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Transient formation of organic gaseous emissions in residential wood combustion with and without an electrostatic precipitator

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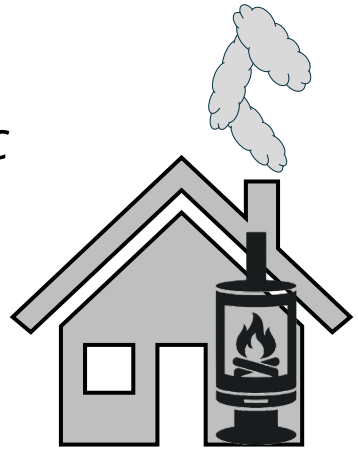
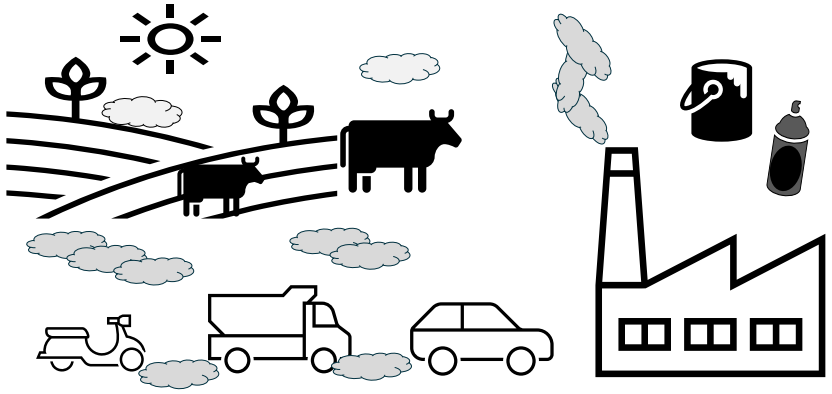
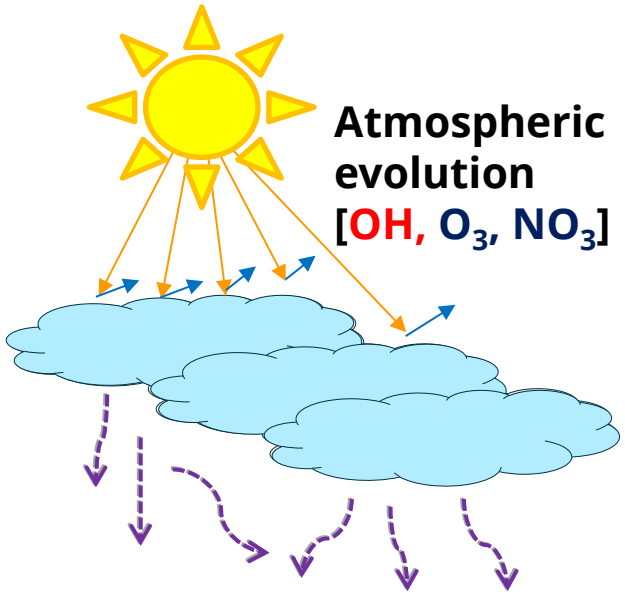
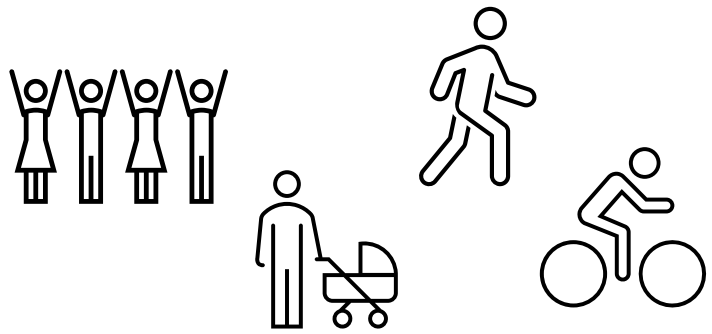
Wood combustion is an important source of volatile organic compounds emissions (VOCs)

In Europe, ~15 % of anthropogenic VOCs from residential heating









→ **Formation of secondary particles and ground-level ozone**

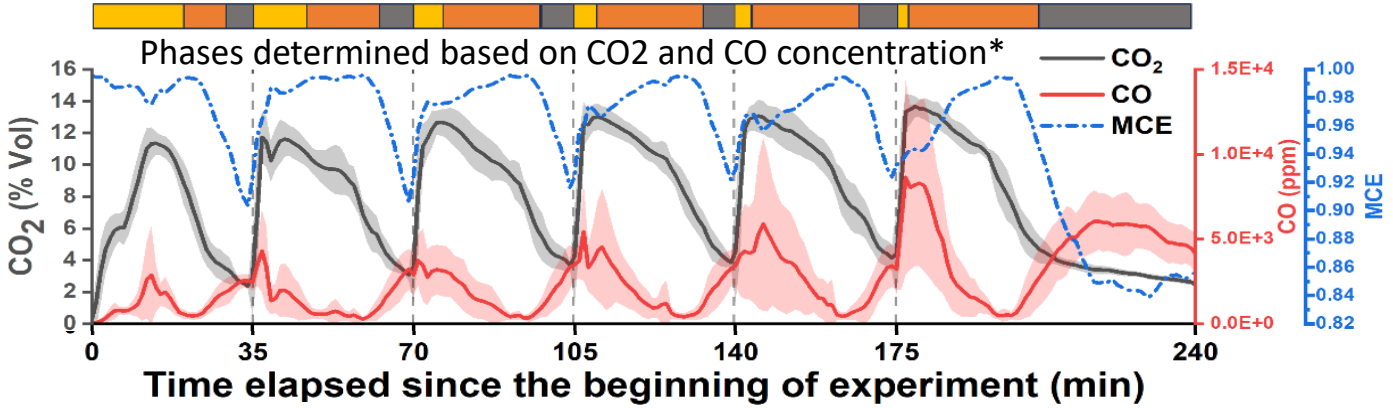
→ **Exposure resulting in adverse health outcomes**
e.g., PAHs emitted from incomplete combustion

Batchwise beech log combustion in a modern chimney stove

IGNITION **FLAMING** **CHAR BURNING**

+ Particle emission reduction by an electrostatic precipitator (ESP; OekoTube-Inside)

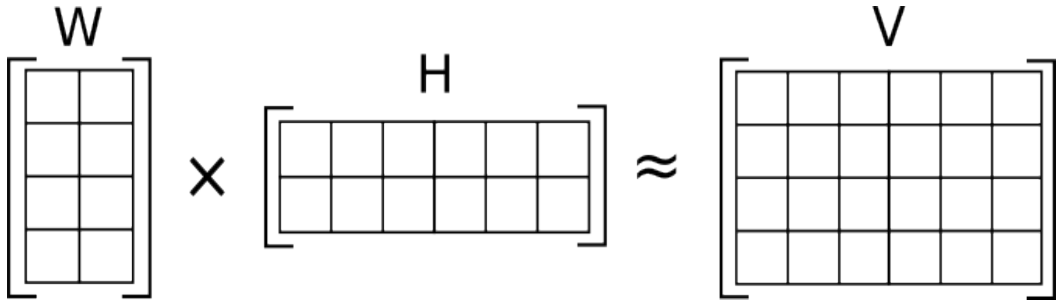
N = 4 × 4 h (6 batches) for both without and with ESP



* Mukherjee, A., et al.: Black carbon and particle lung-deposited surface area in residential wood combustion emissions: Effects of an electrostatic precipitator and photochemical aging. STOTEN. 2024. DOI: 10.1016/j.scitotenv.2024.175840

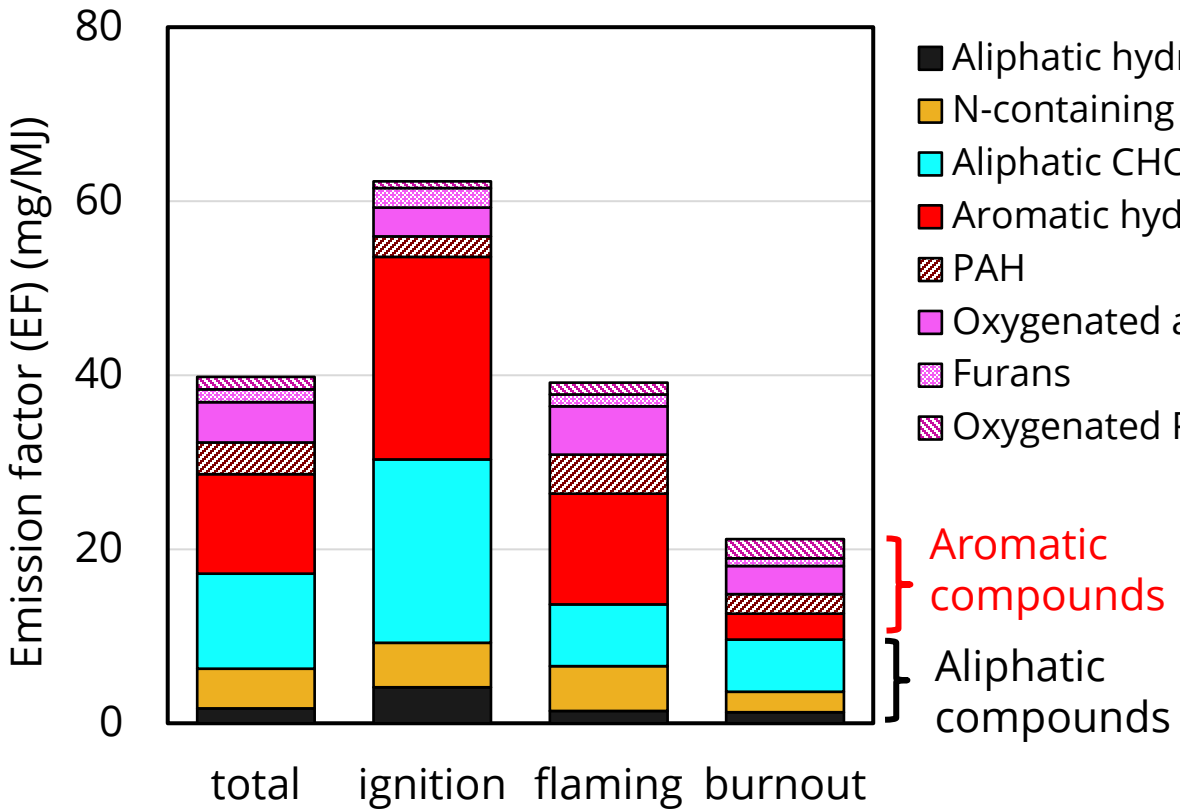
Online measurement of organic gaseous emissions

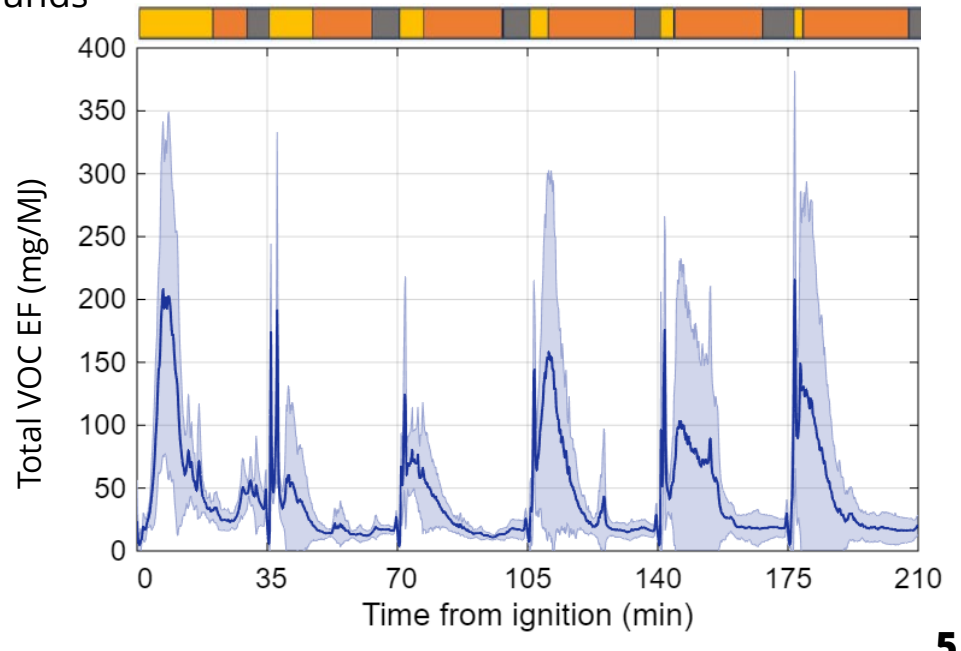
- Volatile organics were monitored by a **proton transfer reactor - time of flight - mass spectrometer** (PTR-ToF 8000, Ionicon) at dilution of 1:60
  - 10 sec time resolution
  - 109 ions identified and classified based on molecular formula
- Non-negative matrix factorization* of the online mass spectra by matlab *nmf()*
  - Factor timeseries × factor scores (contribution of different ions to each factor) ≈ original matrix



Hartikainen A. et al.: Resolving emission factors and formation pathways of organic gaseous compounds from residential combustion of European brown coal. Combustion and Flame. 2024. DOI: 10.1016/j.combustflame.2024.113485

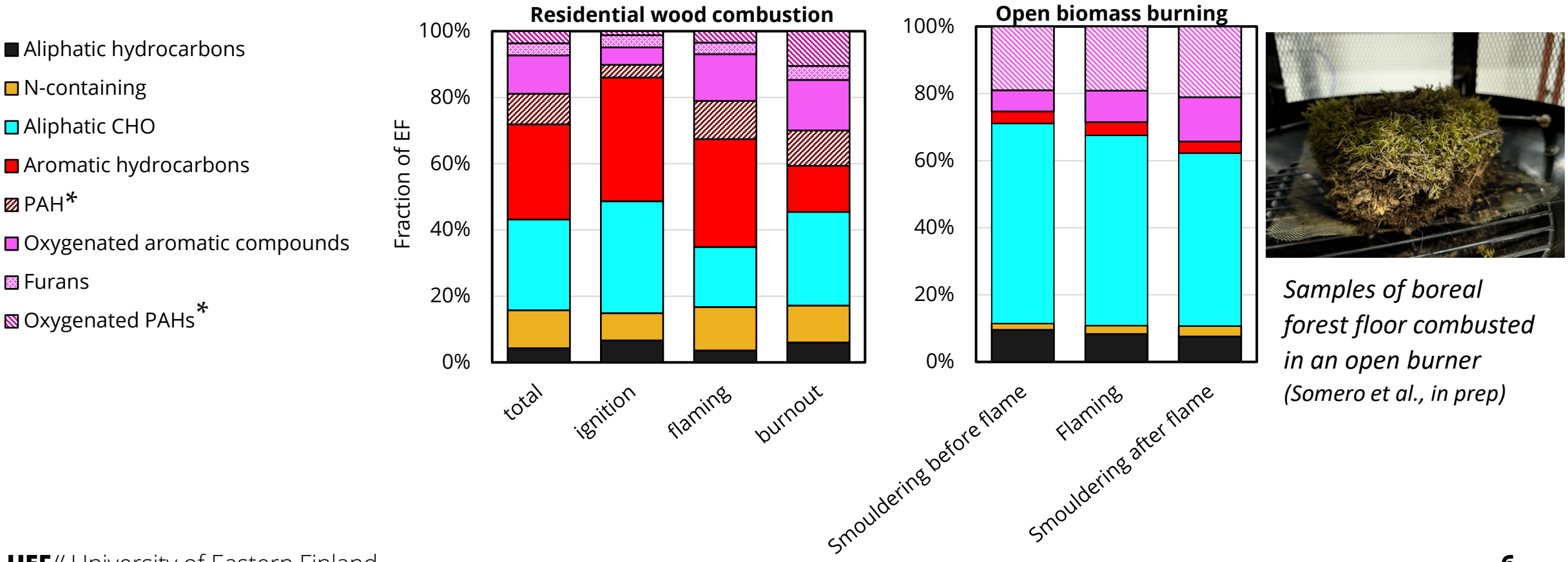
Half of the organic gaseous emissions aromatic





*With sufficient proton affinity →
e.g., small alkanes not included

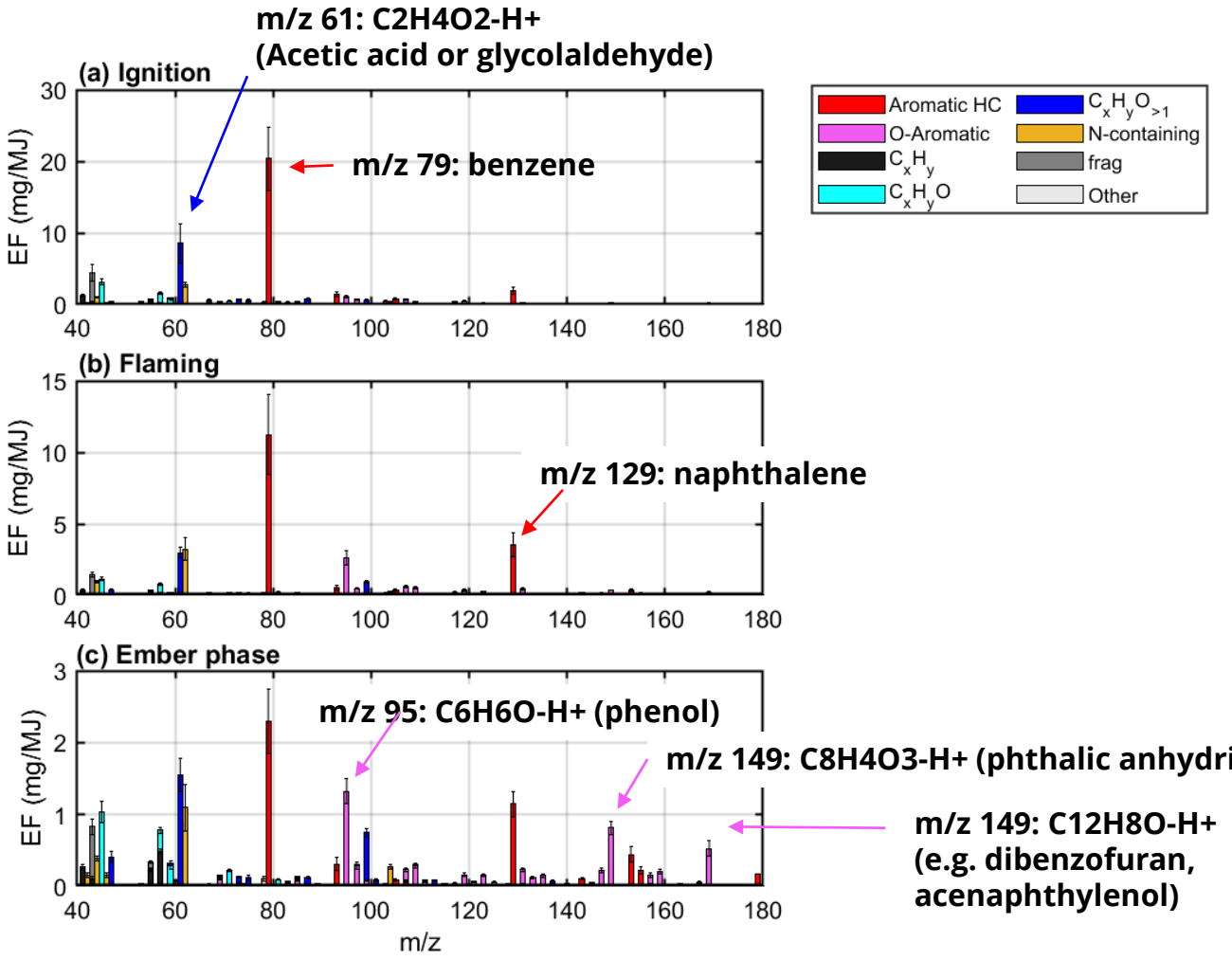
High efficiency combustion → more aromatic hydrocarbons, less oxygenated compounds than from open biomass burning

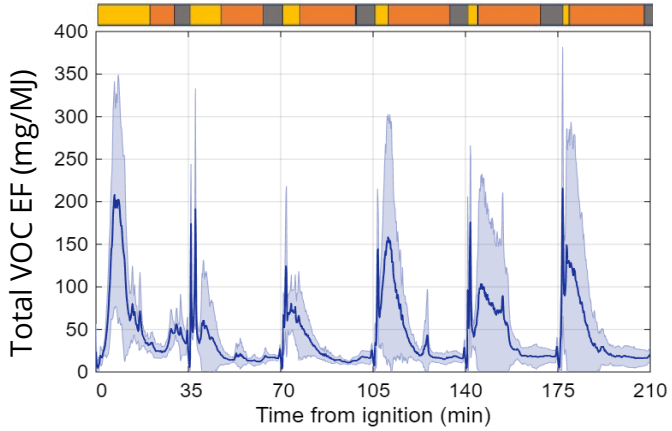


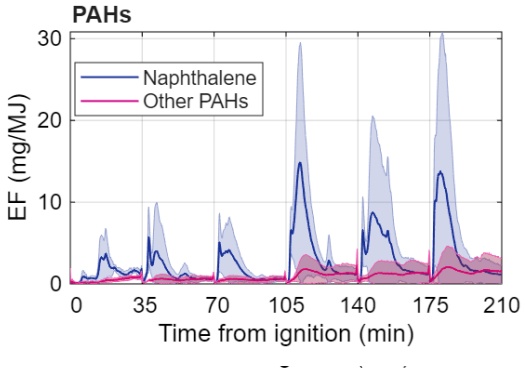
Samples of boreal forest floor combusted in an open burner
(Somero et al., in prep)

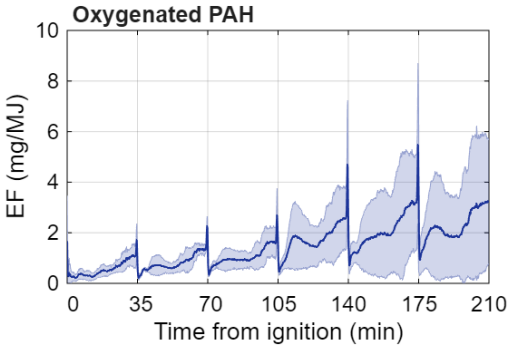
*not separated from other aromatics/O-aromatics for boreal forest floor combustion

Different combustion phase, different VOC profile

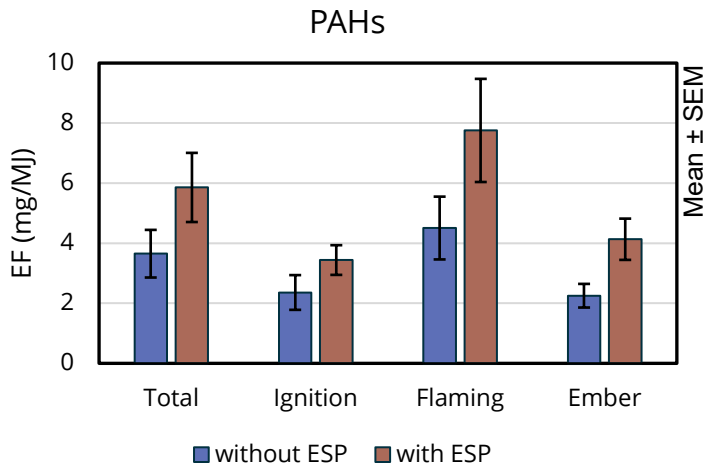


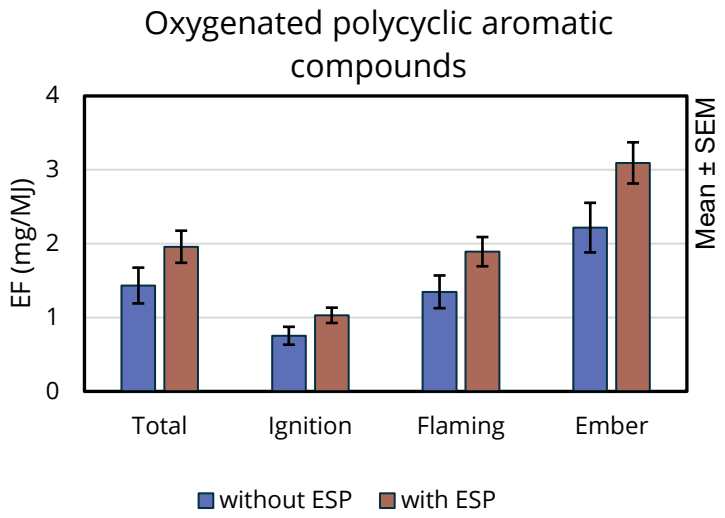


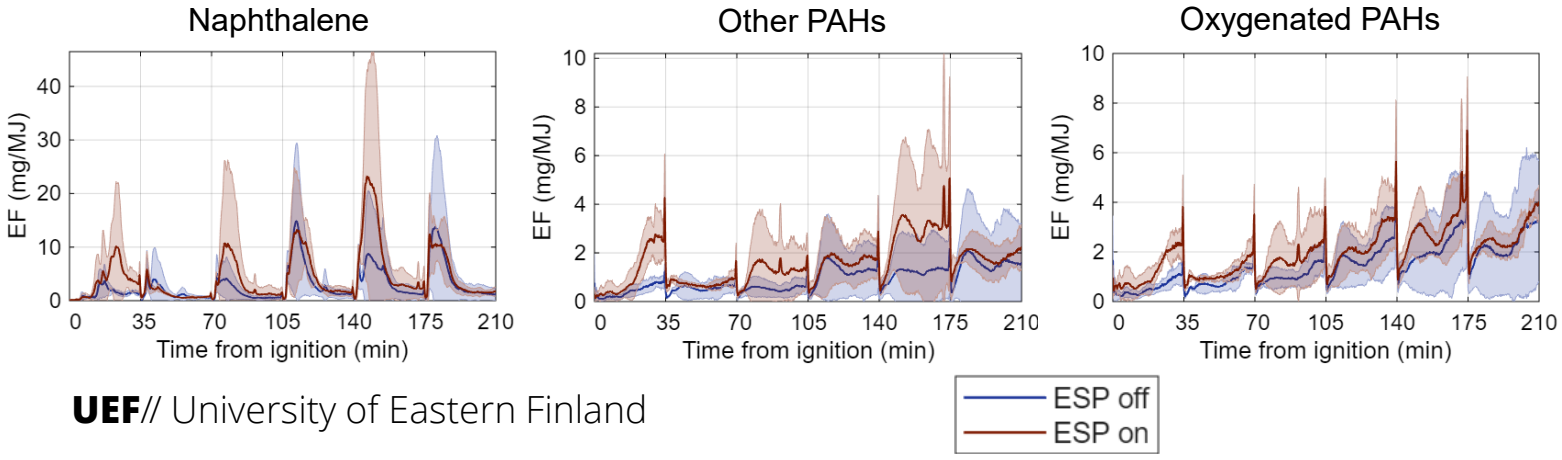




ESP increased the emissions of a few gaseous species, mainly polycyclic compounds





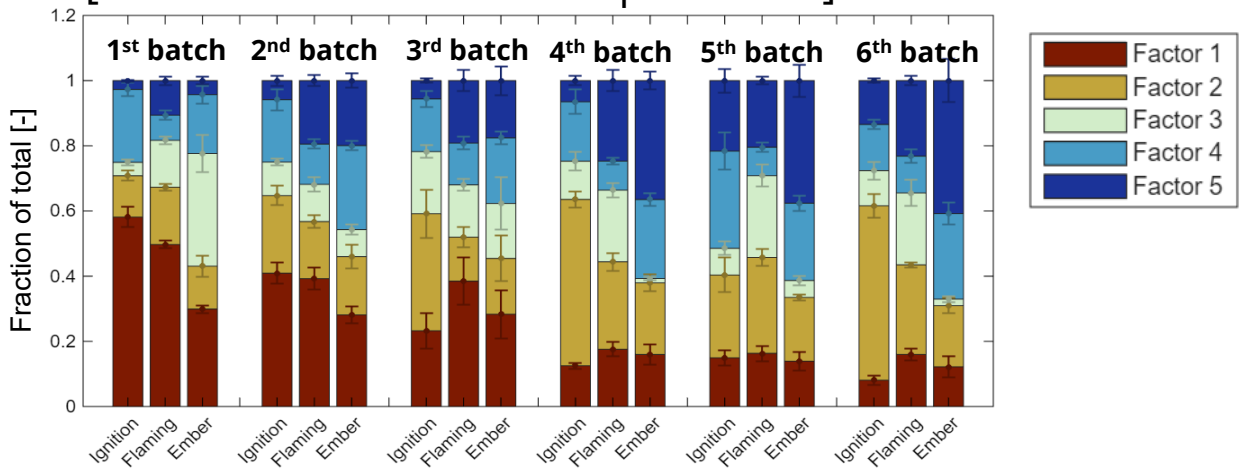


- Overall similar emission contents with or without ESP
- No major differences in the mass spectra of organic particles either (UHR-FTICR-MS; *Schneider et al., 2024**)
- ESP → higher number of oxygenated compounds detected at low intensities
- Statistically significantly higher EFs for polycyclic species at every phase

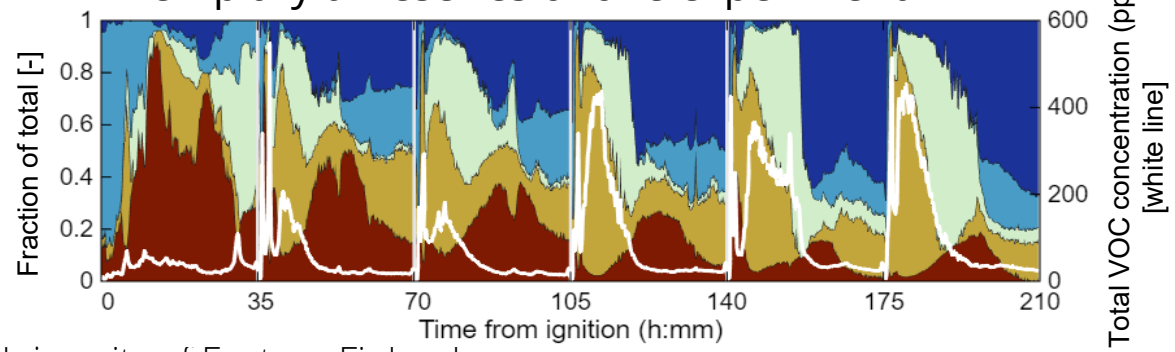
*Schneider E., et al.: Molecular composition of fresh and aged aerosols from residential wood combustion and gasoline car with modern emission mitigation technology. Environmental Science: Processes & Impacts. 2024. DOI: 10.1039/d4em00106k

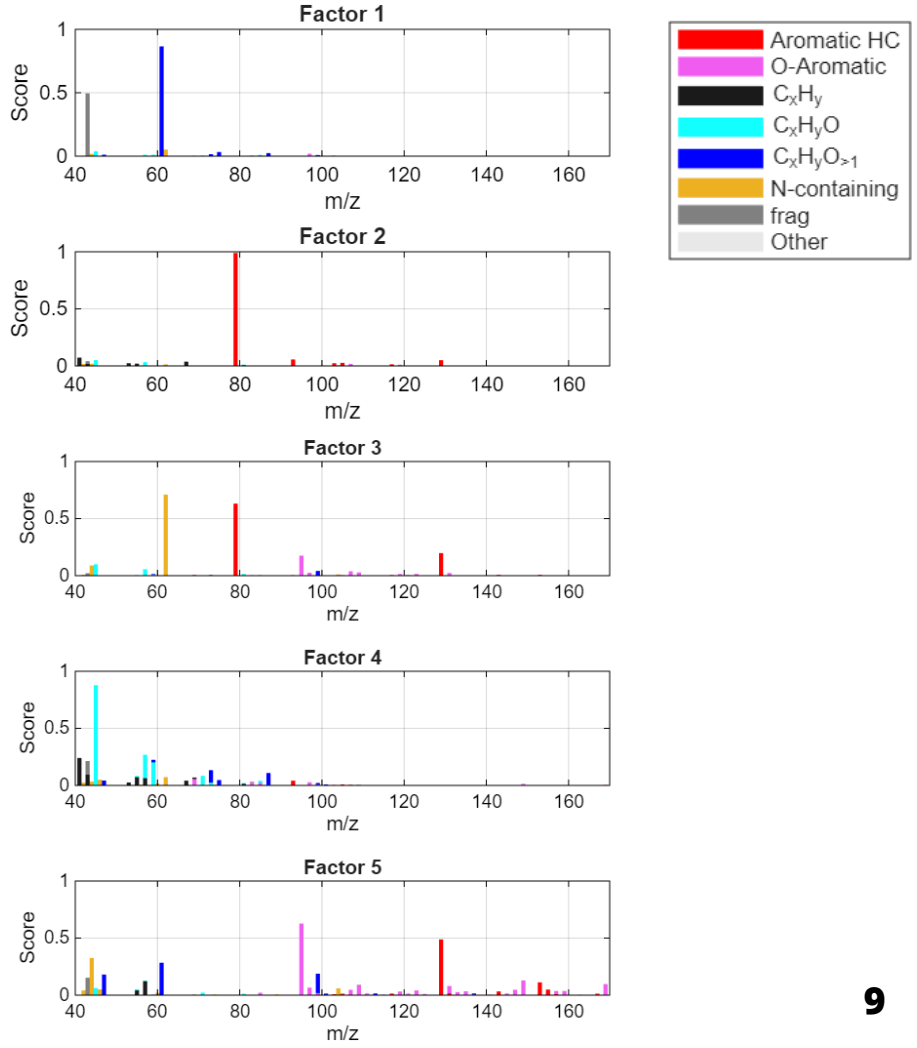
Five distinct factors revealed by factor analysis of spectra timeseries

[mean ± SEM for no-ESP experiments]

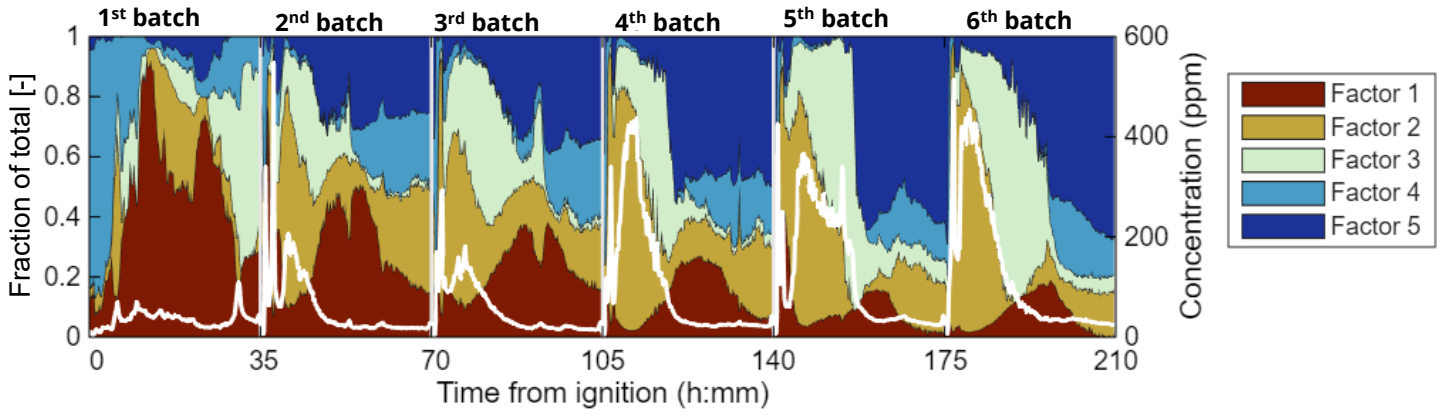


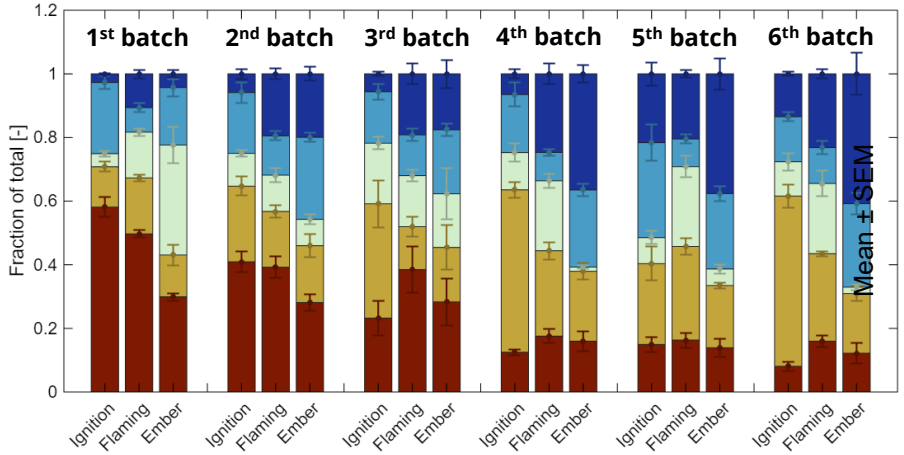
Exemplary timeseries of one experiment

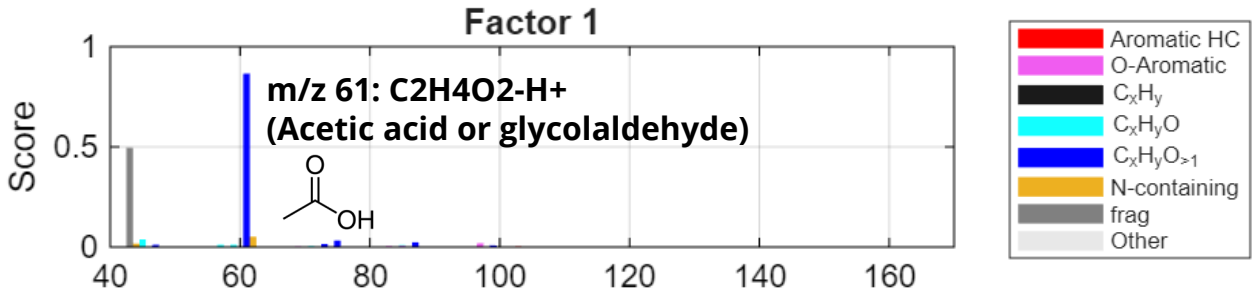




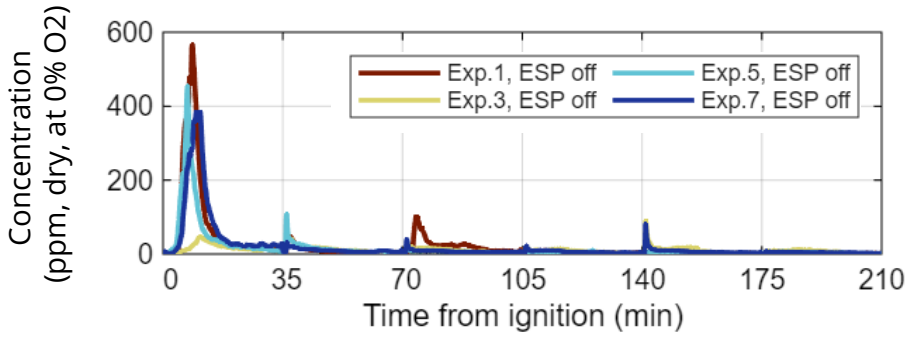
Factor 1: primary devolatilization of the fuel



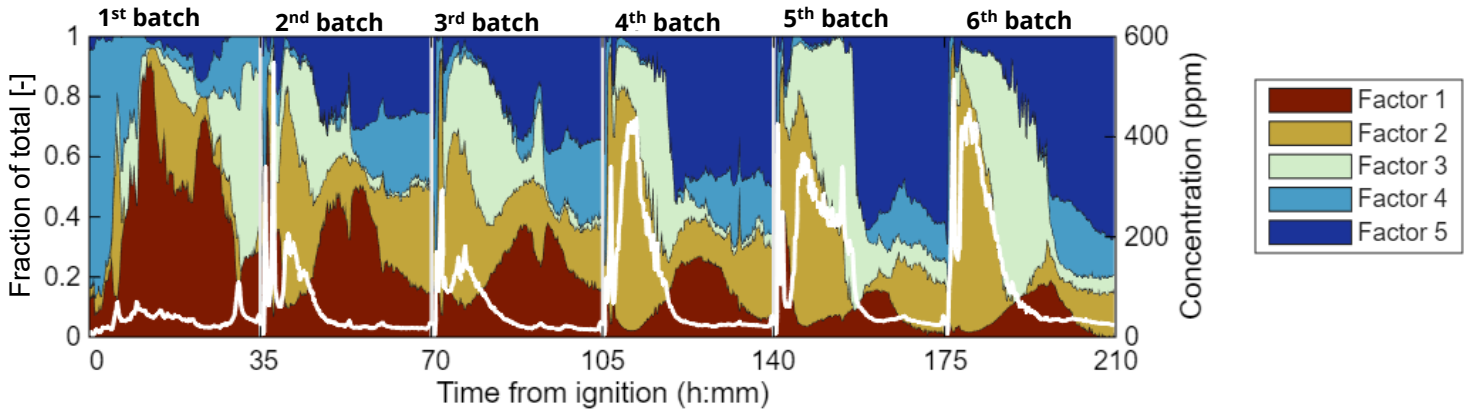


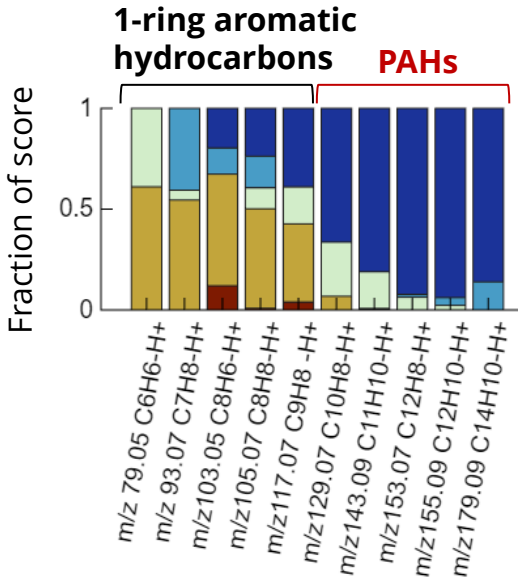


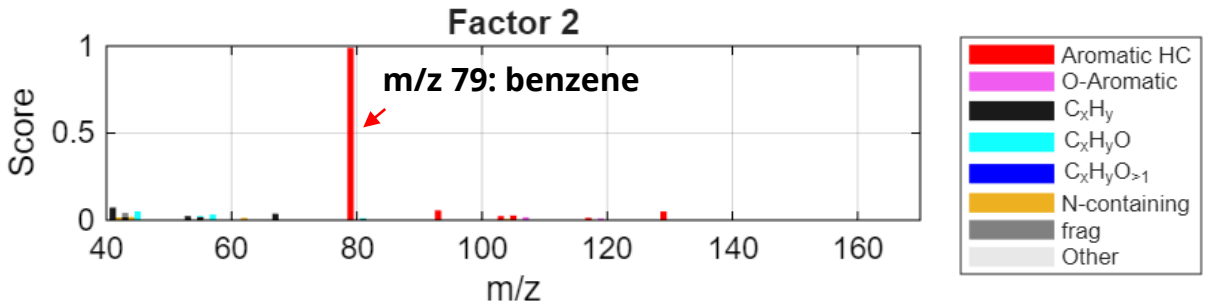
m/z 43: C2H2O-H+; fragment associated with acetic acid or other oxygenated compounds



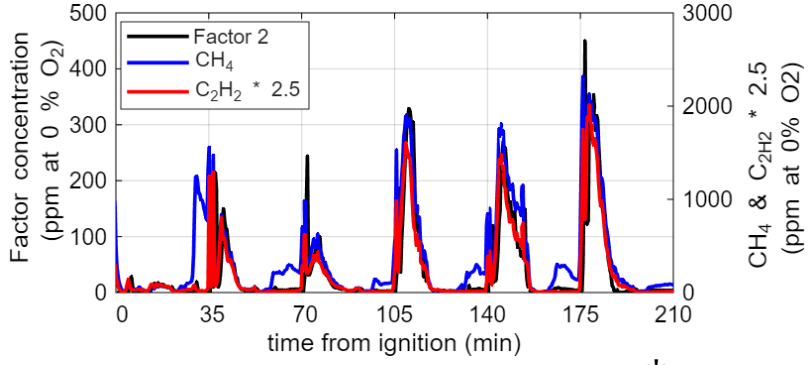
Factor 2: direct pyrolysis products of cyclopentadiene intermediates





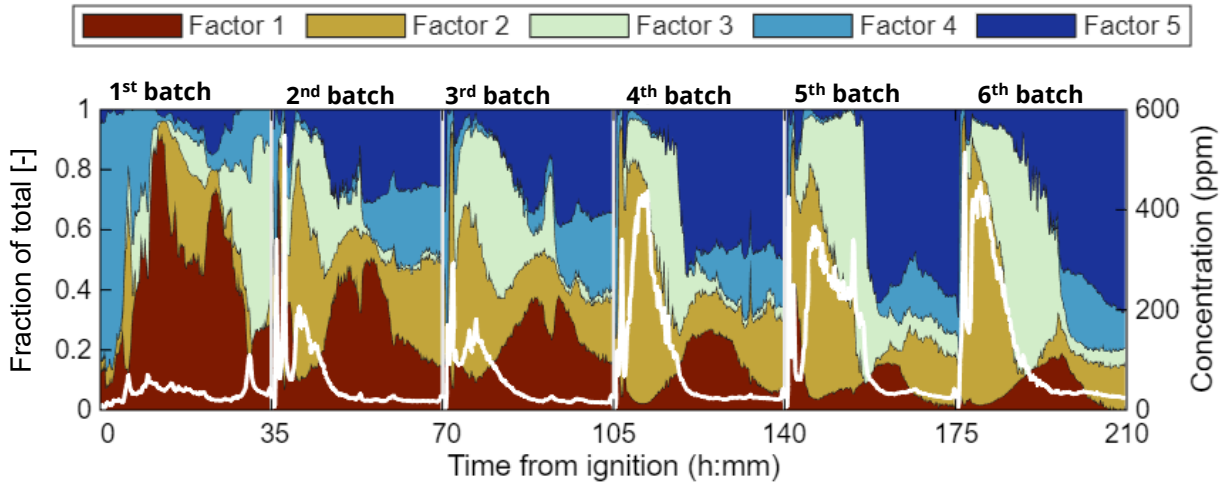


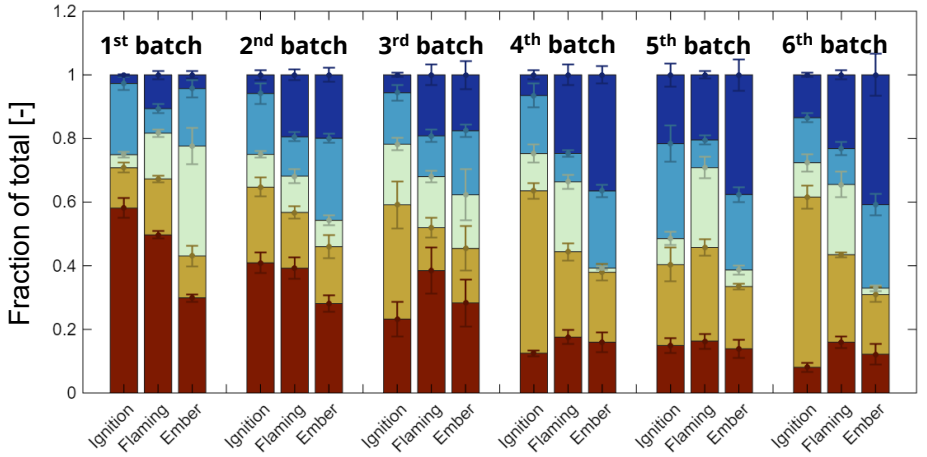
Coincides with acetylene* ← first step in soot production!



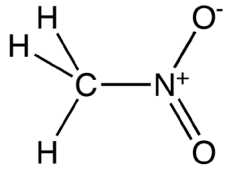
*measured by FTIR

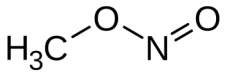
Factor 3: methyl radical oxidation and pyrosynthesis

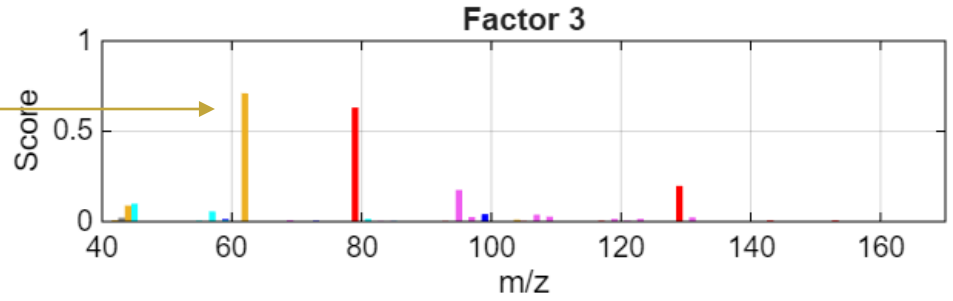


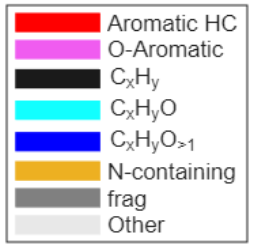


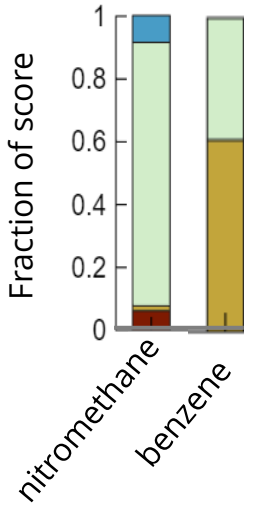
m/z 62: CH3NO2–*H*+
Nitromethane/methyl nitrite



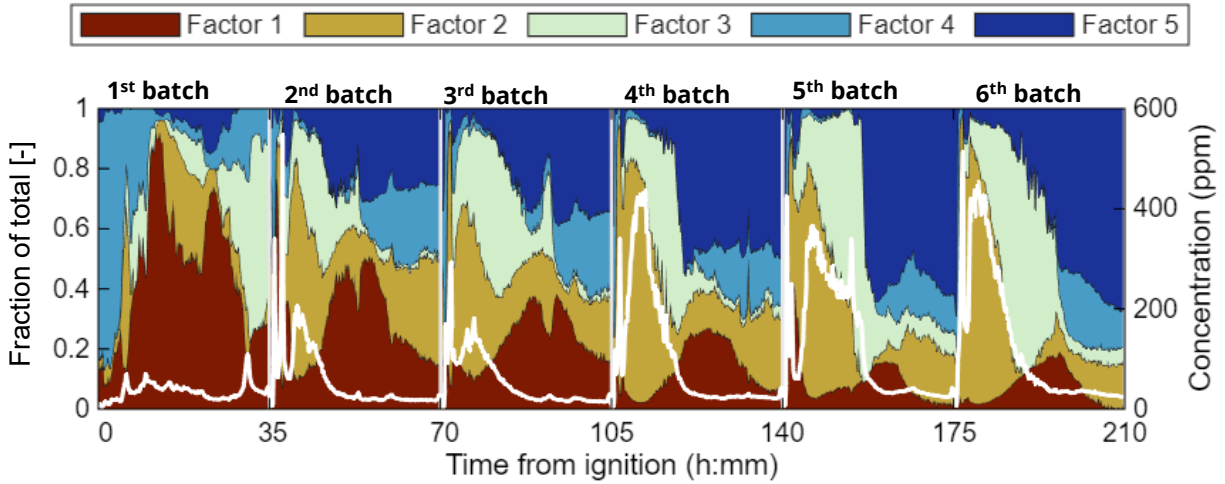


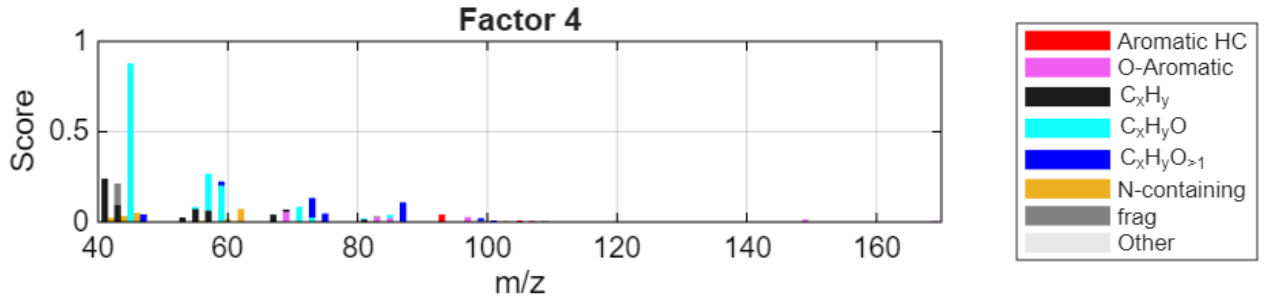


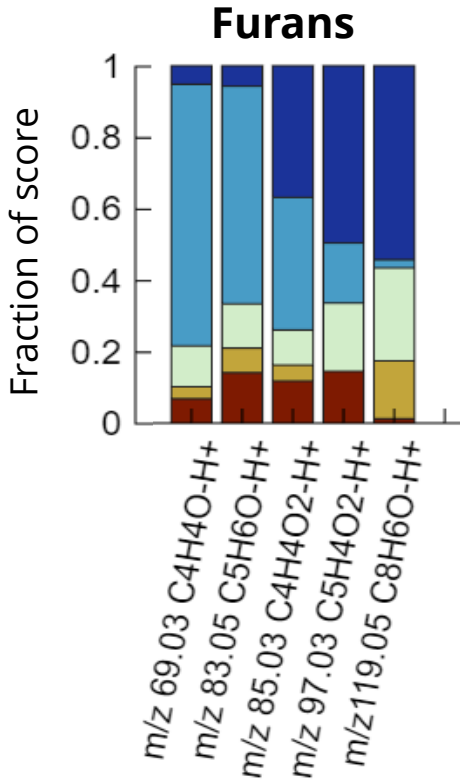


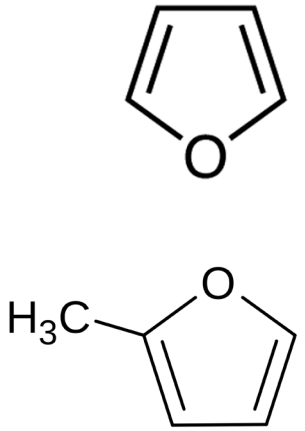


Factor 4: cellulose depolymerization

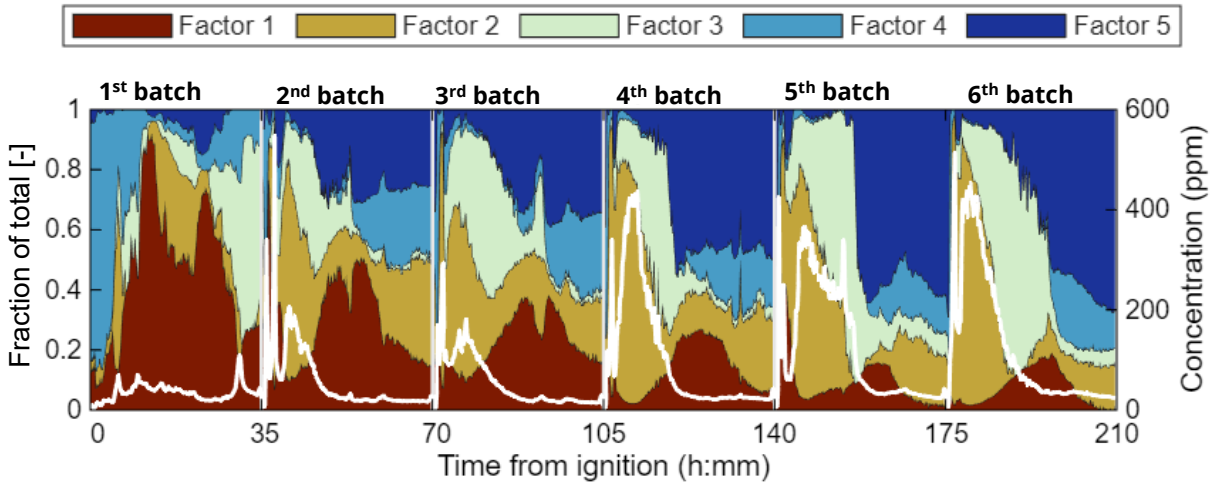


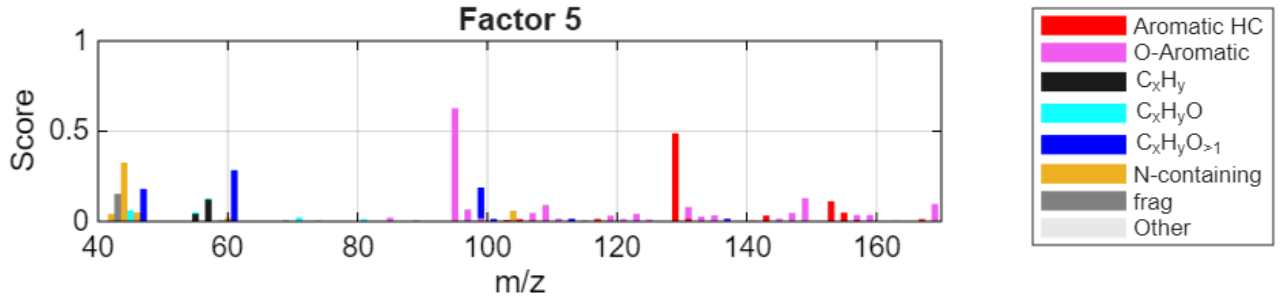


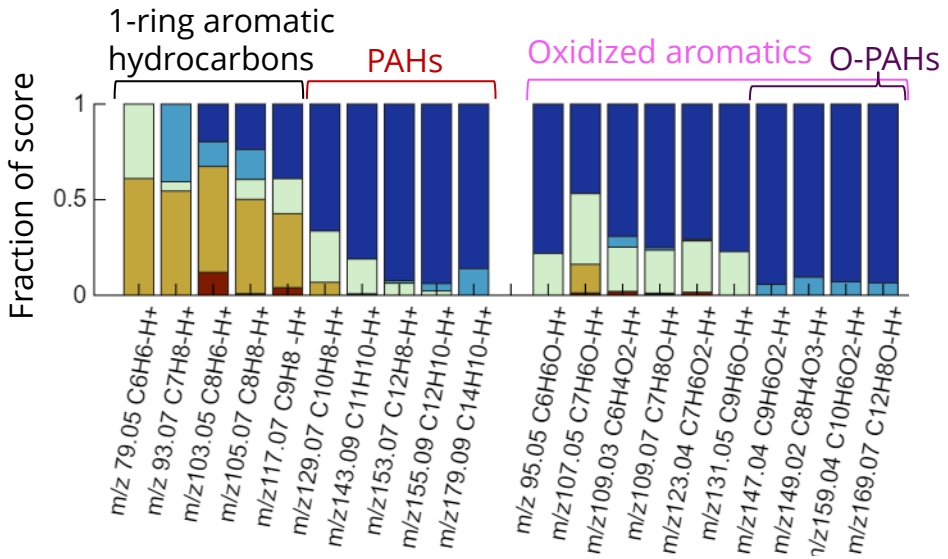




Factor 5: PAHs and oxidized aromatics increasing towards the end of the burn

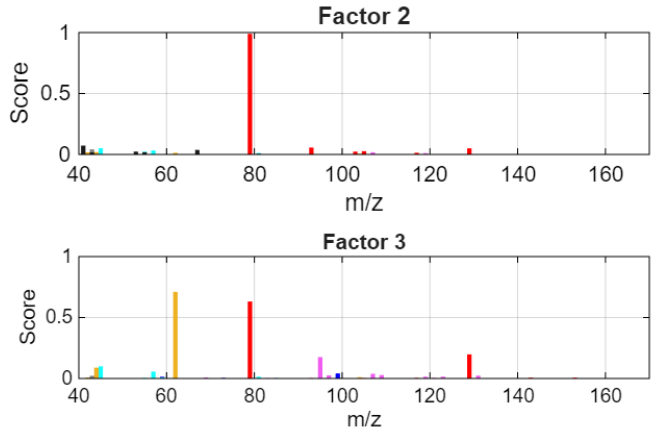






Factors 2 (pyrolysis) and 3 (pyrosynthesis) related to particulate black and brown carbon

measured by aethalometer



BC:

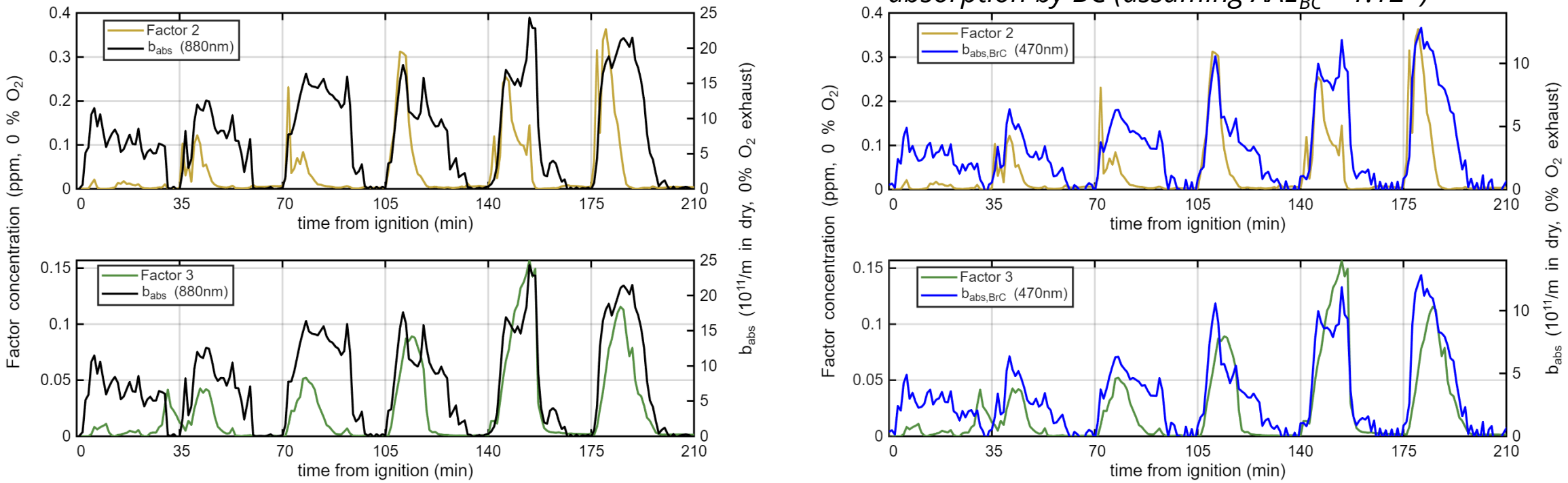
based on absorption coefficient at 880 nm

BrC:

absorption coefficient at 470 nm after subtracting absorption by BC (assuming AAE_{BC} = 1.12)*

F2:

F3:



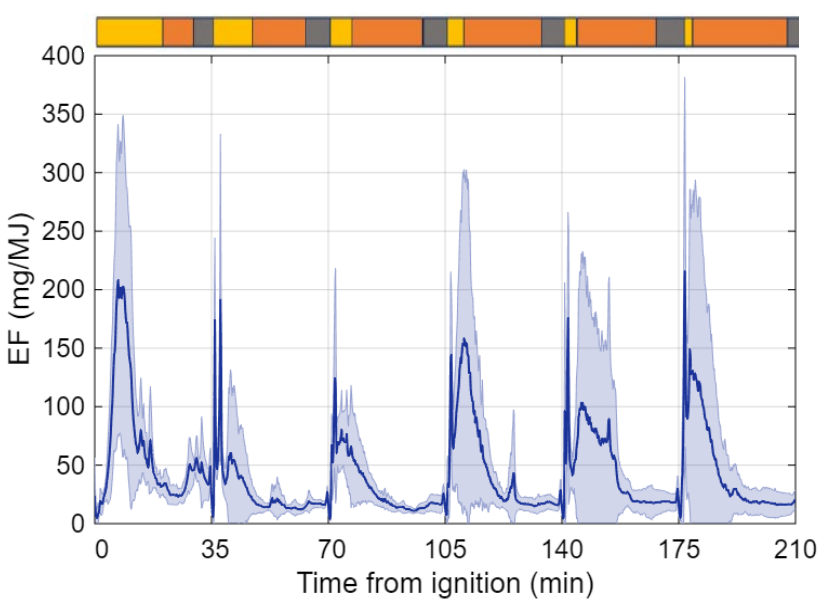
*<u>Basnet S.</u> *et al., 2024, ACP (doi: 10.5194/acp-24-3197-2024)*

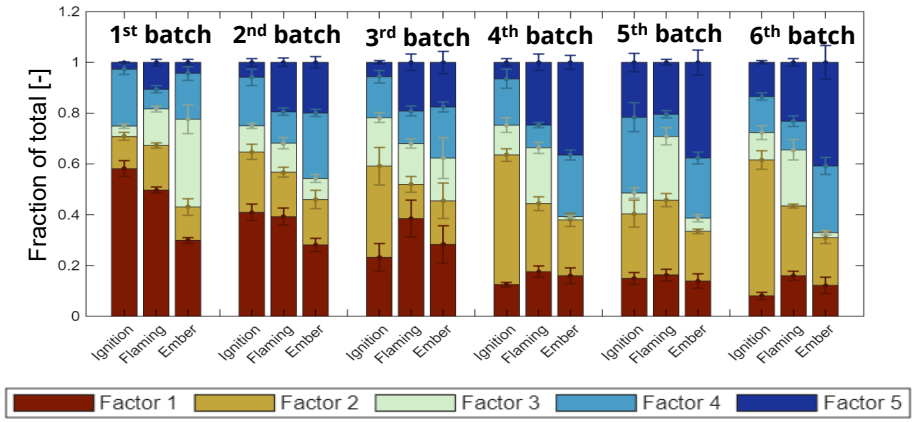
Conclusions

Contact: anni.hartikainen@uef.fi

- Organic emissions from residential wood combustion vary as combustion progresses
  - aromatic hydrocarbons characteristic for high-efficiency combustion
- Emission profile vulnerable to changes in particle (soot) concentration
- Several overlapping formation processes, where emission of specific compounds can be linked together

  → combustion diagnostics of organic PM or soot formation





Thank you!

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  - Helmholtz Virtual Institute of Complex Molecular Systems in Environmental Health - Aerosols and Health (HICE)

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- Jane and Aatos Erkko Foundation BoFF-project

aeroHEALTH HELMHOLTZ International Lab

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J&AE JANE JA AATOS ERKON SÄÄTIÖ

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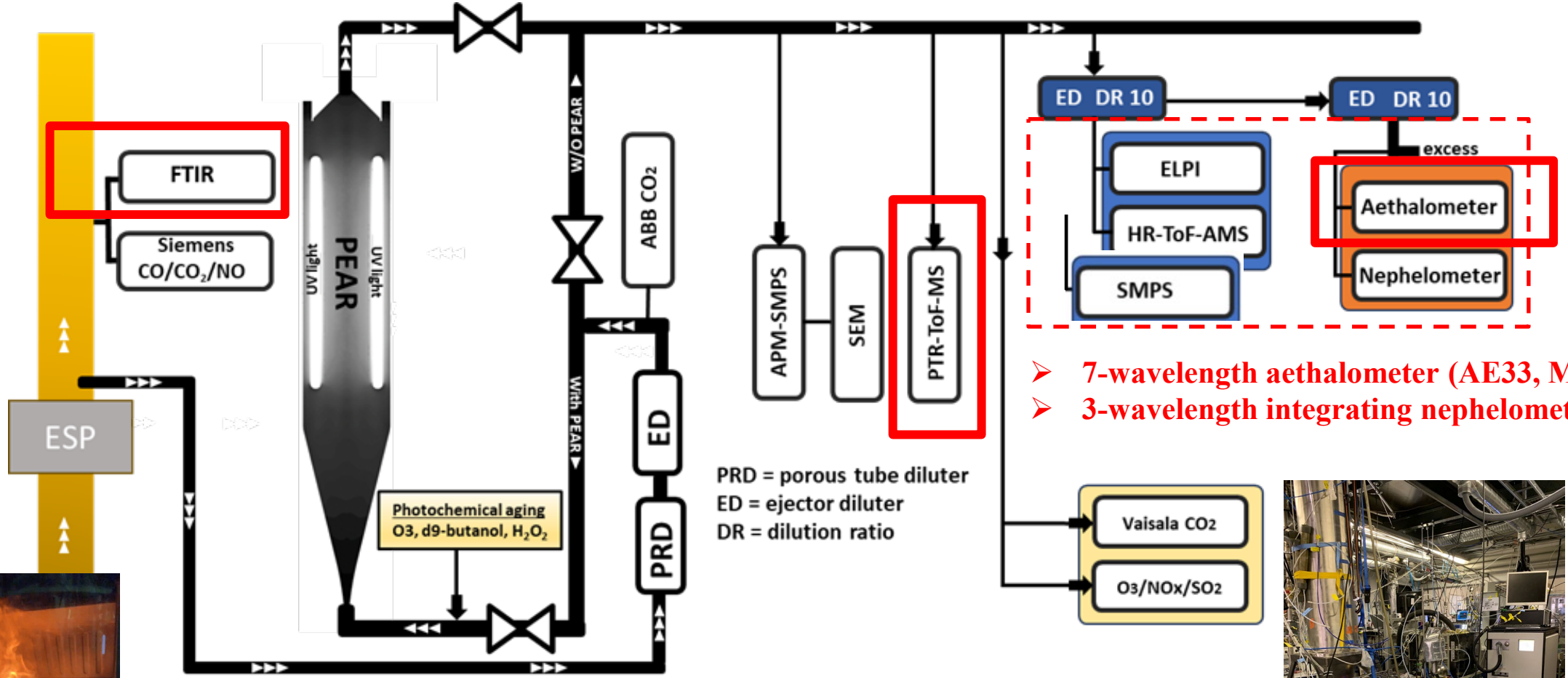
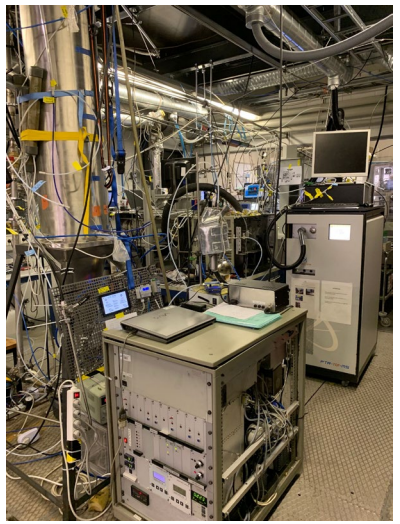
Experimental setup



- 7-wavelength aethalometer (AE33, Magee Scientific)
- 3-wavelength integrating nephelometer (Model 3563, TS



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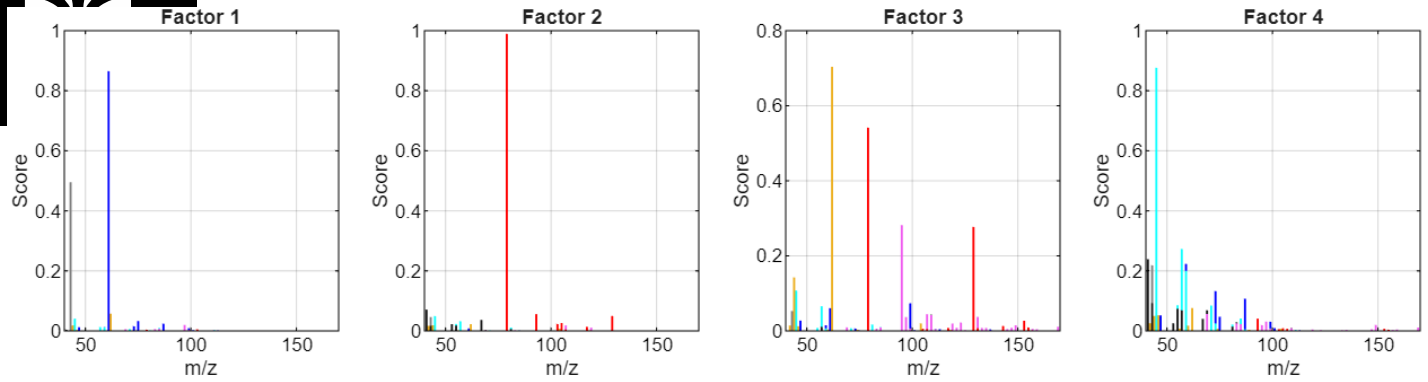
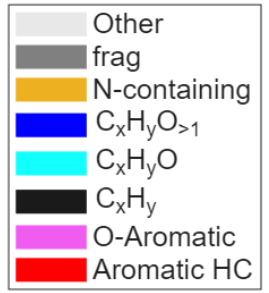
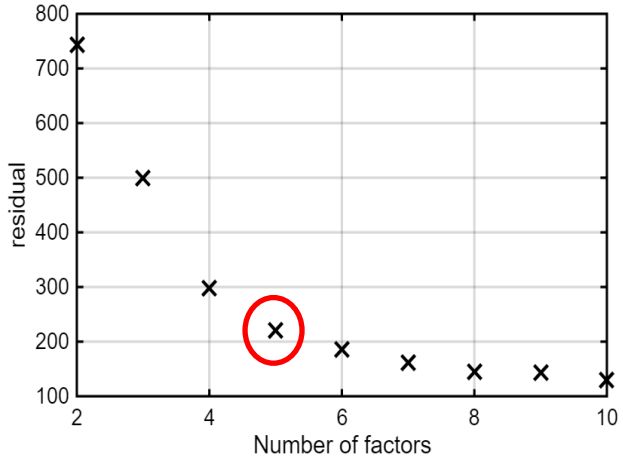
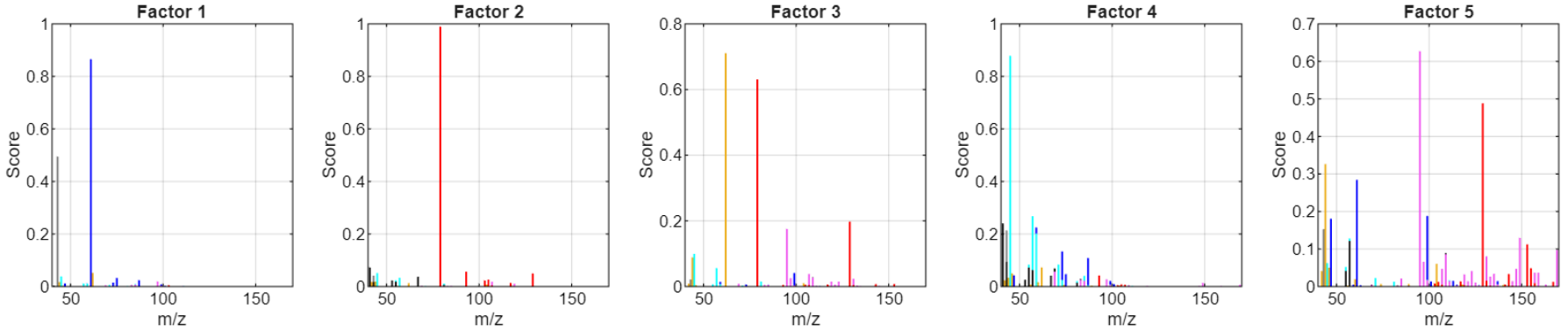
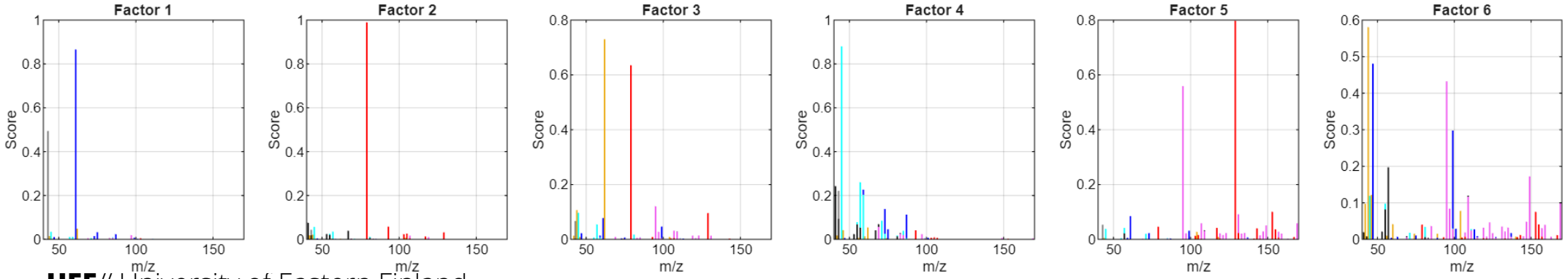
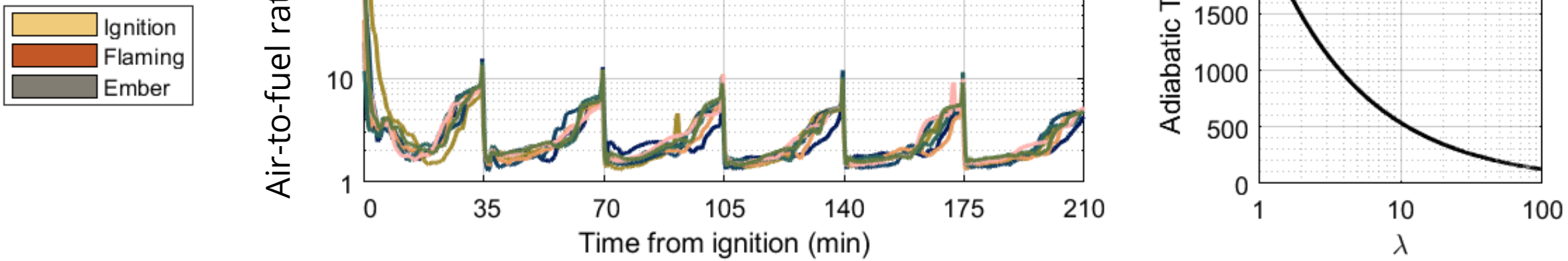

Factor 1
Factor 2
Factor 3
Factor 4
Score
m/z

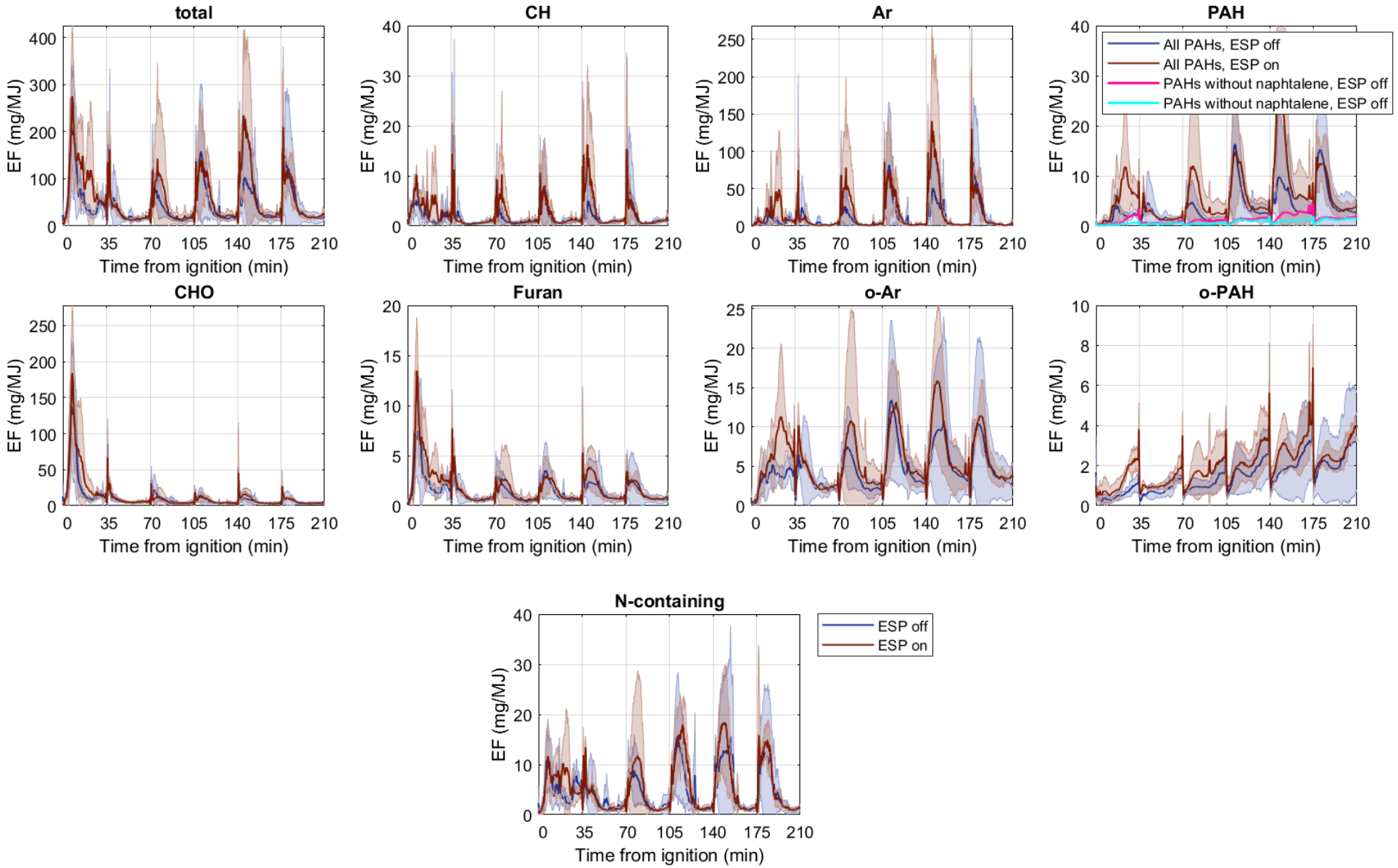

Other
frag
N-containing
CxHyO>1
CxHyO
CxHy
O-Aromatic
Aromatic HC

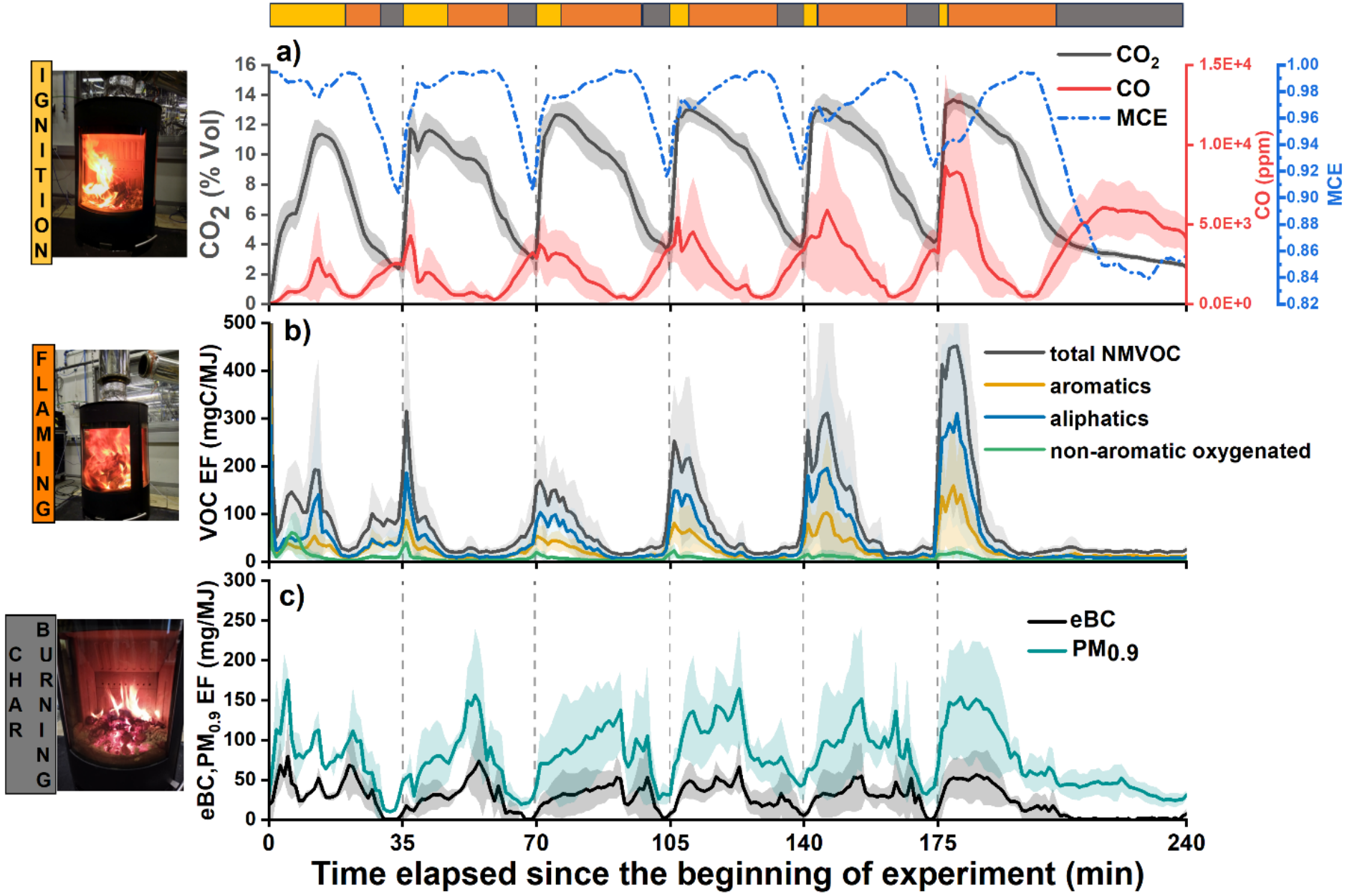

residual
Number of factors


Factor 1
Factor 2
Factor 3
Factor 4
Factor 5
Score
m/z

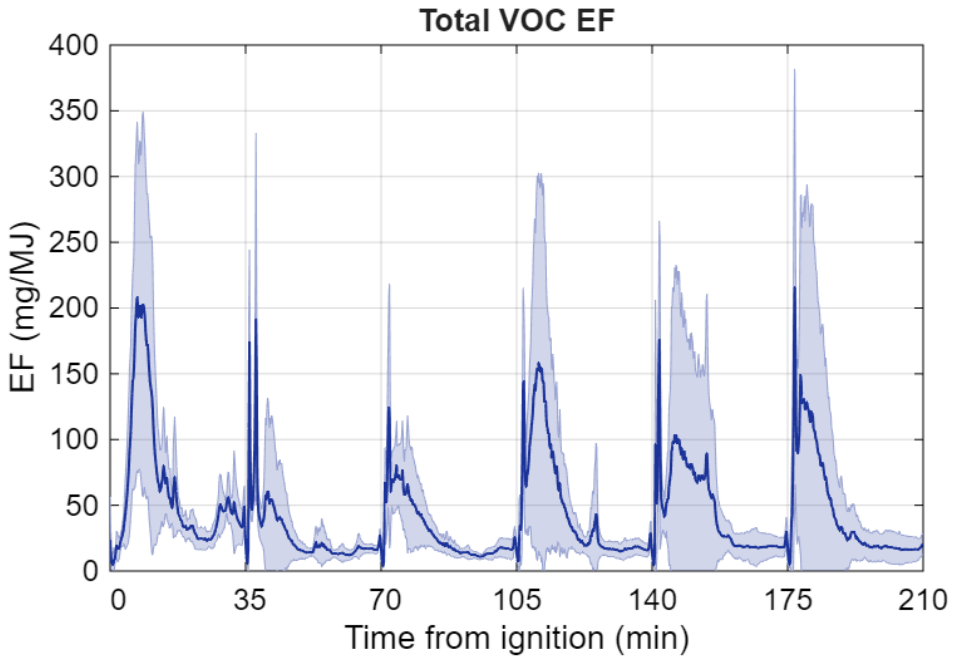

Factor 1
Factor 2
Factor 3
Factor 4
Factor 5
Factor 6
Score
m/z

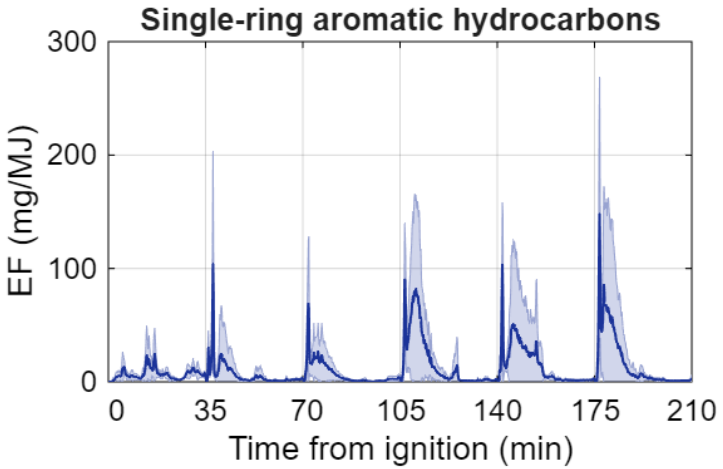

Ignition
Flaming
Ember
Air-to-fuel ratio (λ)
100
10
1
0
35
70
105
140
175
210
Time from ignition (min)
Adiabatic T (K)
2000
1500
1000
500
0
1
10
100
λ

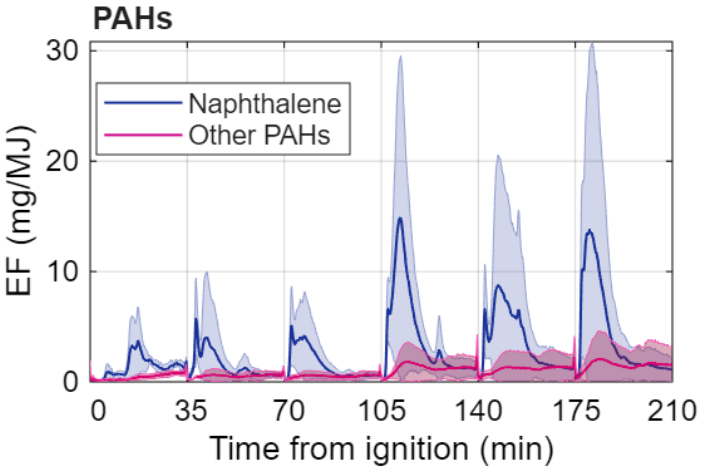

total
CH
Ar
PAH
All PAHs, ESP off
All PAHs, ESP on
PAHs without naphtalene, ESP off
PAHs without naphtalene, ESP off
CHO
Furan
o-Ar
o-PAH
N-containing
ESP off
ESP on
EF (mg/MJ)
Time from ignition (min)

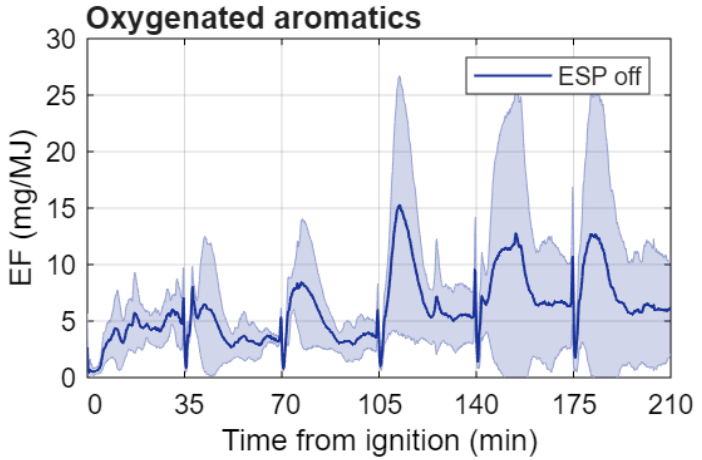

IGNITION
FLAMING
CHAR BURNING
a)
b)
c)
CO2 (% Vol)
CO (ppm)
MCE
CO2
CO
MCE
VOC EF (mgC/MJ)
total NMVOC
aromatics
aliphatics
non-aromatic oxygenated
eBC,PM0.9 EF (mg/MJ)
eBC
PM0.9
Time elapsed since the beginning of experiment (min)

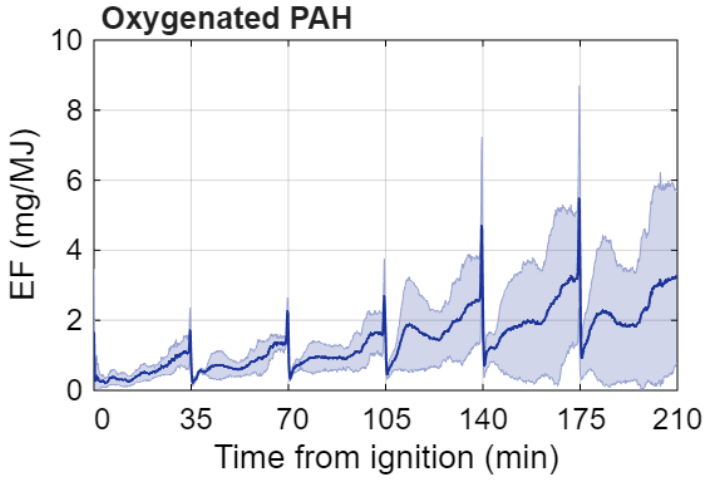
PAH EFs increase towards end of batch

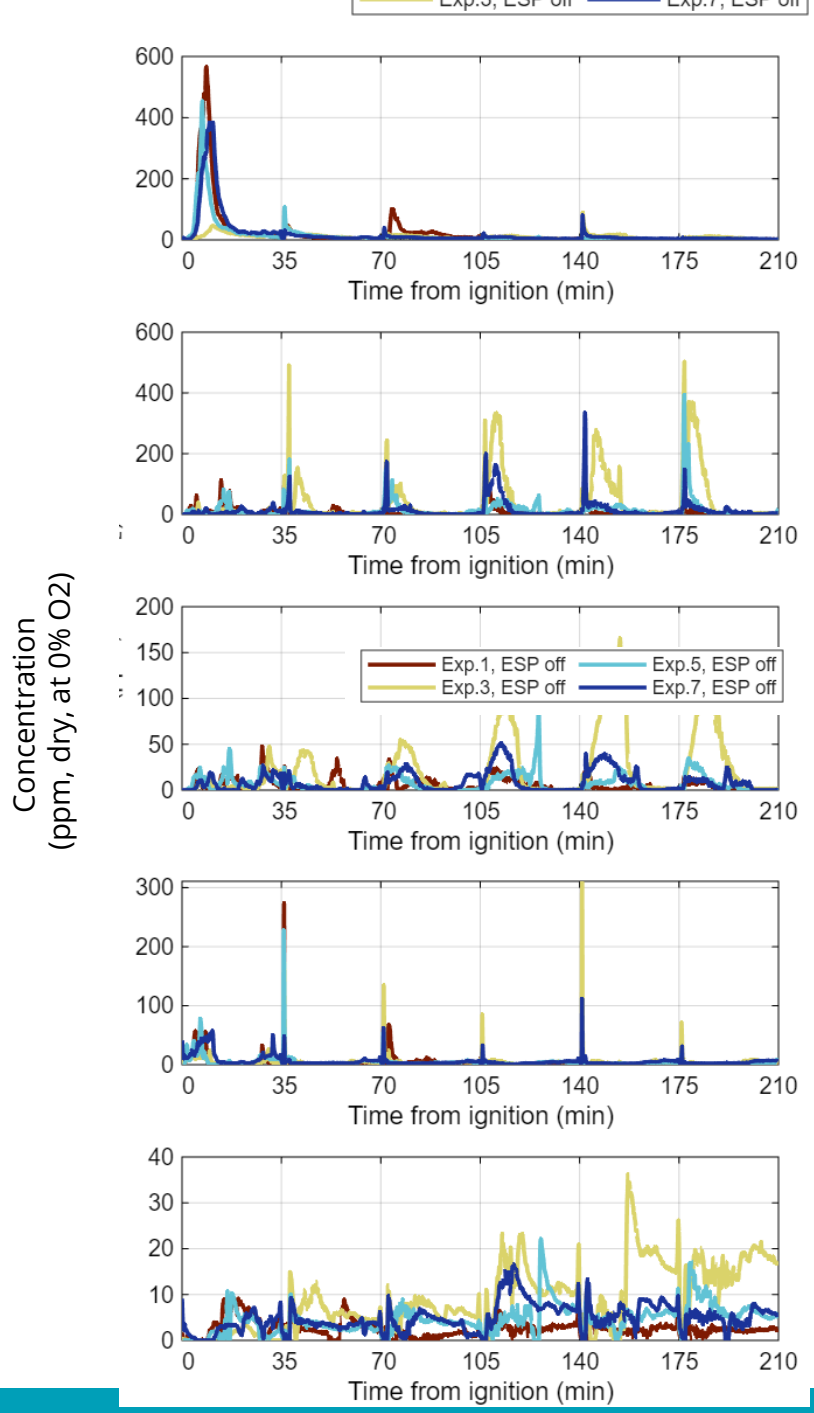




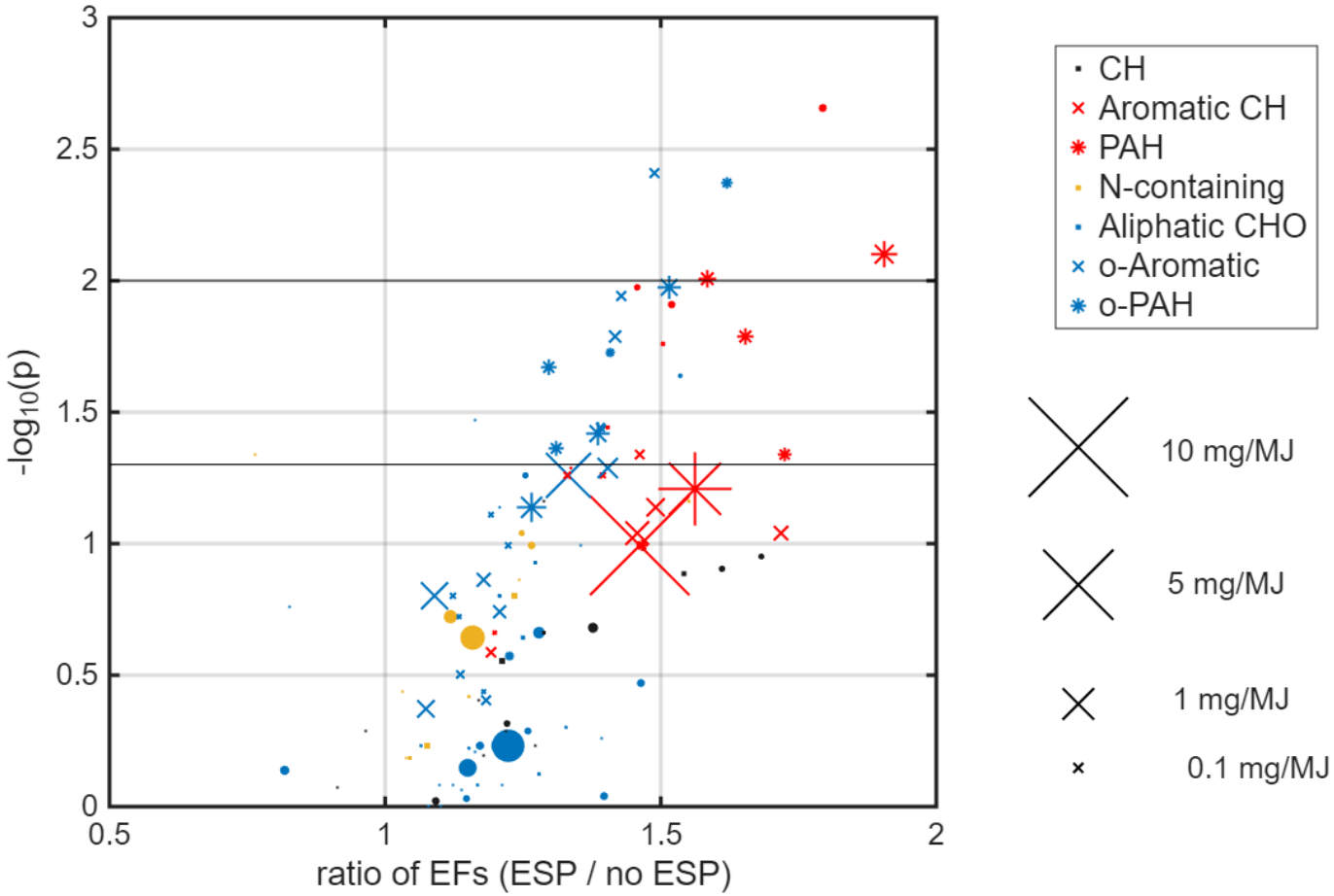


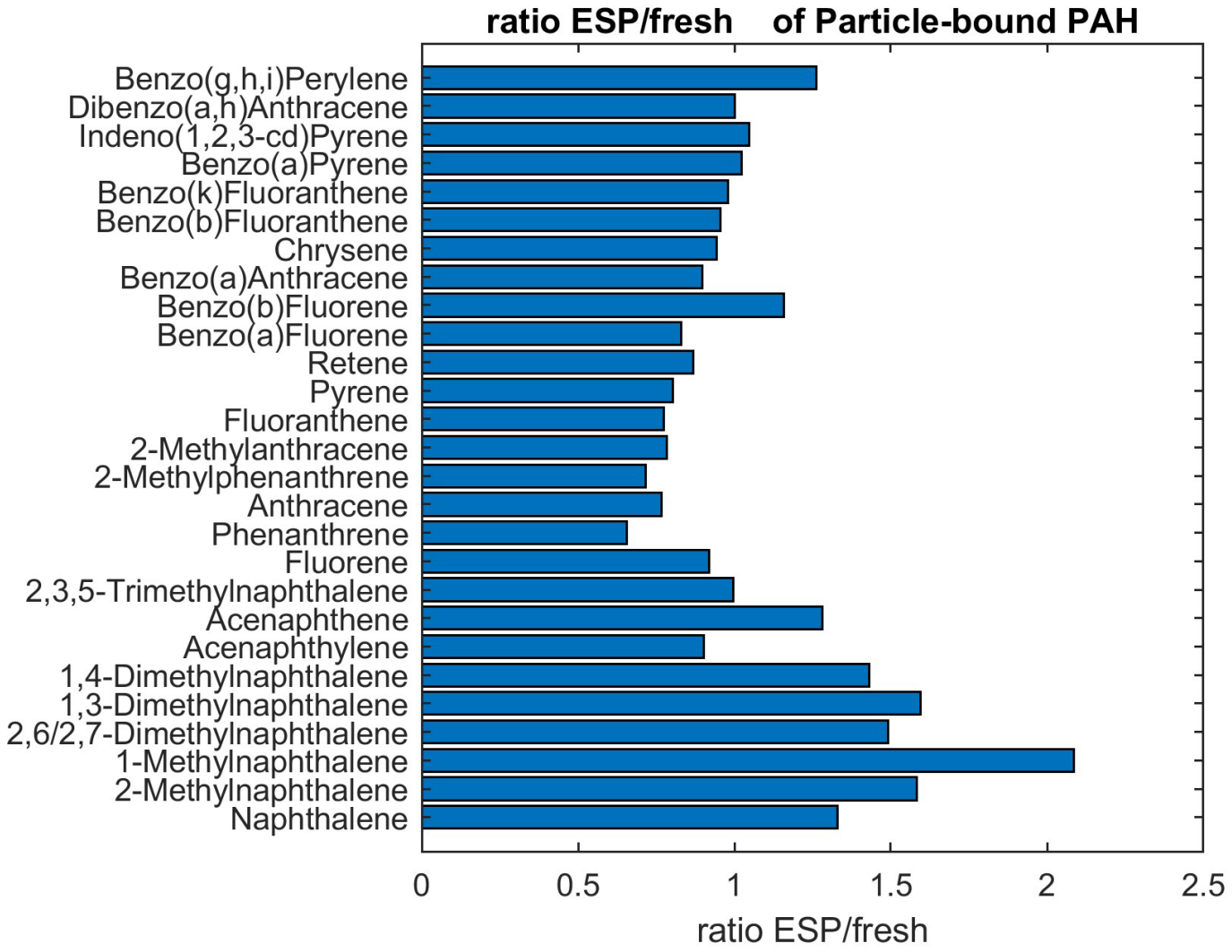





Concentration
(ppm, dry, at 0% O2)
Exp.1, ESP off
Exp.3, ESP off
Exp.5, ESP off
Exp.7, ESP off
Time from ignition (min)

Increase mainly for polycyclic compounds with relatively low EFs




ratio ESP/fresh of Particle-bound PAH
Benzo(g,h,i)Perylene
Dibenzo(a,h)Anthracene
Indeno(1,2,3-cd)Pyrene
Benzo(a)Pyrene
Benzo(k)Fluoranthene
Benzo(b)Fluoranthene
Chrysene
Benzo(a)Anthracene
Benzo(b)Fluorene
Benzo(a)Fluorene
Retene
Pyrene
Fluoranthene
2-Methylanthracene
2-Methylphenanthrene
Anthracene
Phenanthrene
Fluorene
2,3,5-Trimethylnaphthalene
Acenaphthene
Acenaphthylene
1,4-Dimethylnaphthalene
1,3-Dimethylnaphthalene
2,6/2,7-Dimethylnaphthalene
1-Methylnaphthalene
2-Methylnaphthalene
Naphthalene
0
0.5
1
1.5
2
2.5
ratio ESP/fresh