Using a novel chamber to investigate the evolution of single plume from biomass burning

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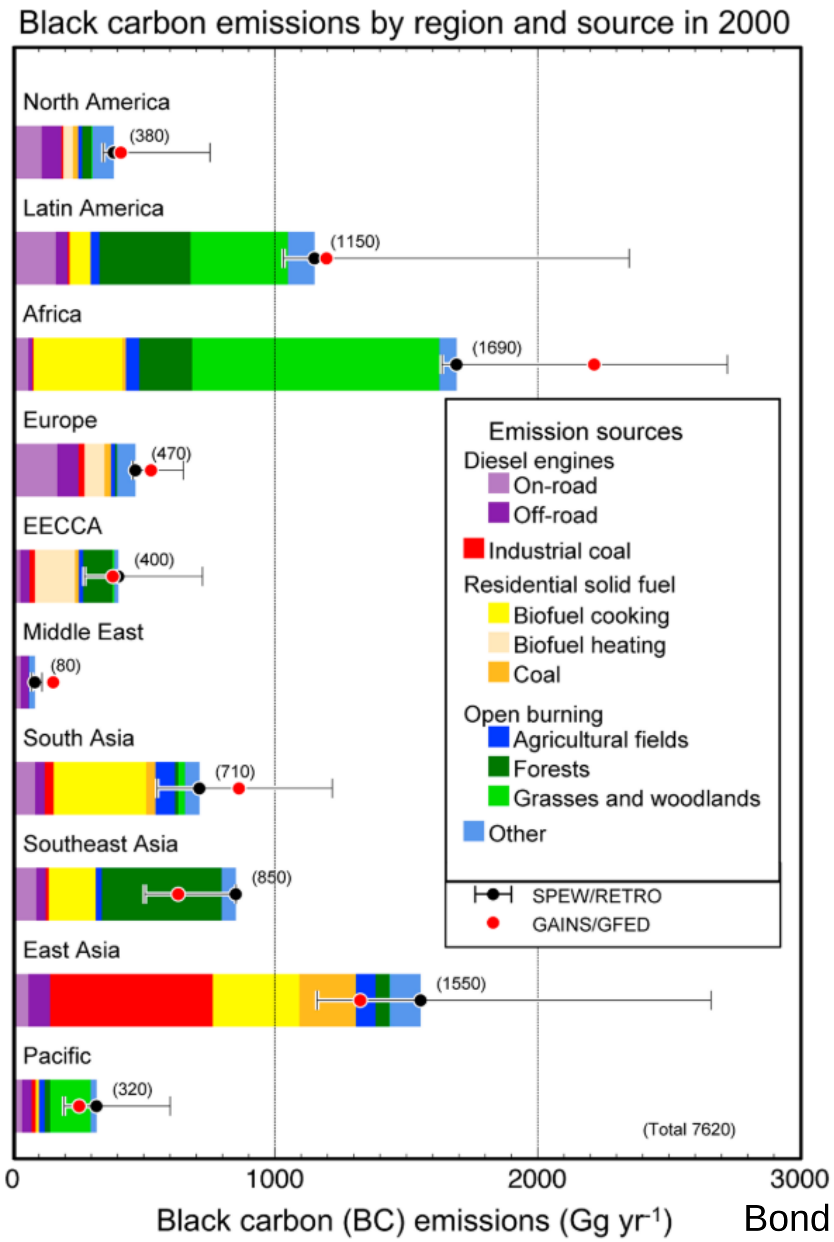
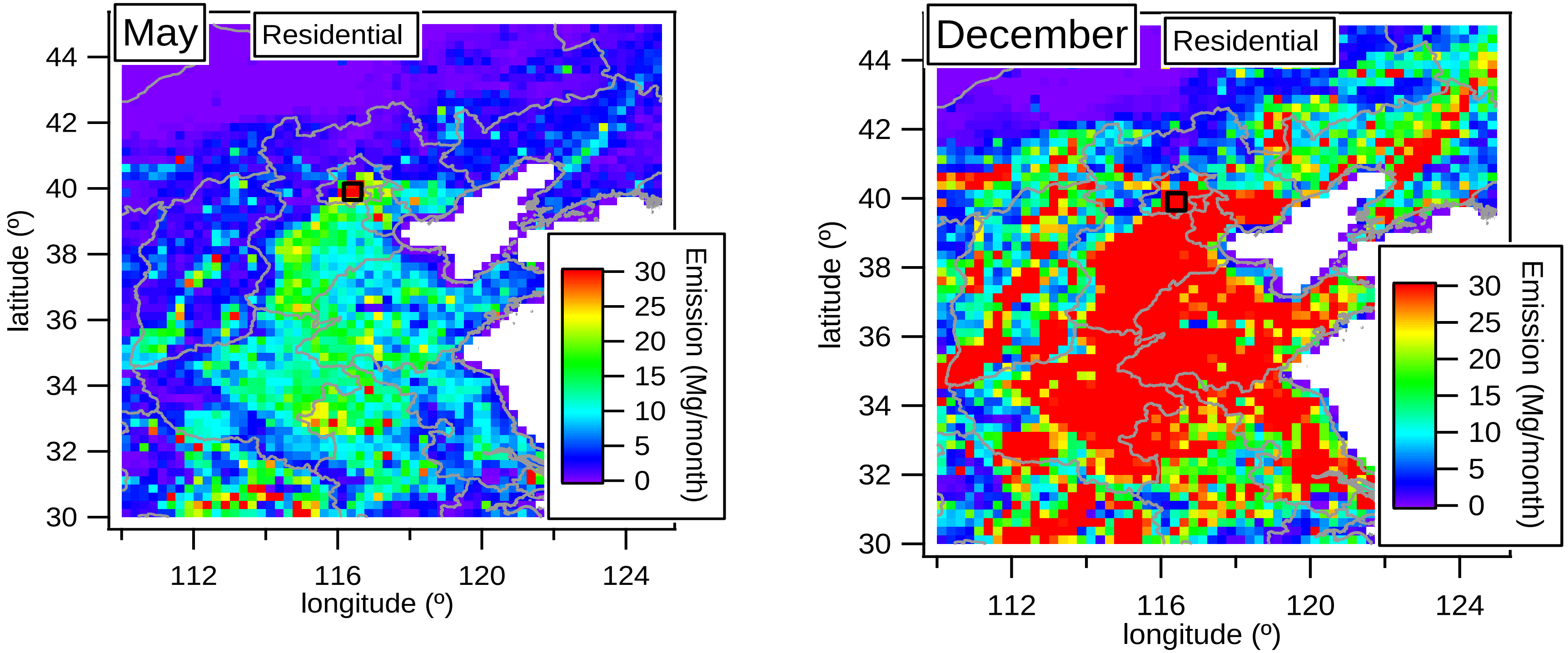
Global distribution of solid fuel burning emissions



Bond et al., 2013

East Asia importantly contributes to emissions from solid fuel burning

Emission inventory of residential solid fuel burning over the NCP



Li, M., Zhang, Q. et al., Atmos. Chem. Phys., 17, 935, 2017.

Solid fuel and burning phases

Wood burning

Coal burning

Purpose of solid fuel burning

Residential

Cooking

Heating

Industrial production and power plant

Burning stoves

Heating stove

CarbonZero

Gyapa

Lucia stove

Burning Phase: Flaming and Smoldering

Flaming (FL)

Smoldering (SM)

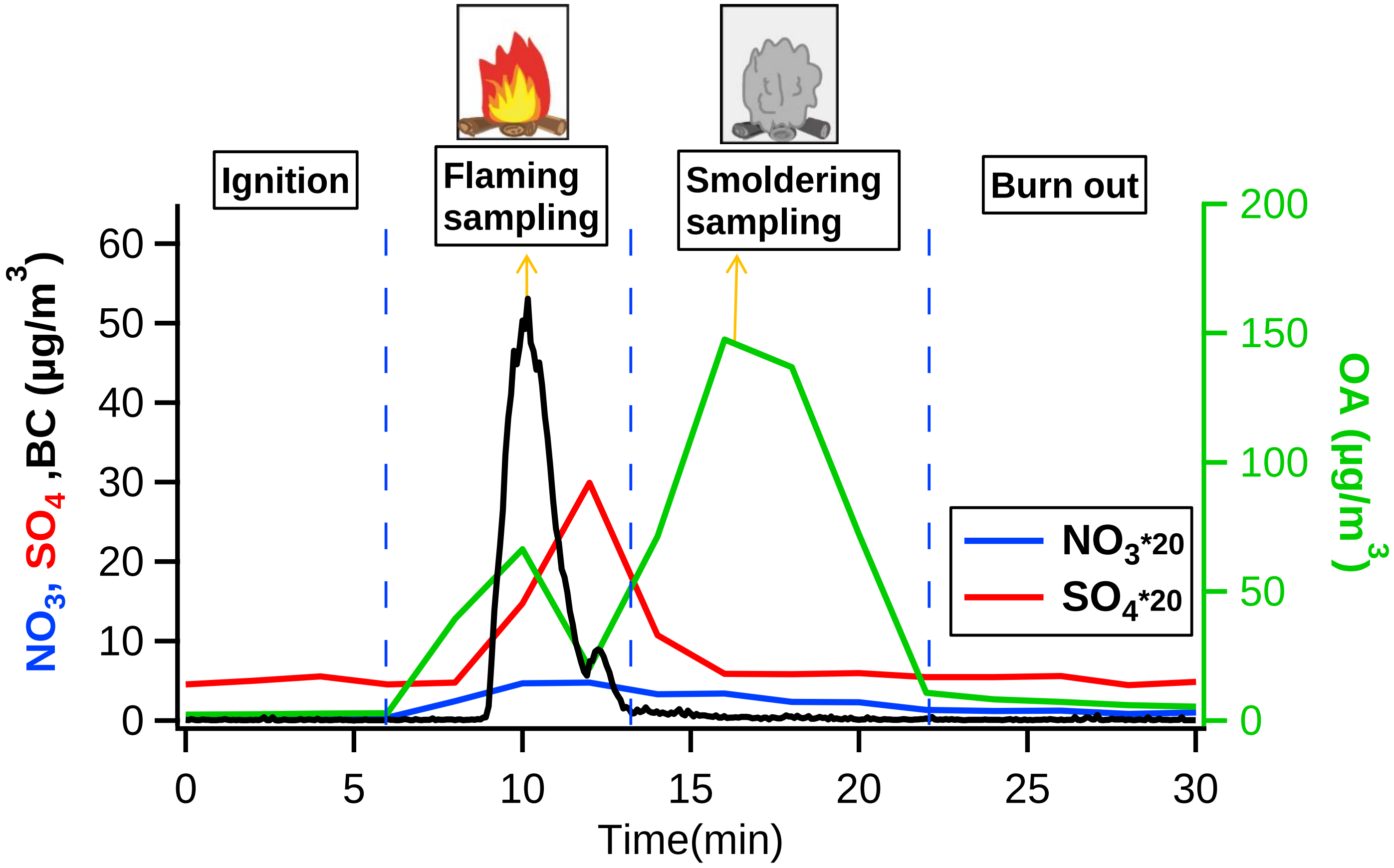
Gas
- CO/CO_2
- NO_x
- SO_2
- VOCs

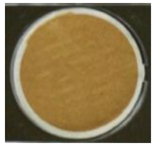
mixing ⟷ transformation

Aerosol
- Organic Aerosol (OA)
- Inorganic Salt
- Soot
- Potassium
- Metal/Mineral

A typical burning cycle of solid fuel burning

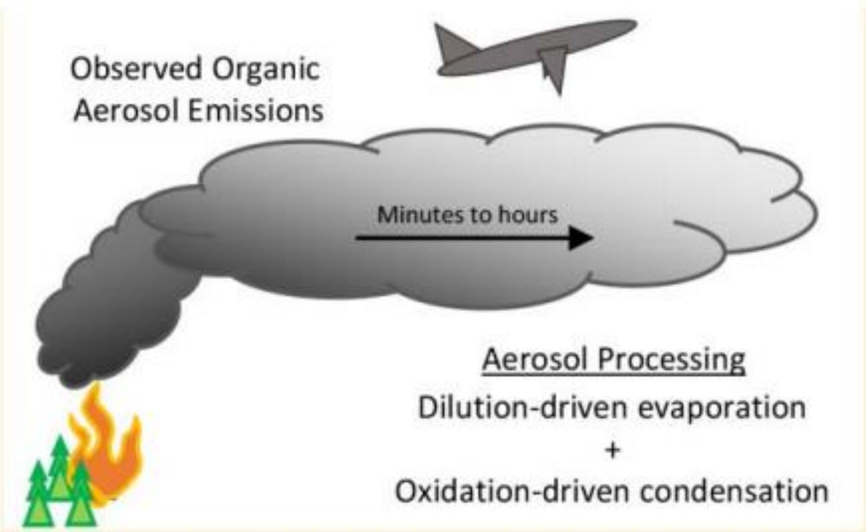
Flaming: BC dominated Smoldering: OA dominated





Importance of evolution

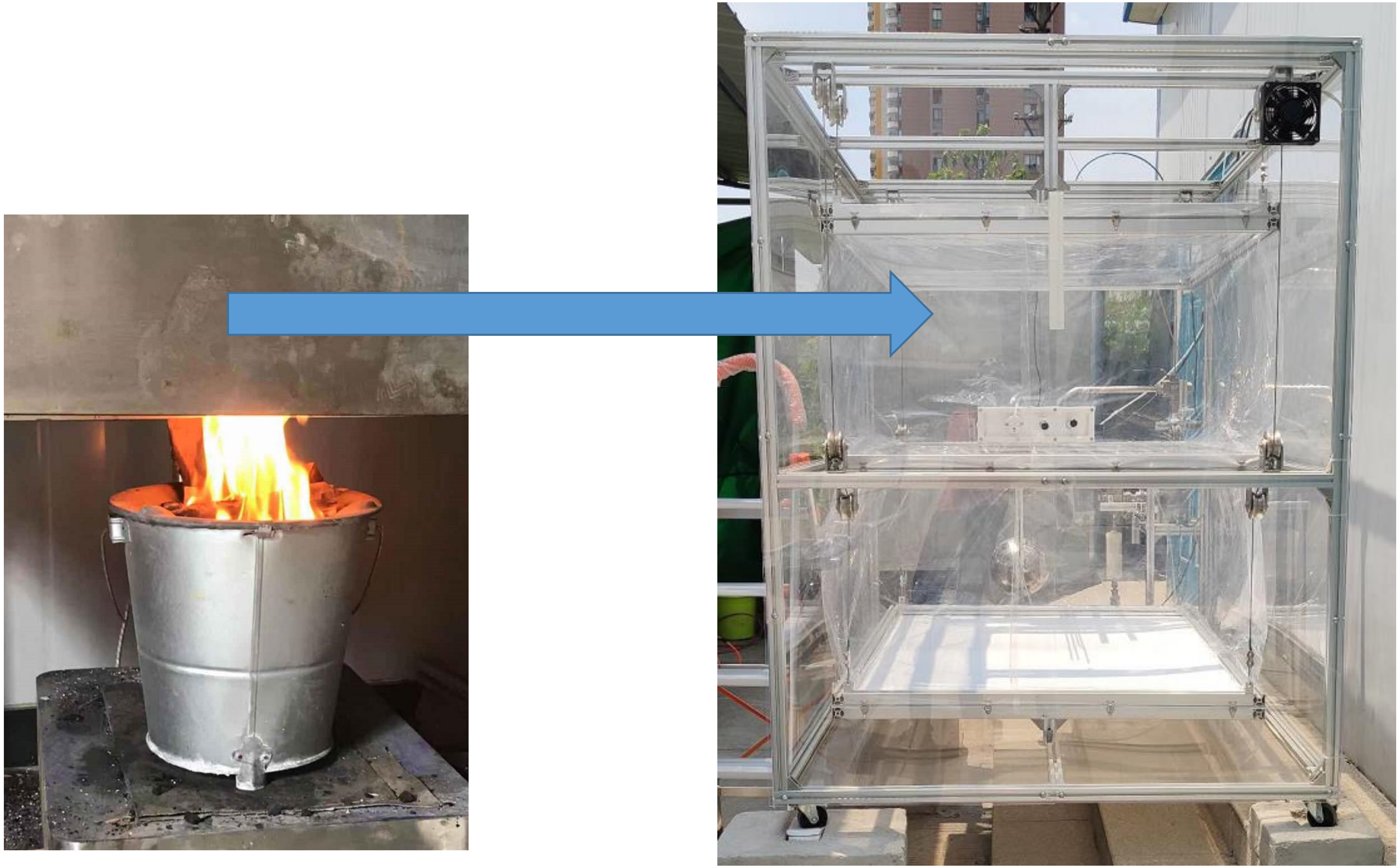
How does biomass burning emission evolve in the atmosphere, and what extent can SOA be generated and what are the properties?



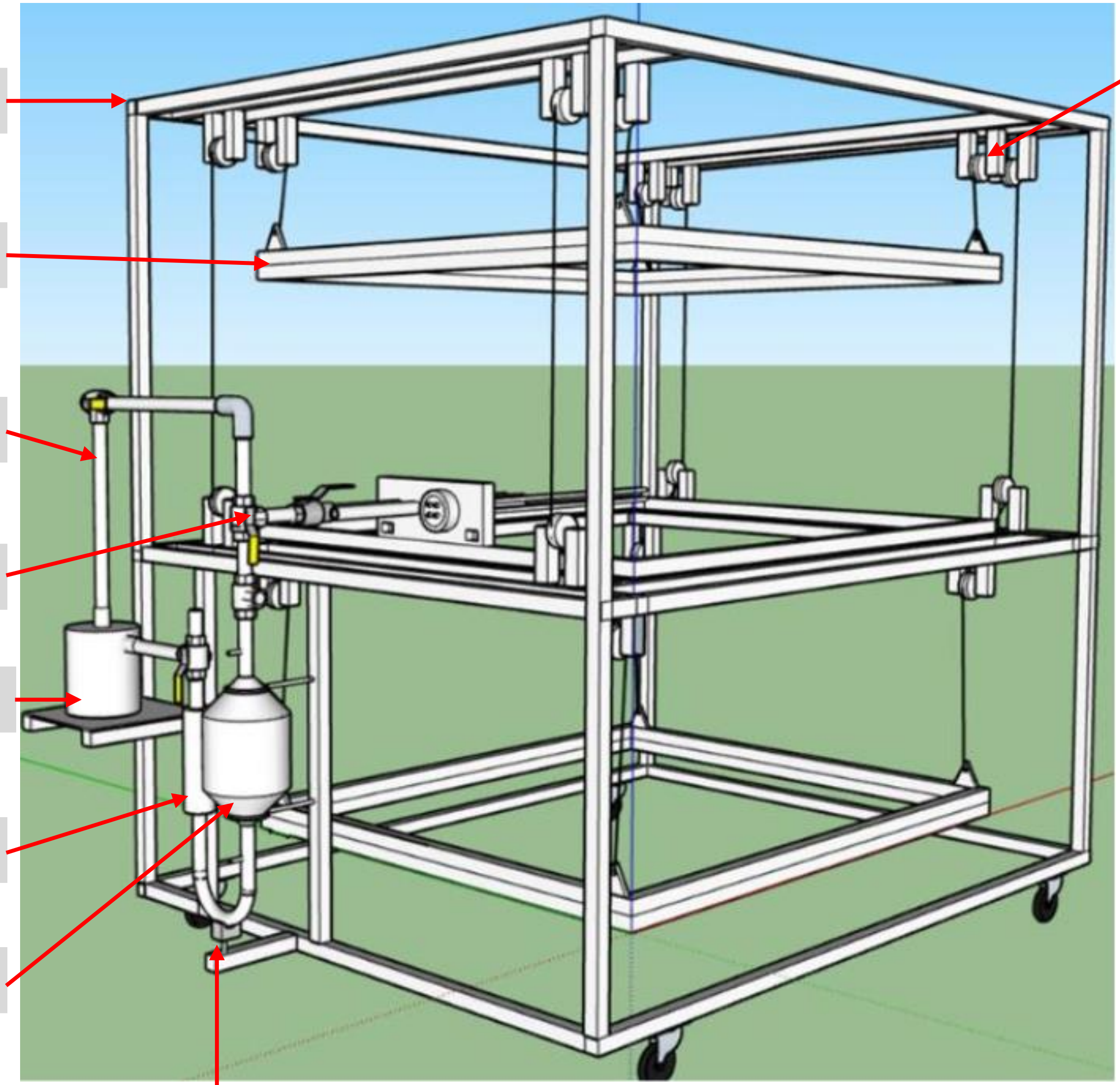
Hodshire. et al., 2019, EST

Emission: evaporation, rapid oxidation and condensation, secondary particulate matter formation.
The burning phase affects the gas and particle precursors.

Plume Evolution Chamber



Structure of our novel-designed chamber

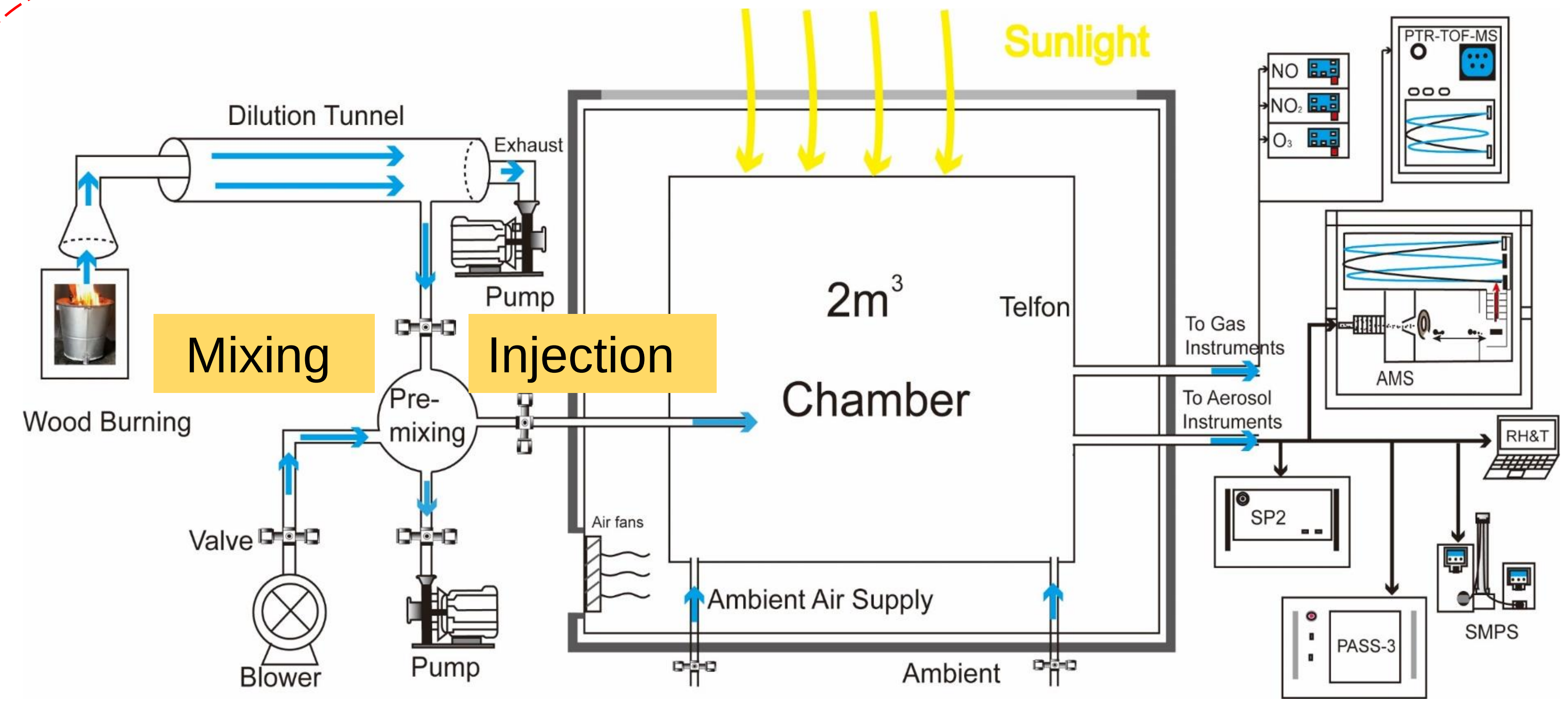


The internal chamber is made of 0.05 mm PTFE film (1.2×1×1.2 m). The external four sides (excluding the top and bottom) of the chamber are covered by transparent acrylic plates (1.8×1.3×1.7m).

Plume Evolution Chamber

Previous laboratory studies
- ➢ Emissions from different combustion phases cannot be effectively isolated.
- ➢ High levels of oxidants injected
- ➢ Artificial UV light

Novel-designed chamber
- ➢ Isolating single plumes from a certain combustion phase.
- ➢ Ambient air oxidants
- ➢ Solar radiation



Instrumentation

- **PASS-3: absorption**
- **SP2: BC mass, size and mixing state**
- **AMS: non-refractory compositions**
- **SMPS: size distribution**
- **CO, NOx, O_3**
- **PTR-TOF-MS: VOCs**

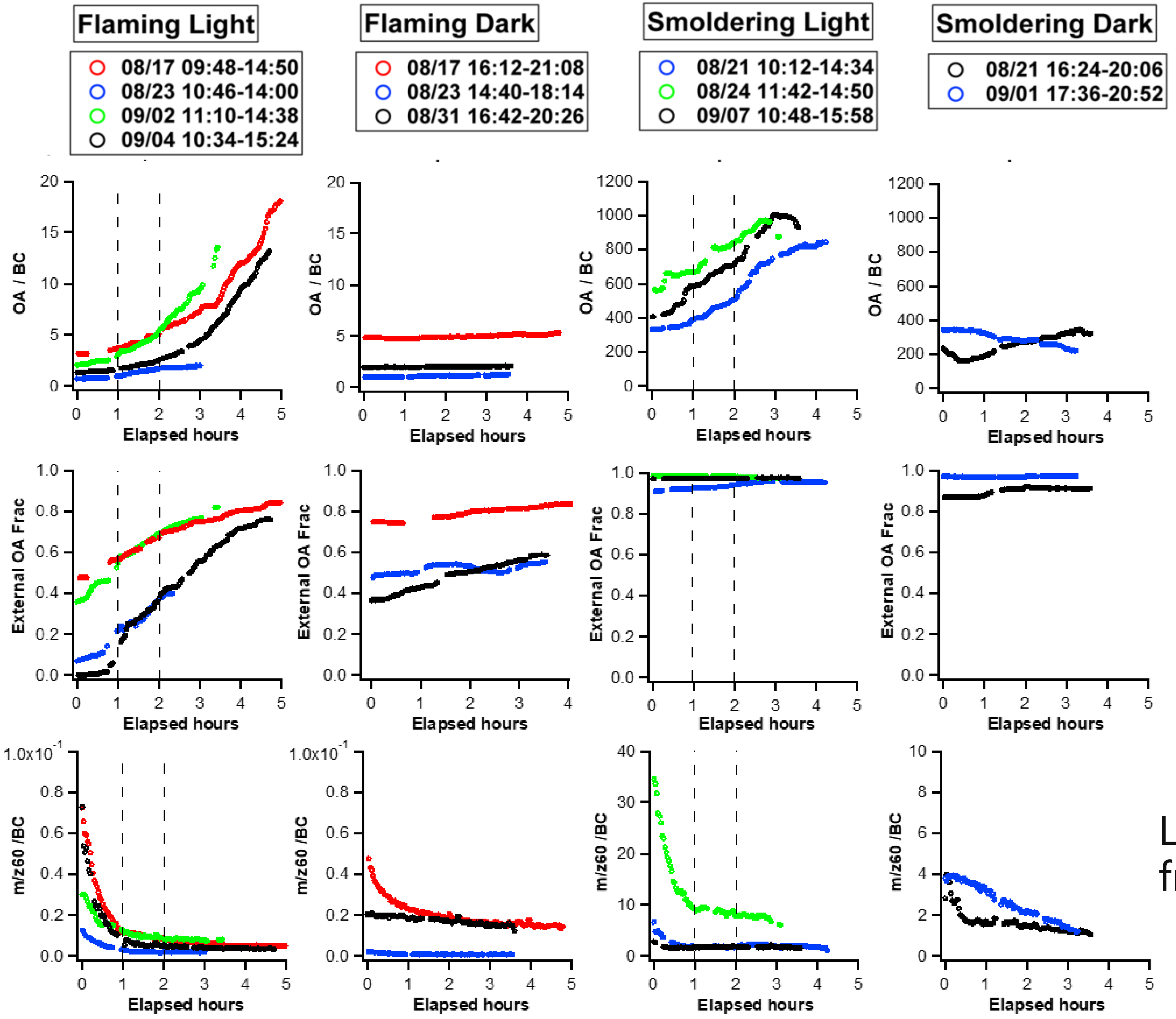
Evolution of Aerosol Chemical Properties

Evolution for burning phases and solar radiation

SOA formation

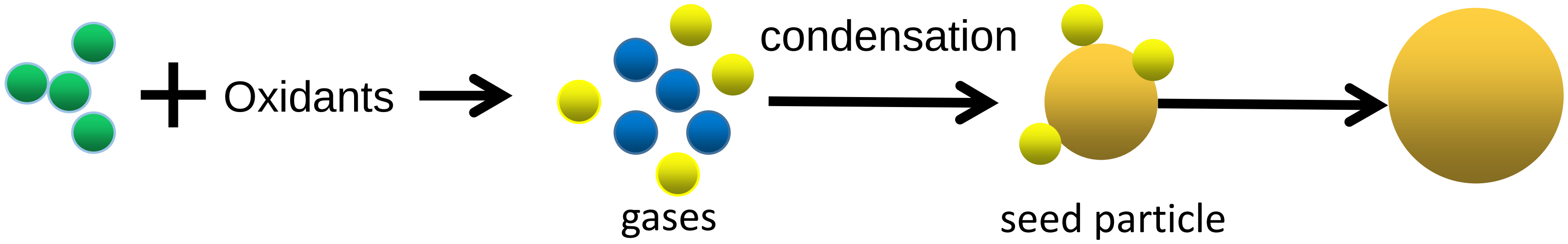
OA externally mixed with BC

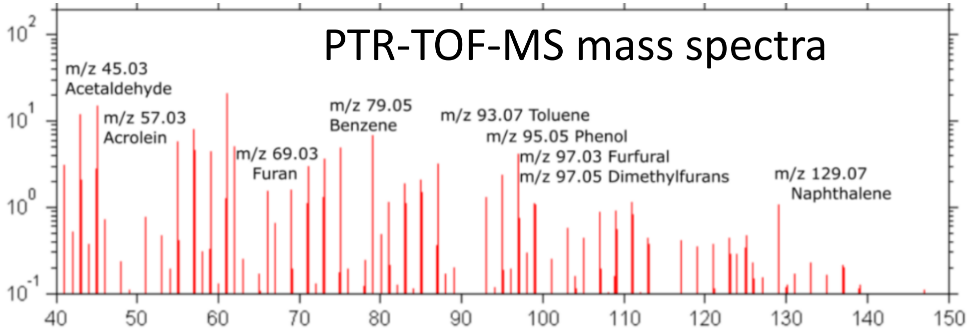
Evaporation process

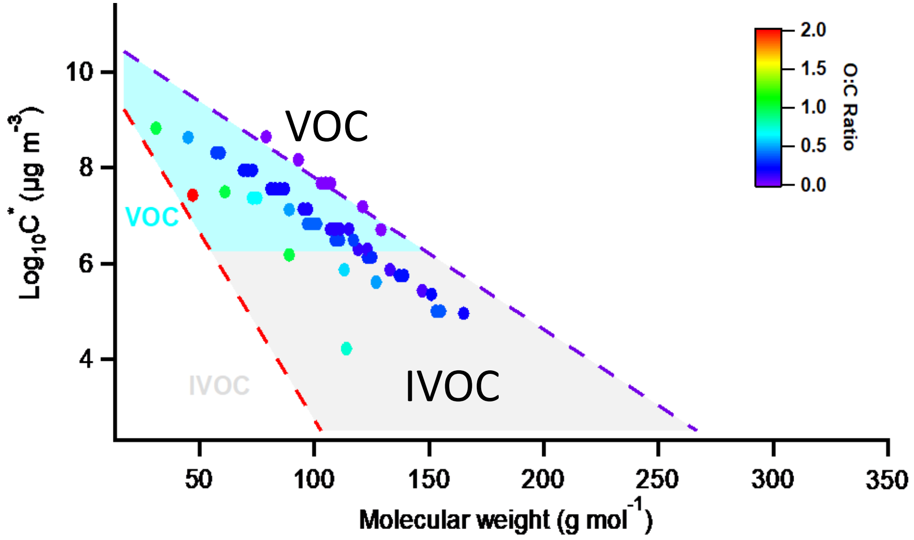


Levoglucosan fragment

Oxidation of VOCs and aerosol

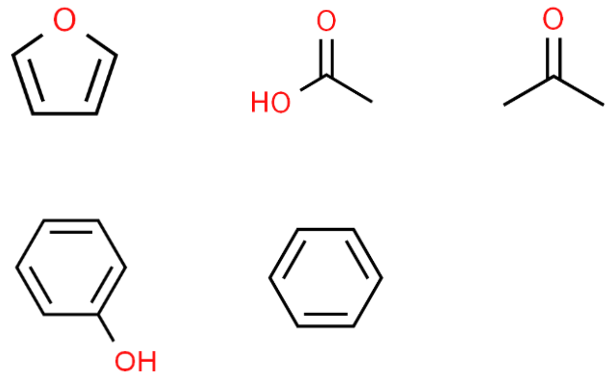






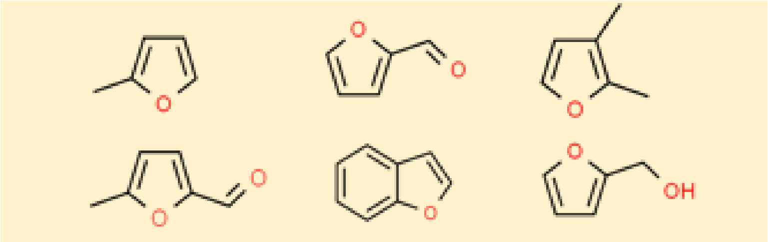
109 species

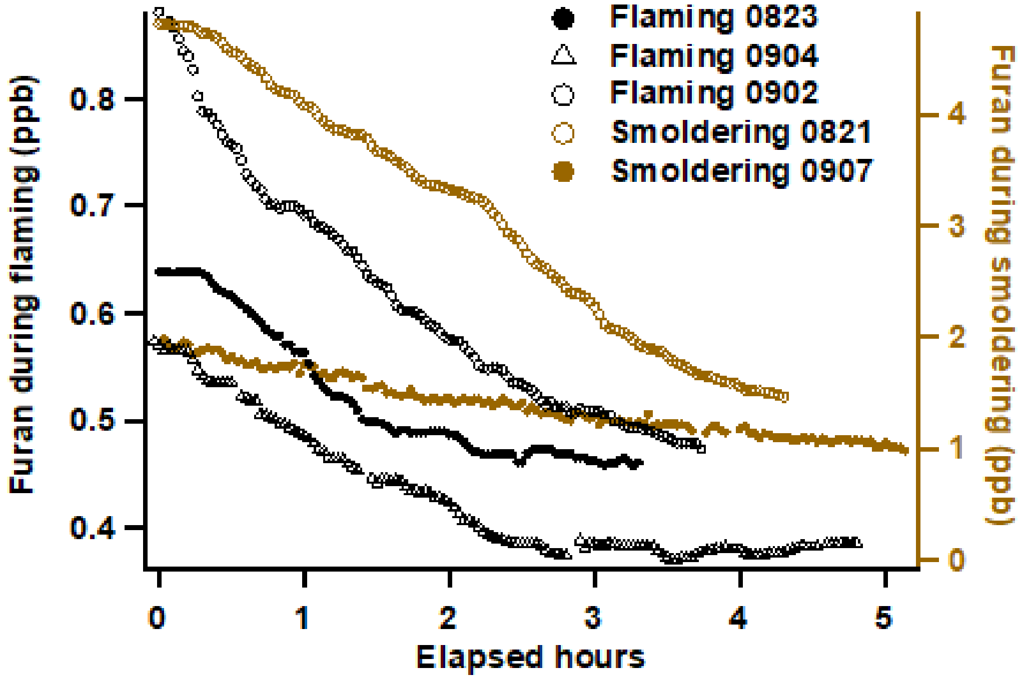
VOCs: furanic, carboxylic acids, carbonyl compounds (aldehydes and ketones), phenols and aromatic compounds.



Evolution of VOCs

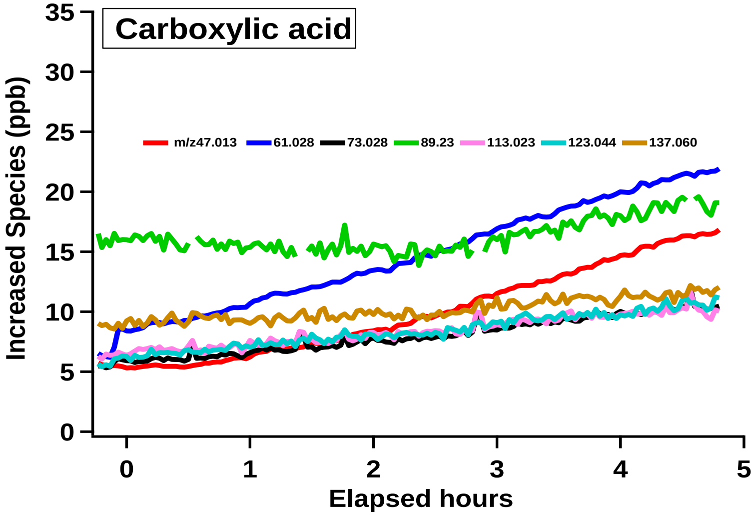
Furanic compounds:
Important precursor



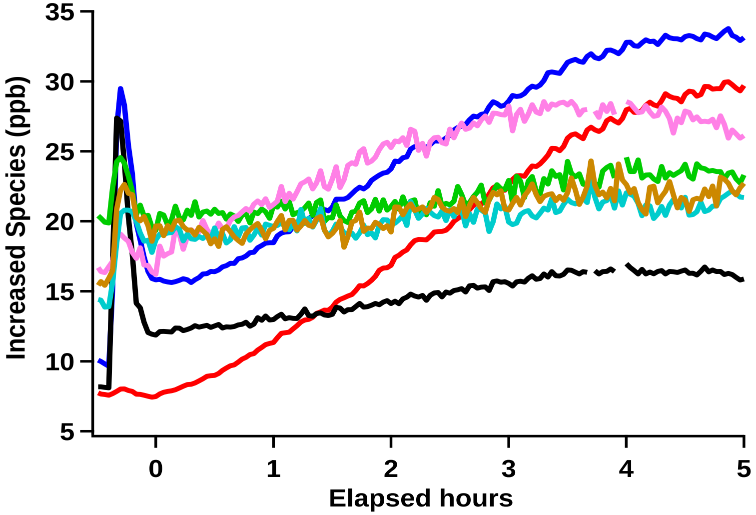


Oxidation products:

Flaming

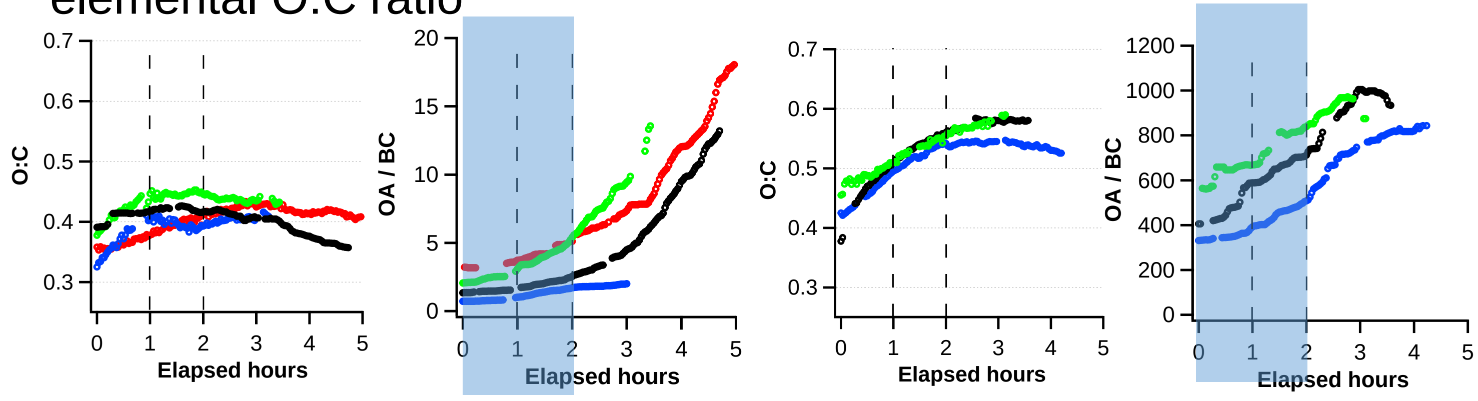


Smoldering

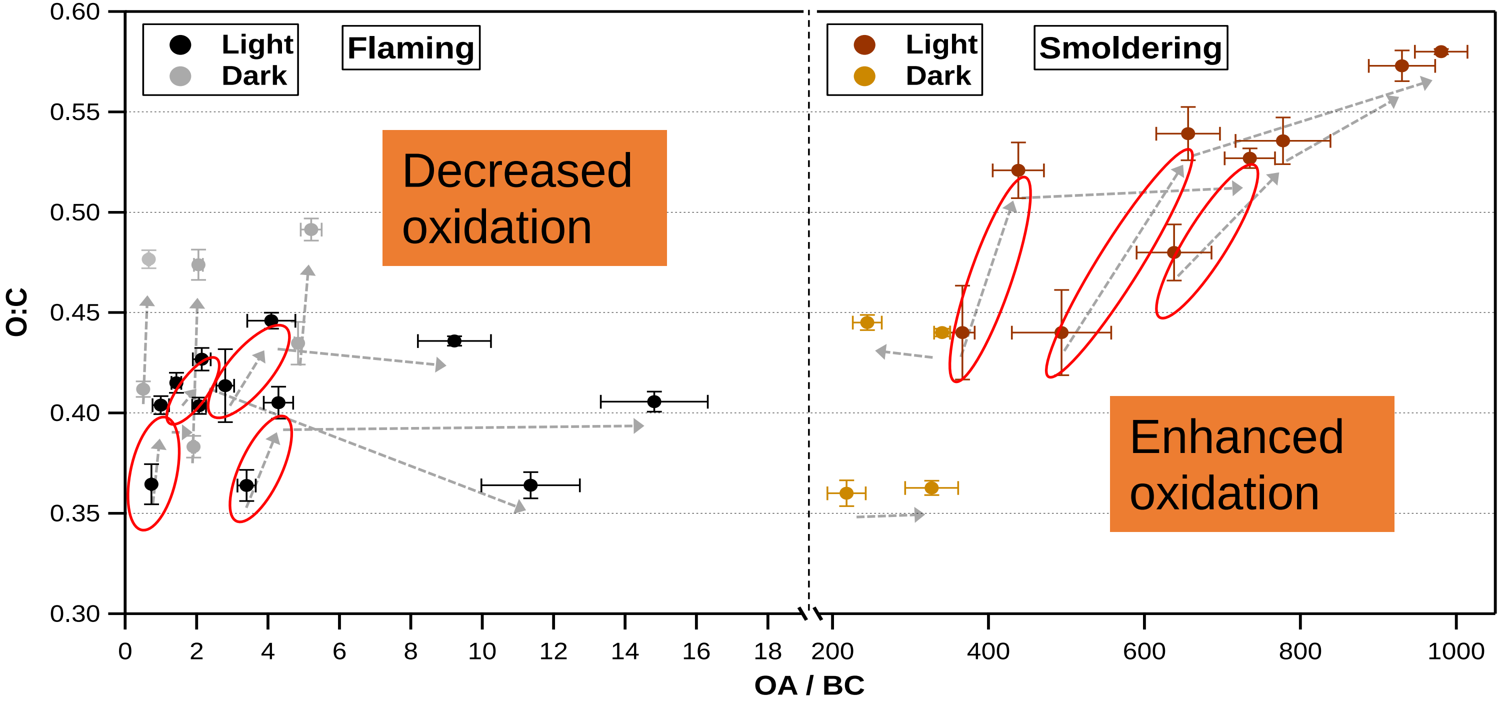


Organic aerosol oxidation

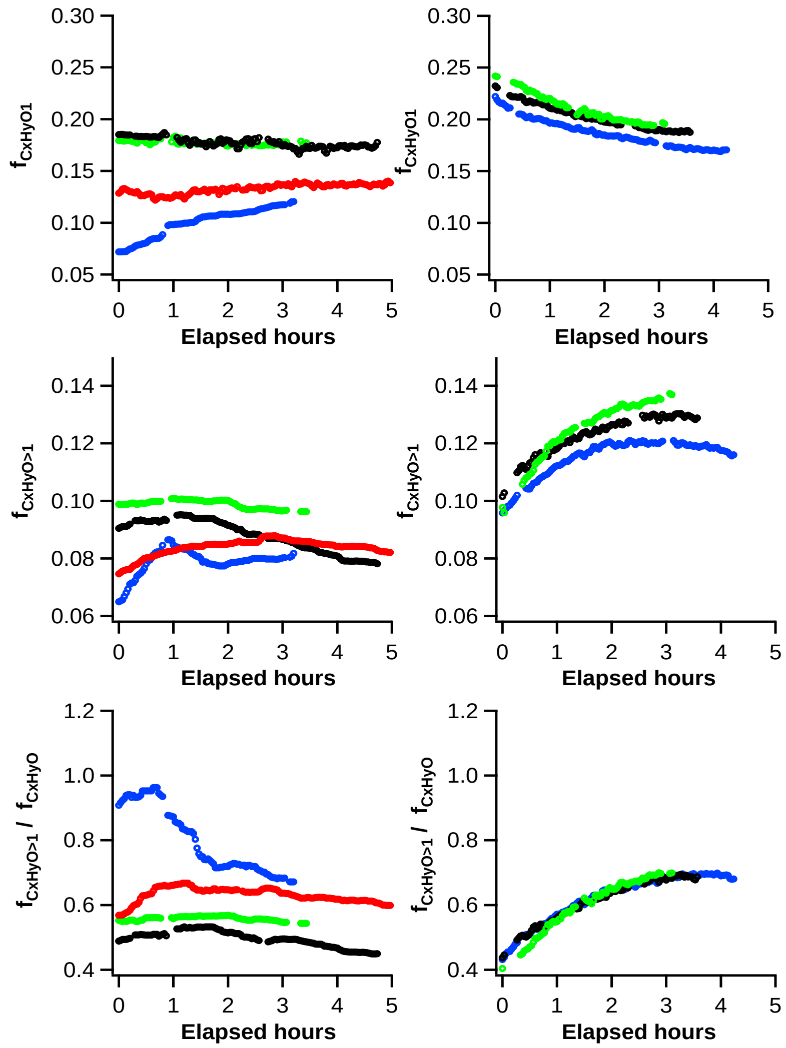
elemental O:C ratio



On OA/BC basis



Flaming **Smoldering**

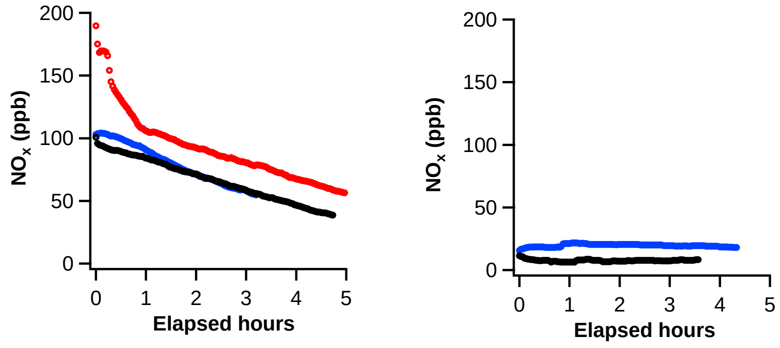


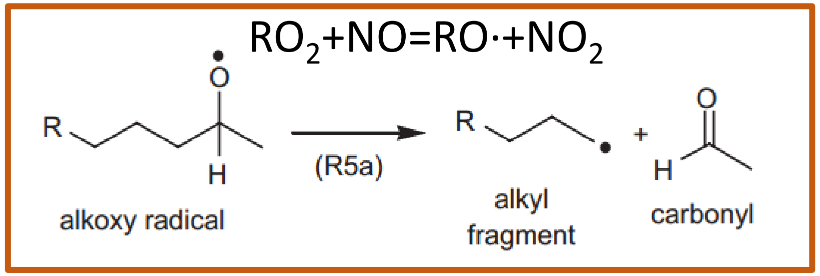
OA after evolution contained a higher elemental fraction of oxygen for SM than FL

Causality for higher oxidation of smoldering OA

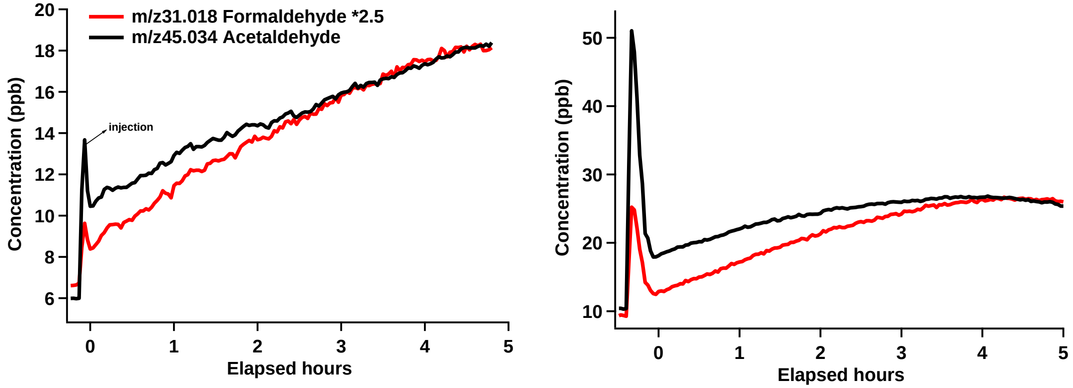
Fragmentation

Flaming Smoldering



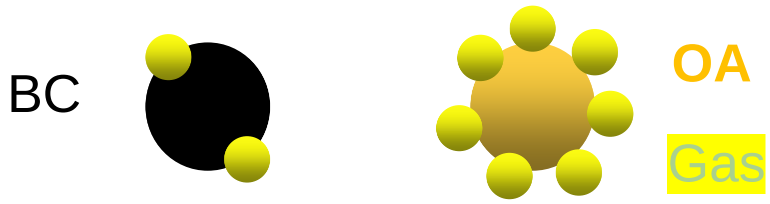


Flaming Smoldering



Seed particle effect

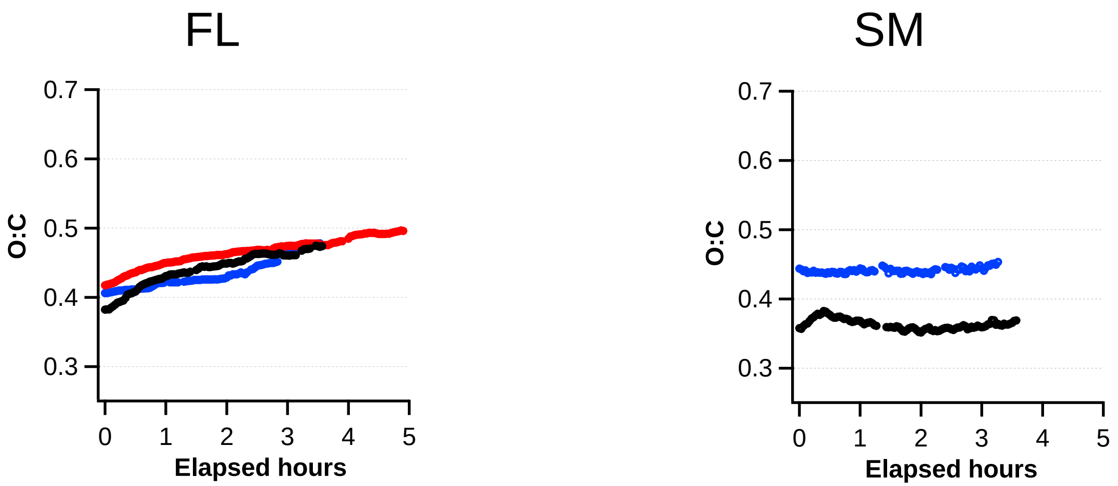
Flaming Smoldering

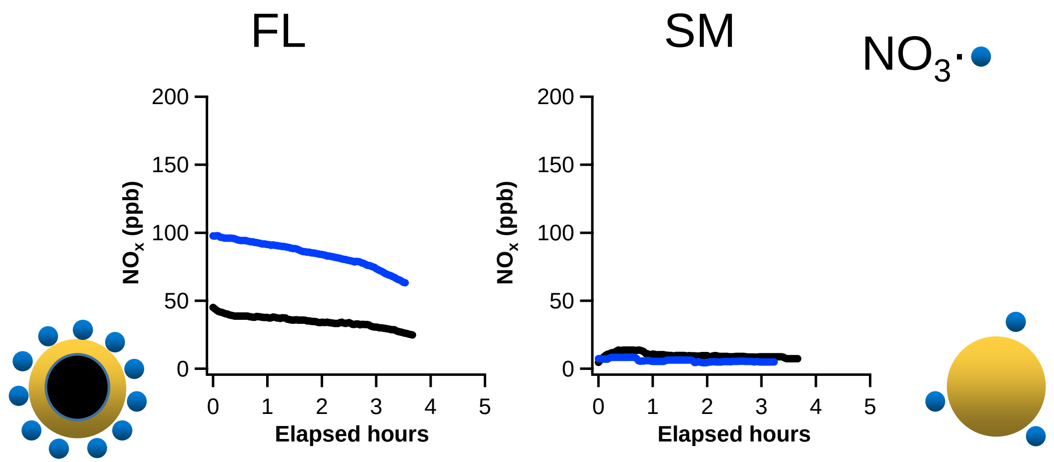


- SM plumes produced more VOCs and semi-volatile VOCs, which can be condensed in a shorter time.
- FL plumes was dominated by BC but SM was dominated by OA.

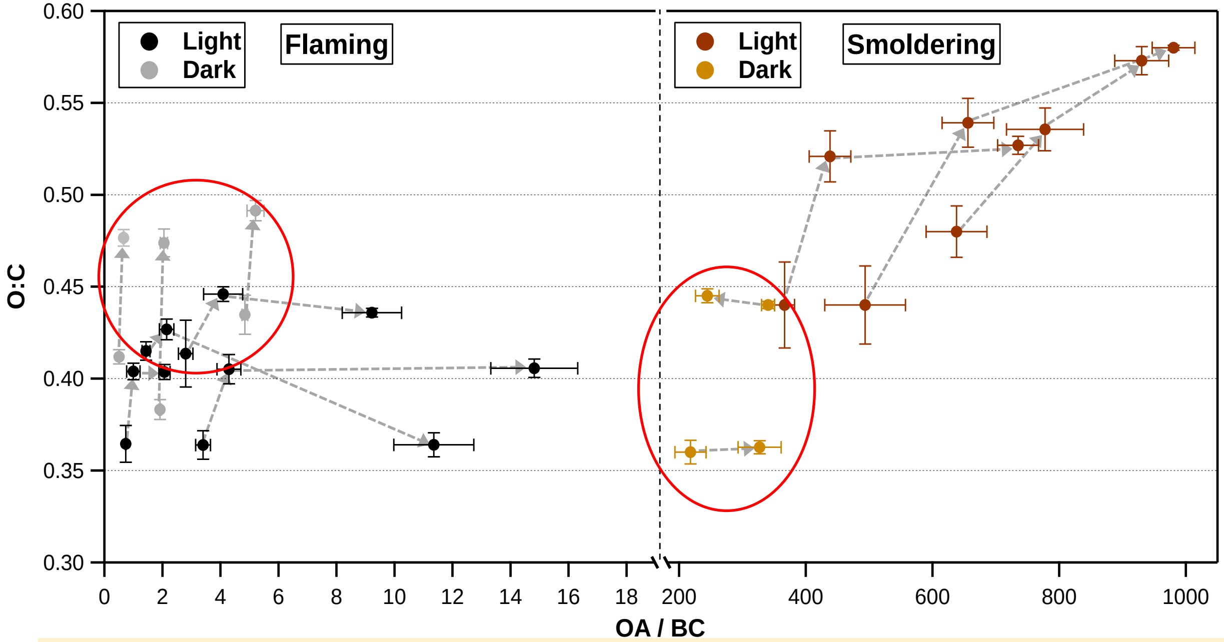
Organic particle substrate may absorb or adsorb more gas and result in enhancement of condensation.

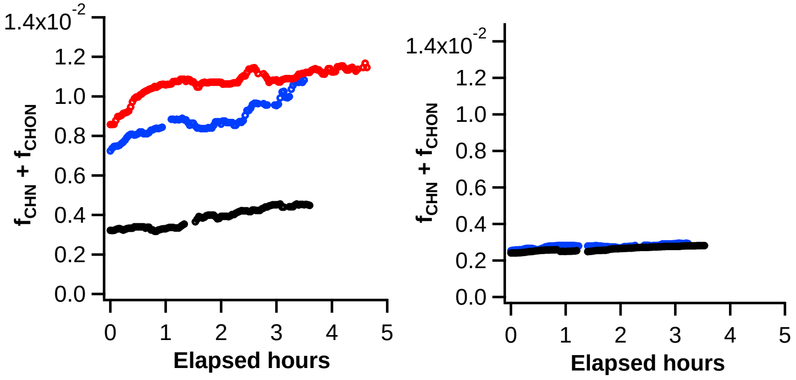
Dark ageing





Nitrate radicals (NO3·) oxidation



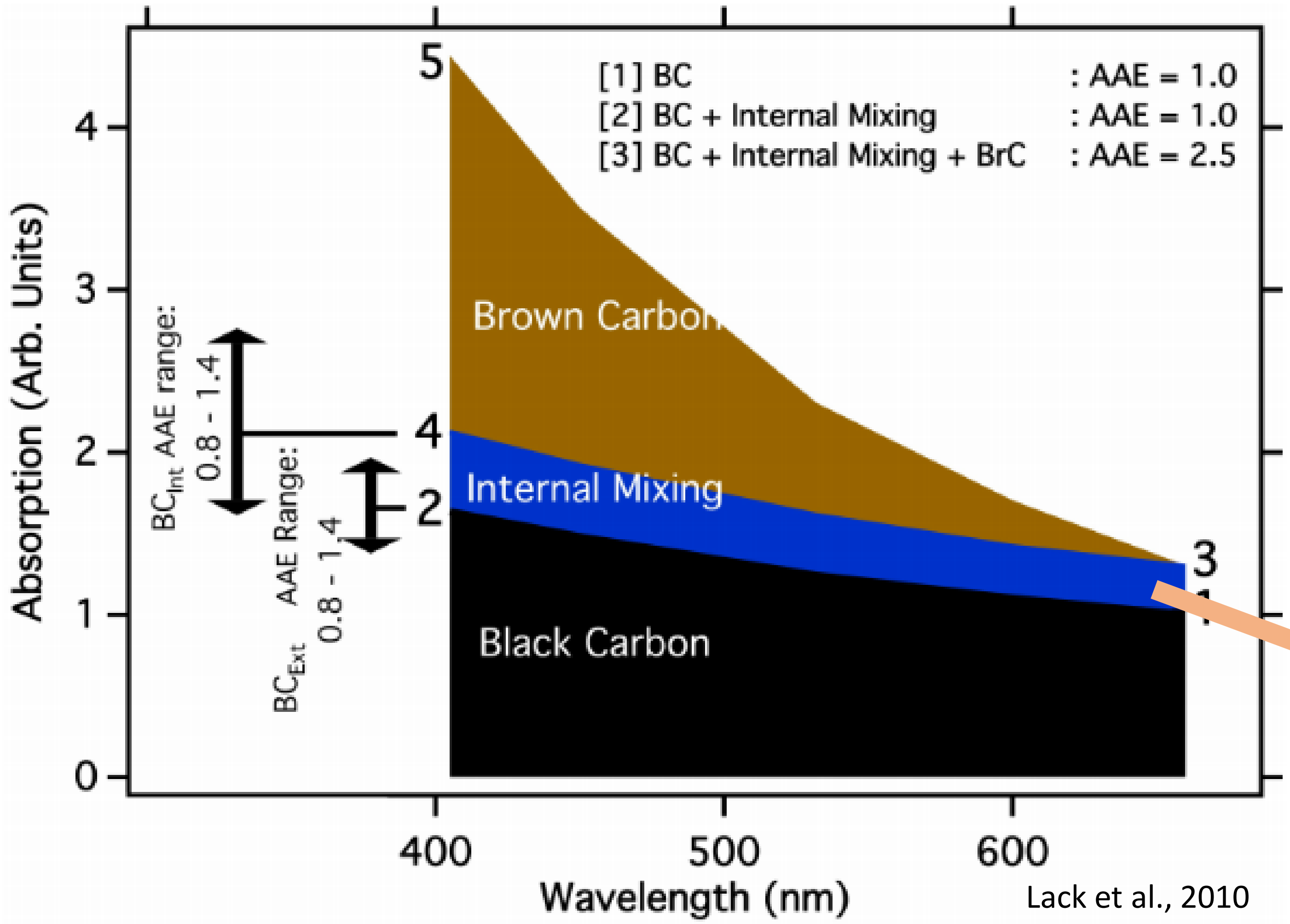


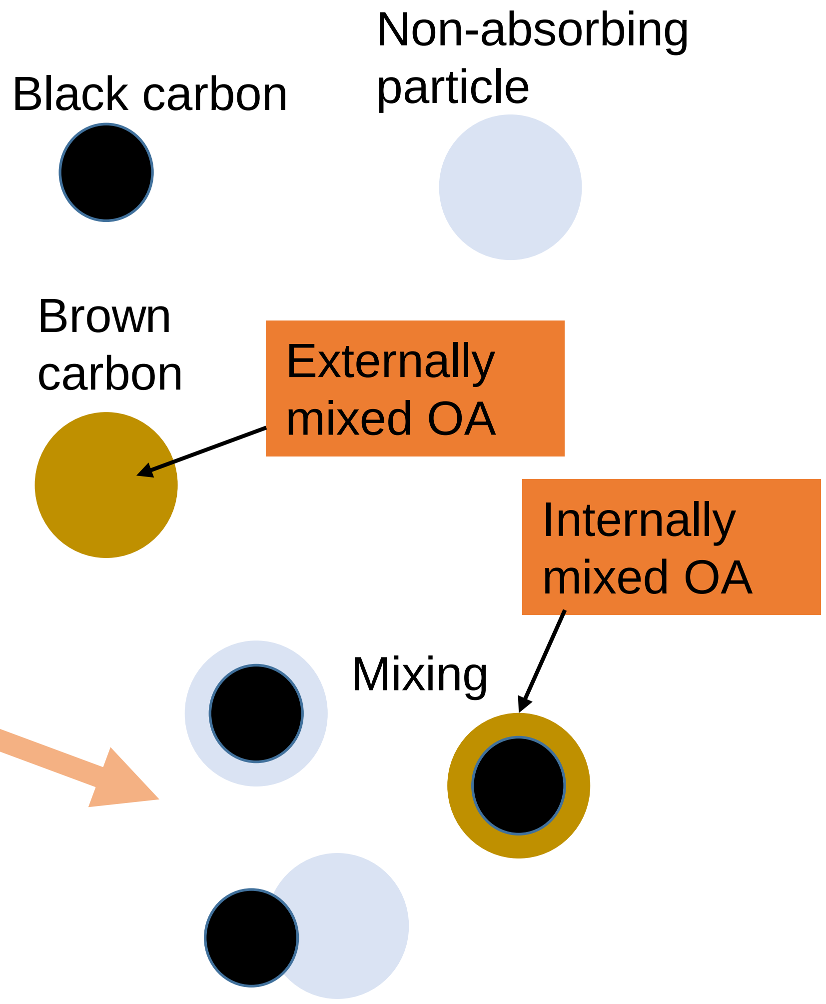
N-containing ions resulting from organic nitrate (ON)

No apparent OA/BC change during the dark.
O:C consistently increased by 0.09 for FL dark, while without apparent change for SM dark.

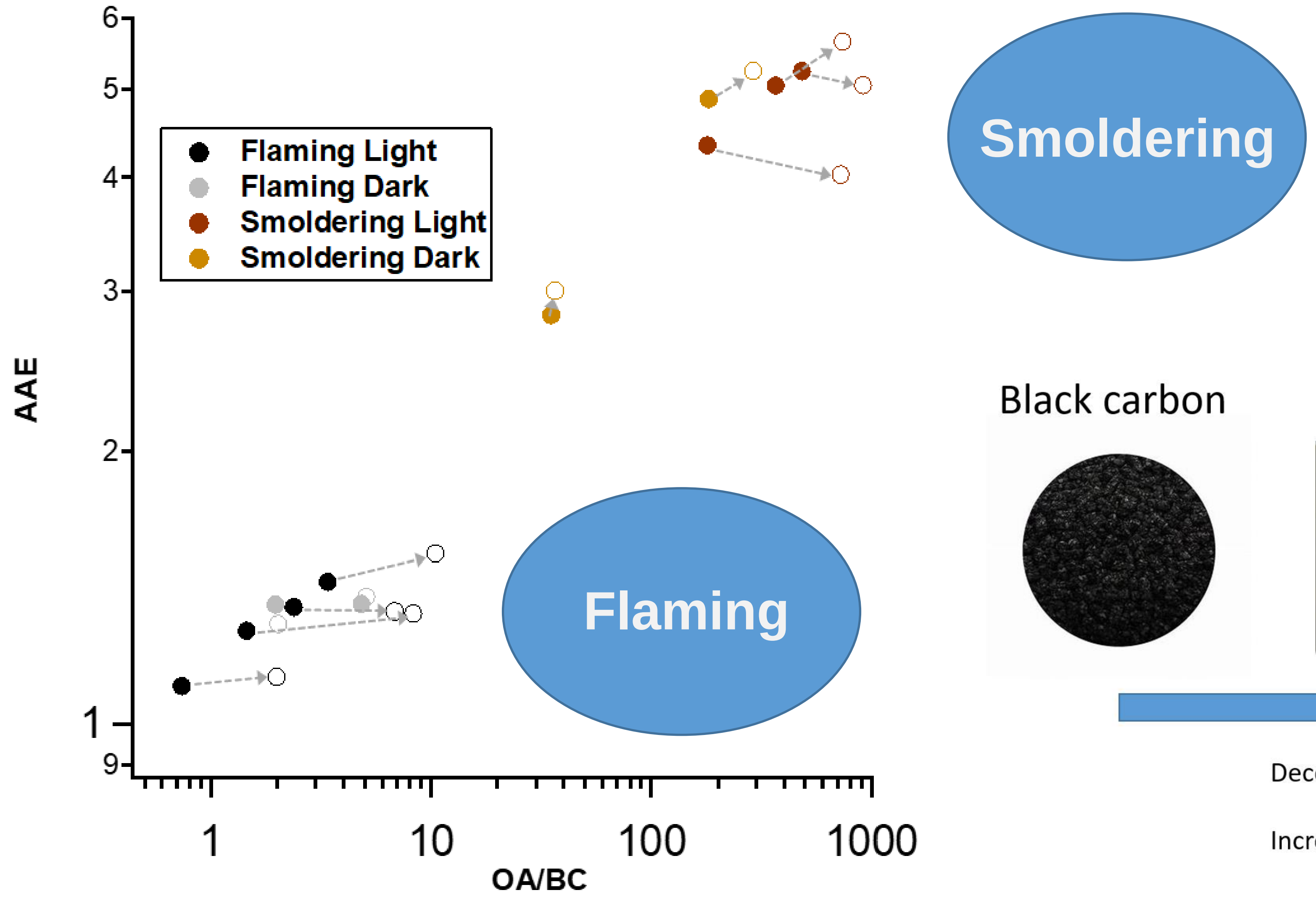
Evolution of Aerosol optical Properties

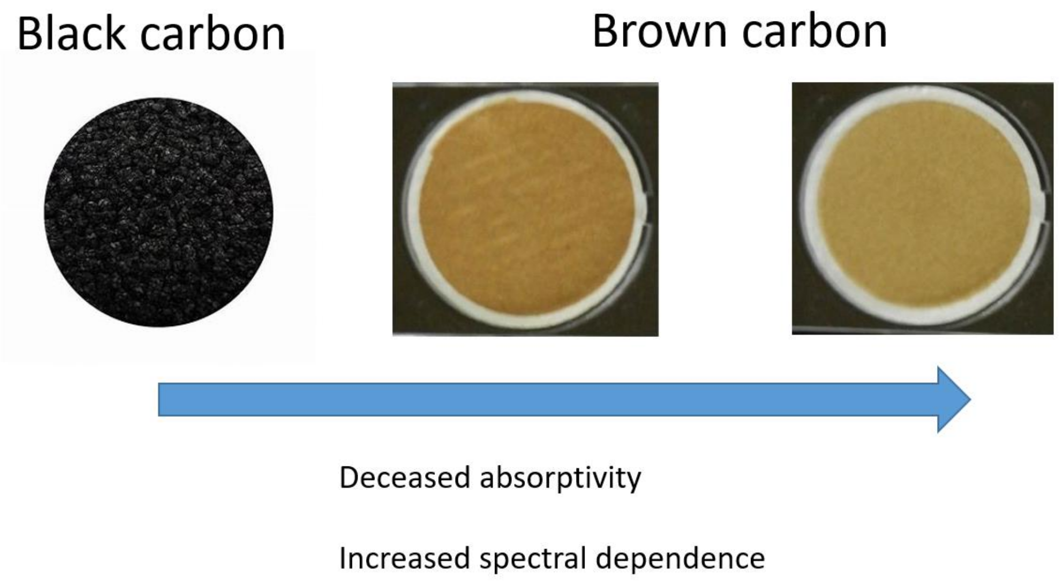
Spectral absorption of BC and BrC



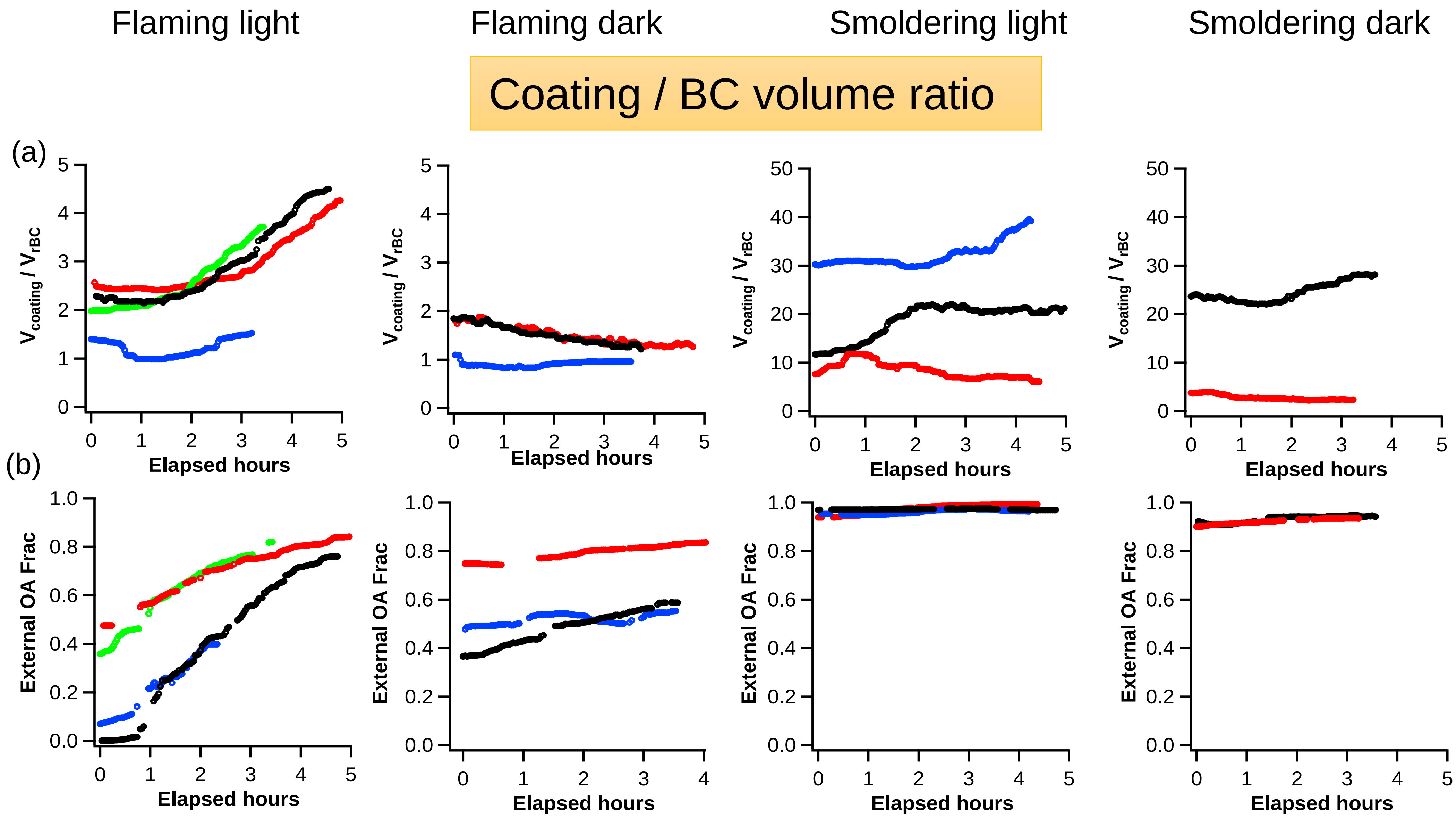


Contrasting features between burning phases





Evolution of mixed state of carbonaceous aerosols

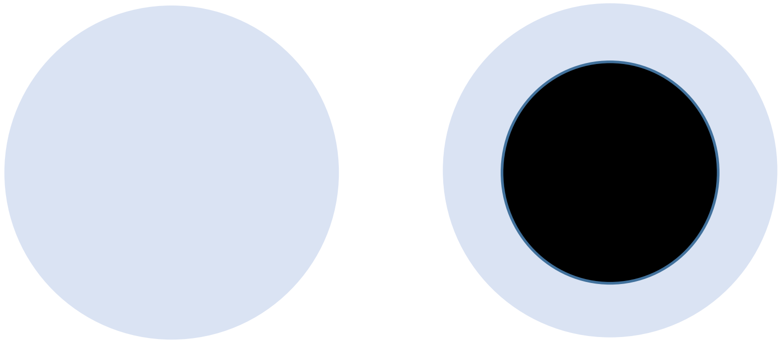


External OA fraction

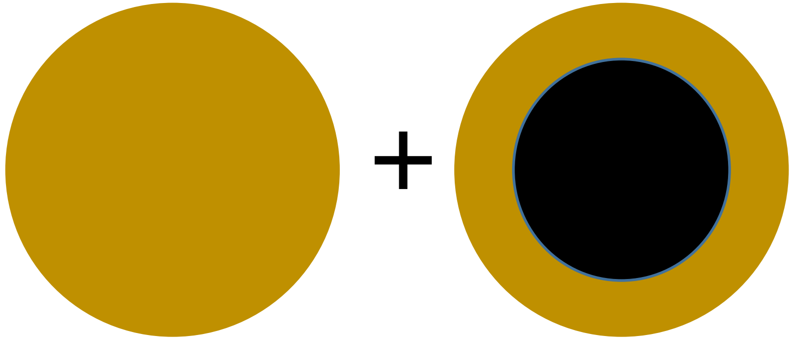
Mixed state of black carbon and brown carbon

Refractive index

RI=n+k *i*

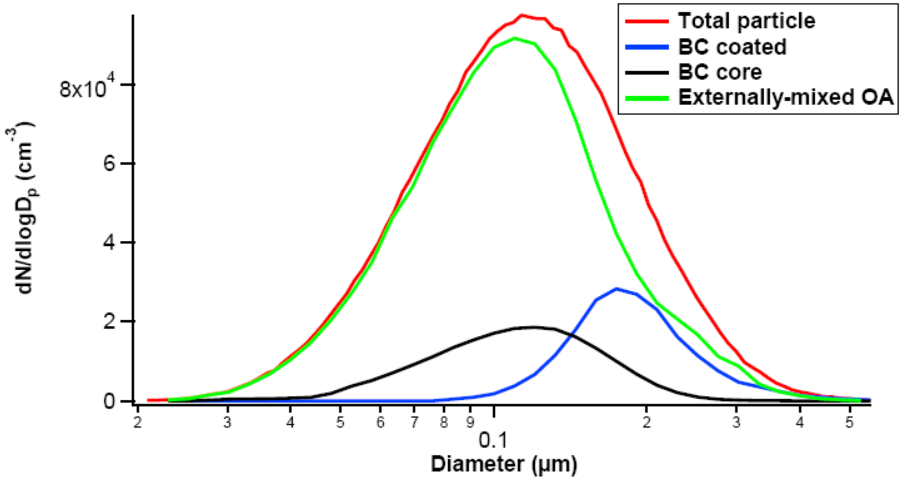


✕ k=0

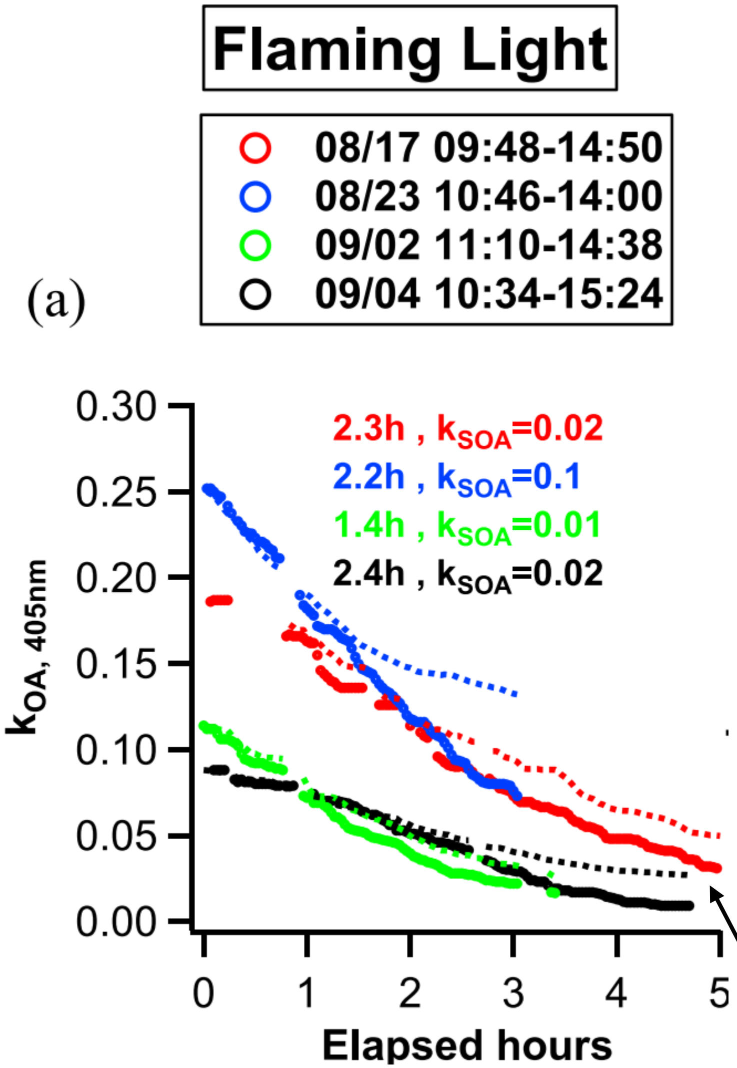


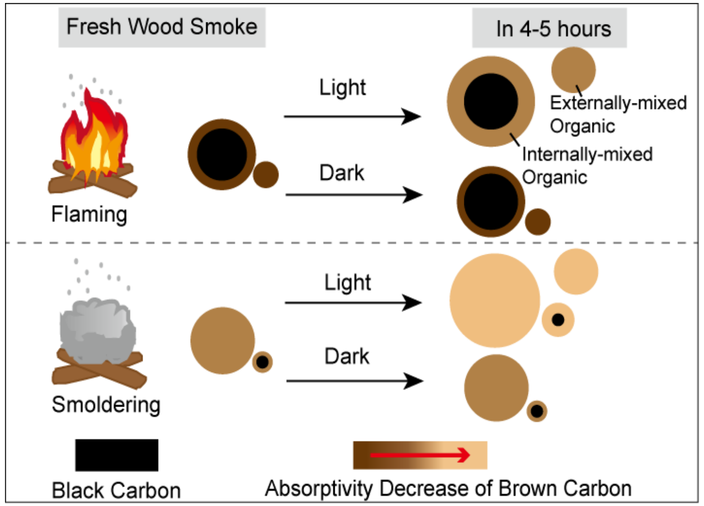
k>0

Externally-mixed OA   Internally-mixed OA



Absorptivity of OA





k of primary OA

k of secondary OA

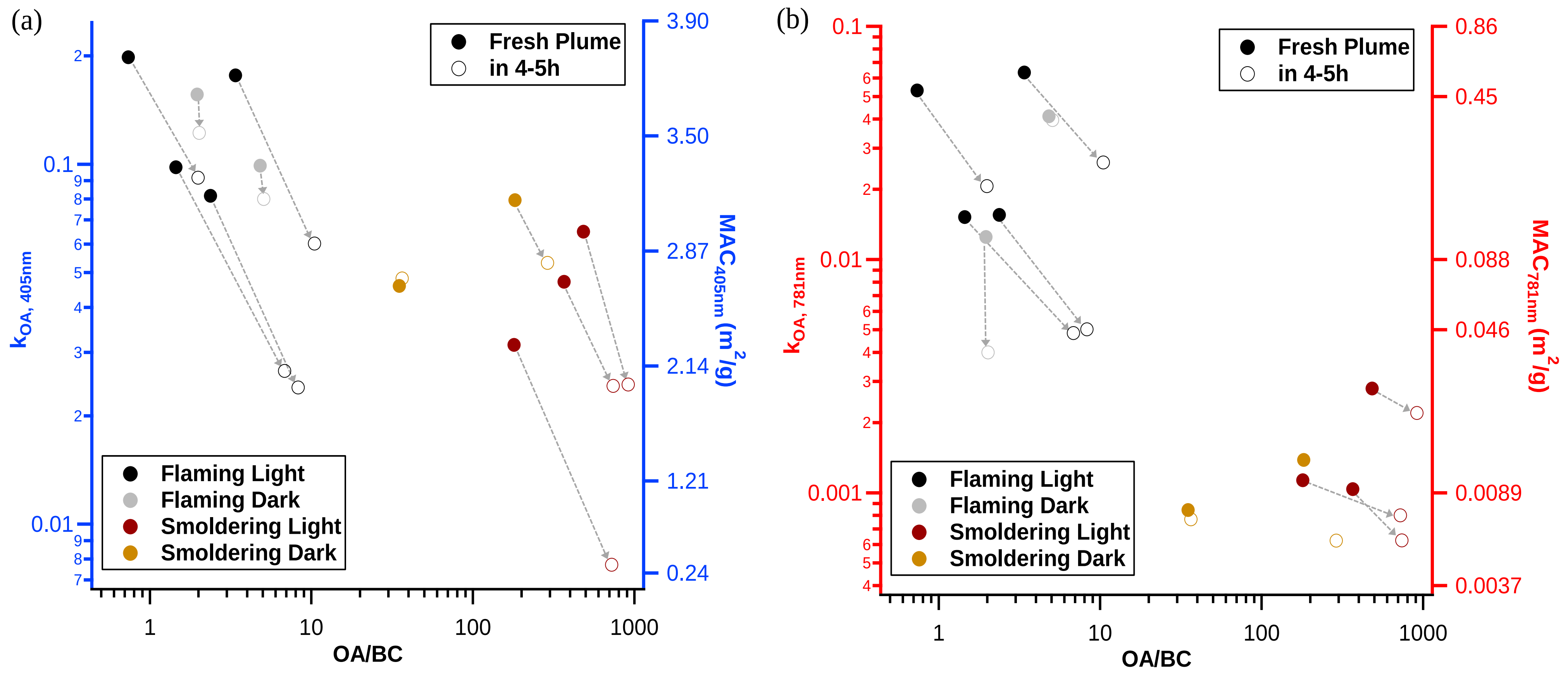
$$k_{\mathrm{OA,t}} = \varepsilon_{\mathrm{POA,t}} k_{\mathrm{POA}} + \varepsilon_{\mathrm{SOA,t}} k_{\mathrm{SOA}};$$

$$\frac{\varepsilon_{\mathrm{SOA,t}}}{\varepsilon_{\mathrm{POA,t}}} = \frac{(\mathrm{OA/BC})_t}{(\mathrm{OA/BC})_{\mathrm{initial}}} - 1$$

Volume ratio of secondary and primary OA

Dash lines: Decreased absorptivity due to SOA formation

Absorptivity mapped on OA/BC



OA from flaming conditions showed a higher absorptivity than from smoldering conditions.
Absorption parameters can be parameterized by OA / BC.

Conclusion

The near-source evolution of biomass burning emission from different burning phases should be considered.

- Smoldering plumes had faster secondary OA formation and higher oxidation than flaming.
- Absorbing OA (the brown carbon) from flaming conditions showed a higher absorptivity than from smoldering conditions.
- The absorptivity of OA had a half-decay time of 2−3 h due to SOA formation and photobleaching of chromophores.

Dantong Liu*, Siyuan Li, Dawei Hu, Shaofei Kong*, et al. Evolution of Aerosol Optical Properties from Wood Smoke in Real Atmosphere Influenced by Burning Phase and Solar Radiation, *ES&T*, 55(9), 5677–5688, 2021.

Siyuan Li, Dantong Liu*, Dawei Hu, et al.: Evolution of organic aerosol from wood smoke influenced by burning phase and solar radiation, *JGR*, 126(8), 2021.


THANK YOU.
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