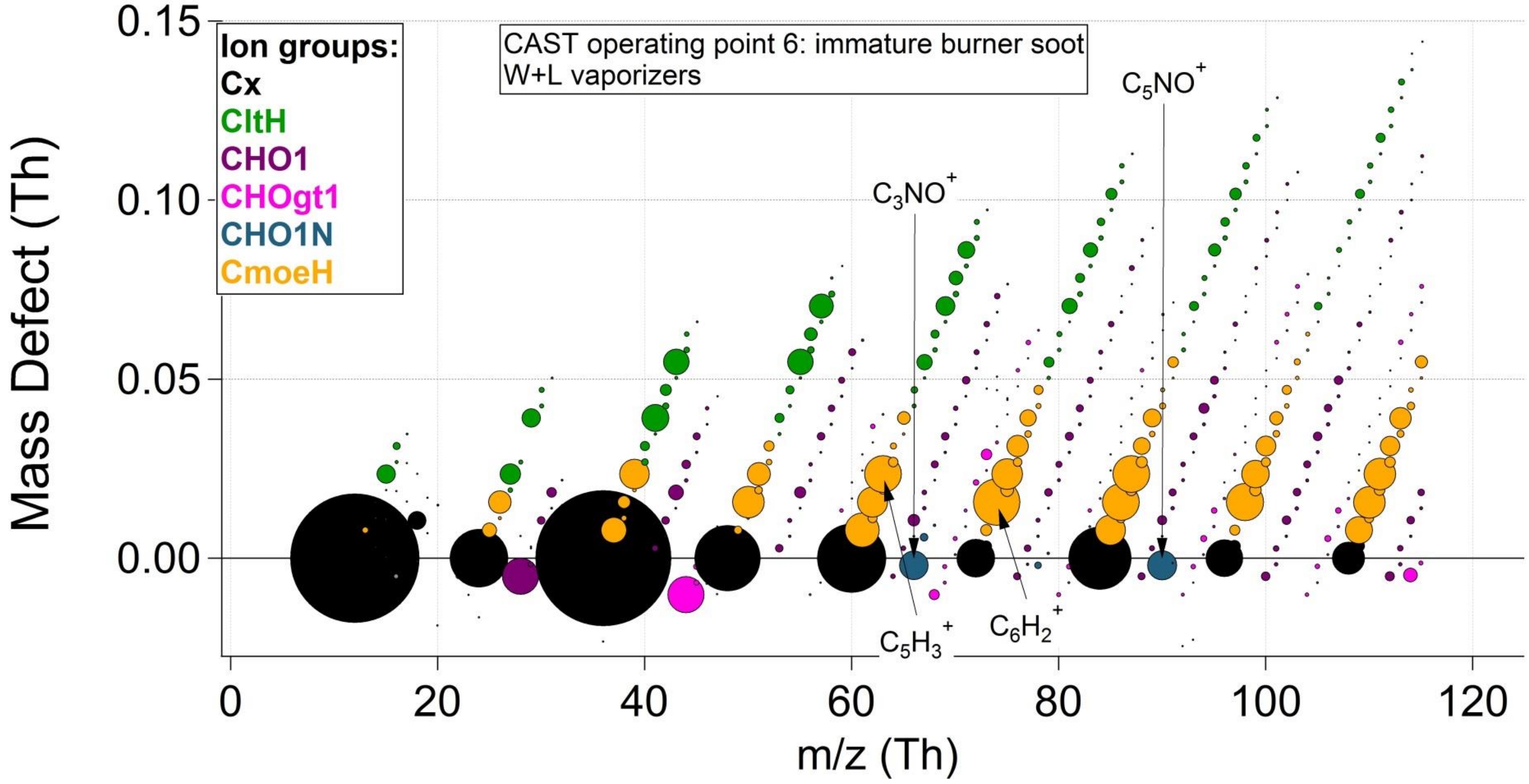



Malmborg et al., submitted

# CAST burner soot W



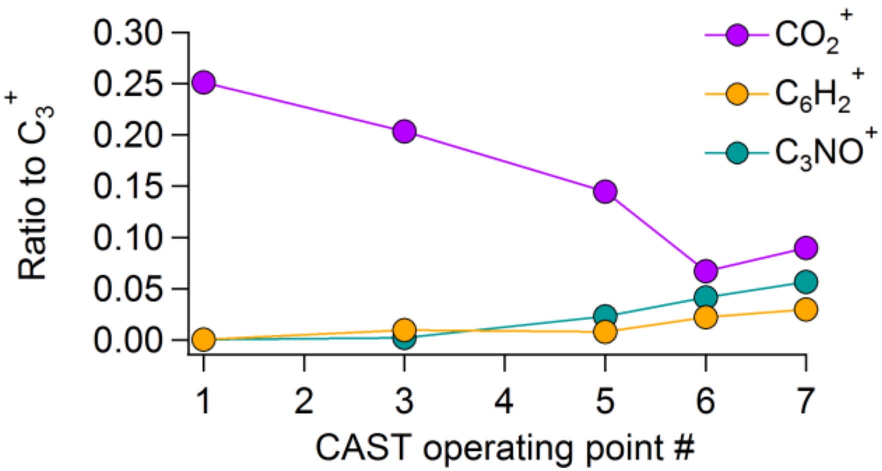
# CAST burner soot W+L

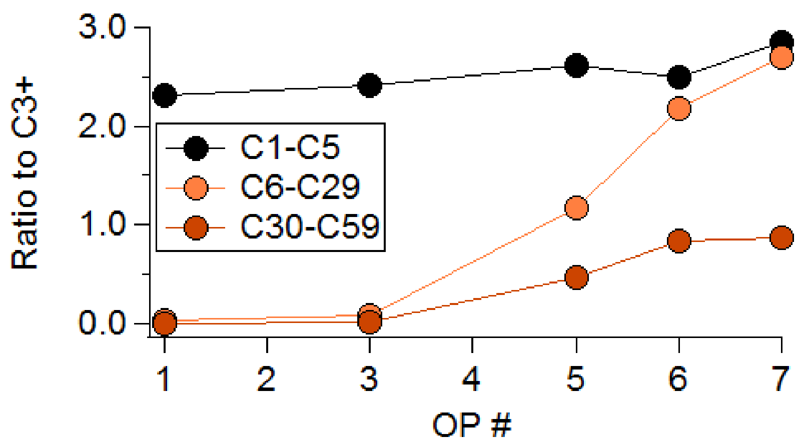


# CAST burner soot evolution

Soot maturity

Oxygen in
Hydrogen out



# Caveats

- Due to lack of reference method, the SP-AMS sensitivity towards different classes (e.g. oxygen and hydrogen containing refractory components) is not known. As shown on earlier users meetings, instrument-to-instrument variability is high but trends are reproducible.
- Mass spectral interferences from $N_2^+$ often preclude the detection of $rCO^+$, which has been the main ion of mixed elemental composition when quantifiable. In lab studies this can be mitigated using high loadings or Ar dilution.
- The mode of vaporization affects the fragmentation of the vapors, which complicates the comparison between W+L and L. However, L gives larger organic fragments than W, thus $rCO_2^+$ etc. are masked, not inflated.
- Collection efficiency (and RIE?) is increased for W+L.

# Conclusions

- In addition to pure carbon ions, ions of mixed elemental composition were generated from the refractory soot materials studied:
- Carbon- and oxygen- containing ions($rC_xO_y^+$) were observed in all cases, with rCO+ as main ion.The origin is likely incomplete in-flame oxidation of soot. Therefore $rC_xO_y^+$ could help elucidate the mechanisms of soot oxidation.
- $rC_xH_y$+ ions were generated by wood smoke (chiefly $rC_2H_2^+$) as well as from the flame soot generator (e.g. $rC_6H_2^+$ and $rC_5H_3^+$). They likely originate from immature soot, where the loss of hydrogen from the precursors was incomplete. $rC_xH_y^+$ may prove helpful for understanding soot formation, which together with oxidation governs soot emissions.
- $rC_3NO^+$ and $rC_5NO^+$ were generated by the less mature burner soot.
- For the real-world sources studied, $rC_xO_y$+ has been more abundant in the mass spectra than $rC_xH_y^+$, which suggests oxygen is more abundant than hydrogen (mass wise) when the soot is large enough to be detected efficiently.
- As nitrogen was not found in the wood smoke or diesel exhaust, it may be very transient and/or possibly rare in real world soot.

# Outlook

O, H and possibly N containing ions, as well as the distribution of carbon clusters from refractory soot constituents could help:

Elucidate soot formation and oxidation (destruction)

Characterize soot from different sources, possibly with different impacts (chemistry in the lung and atmosphere, optical properties, susceptibility to exhaust treatment.)