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Secondary nanoparticles formation and composition from open and residential wood combustion

ETH-Conference on Combustion Generated Nanoparticles, 24 June 2021

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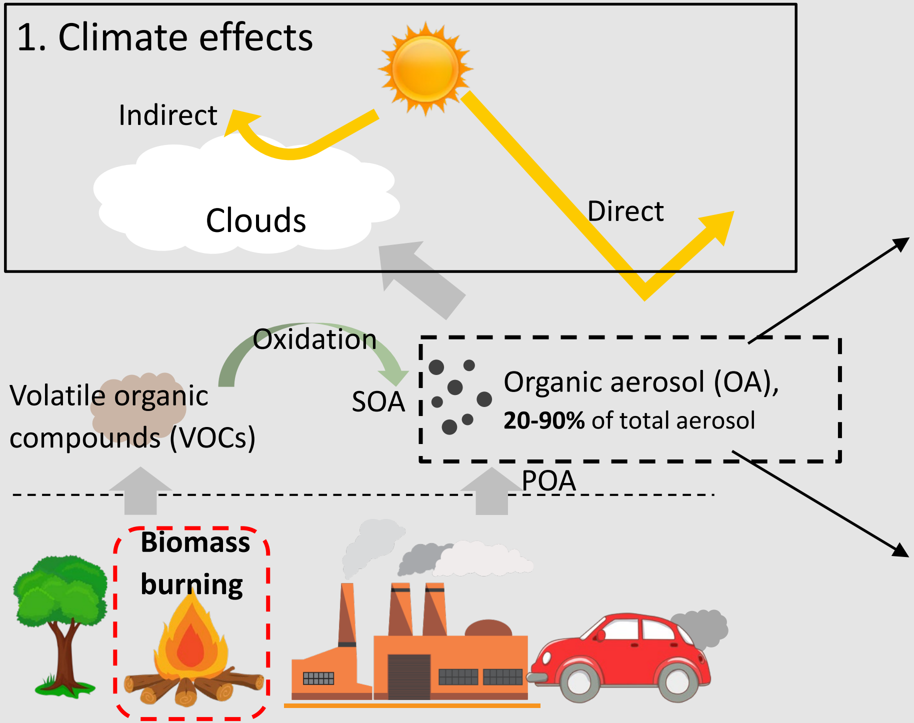
Biomass burning organic aerosols



Organic aerosol (OA),
20-90% of total aerosol

Biogenic and anthropogenic emissions

2. Air quality

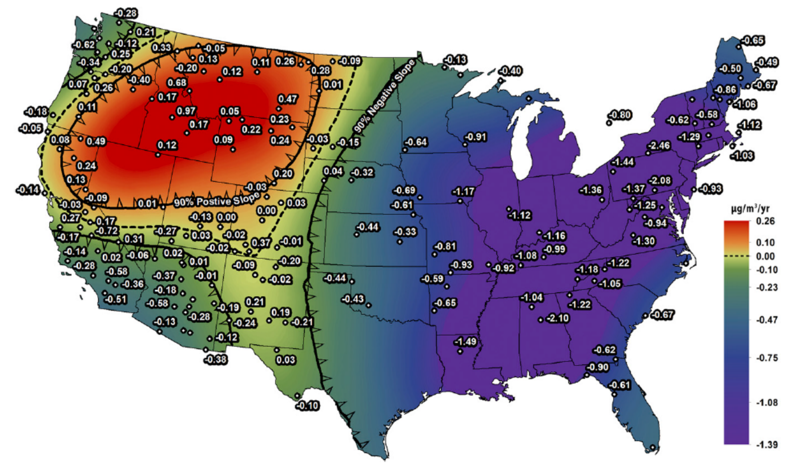
Aerosol can reduce **visibility** by scattering and absorbing lights

3. Human health

Exposure to aerosol is one of the leading causes for **premature death**

Forest fire and residential wood combustion

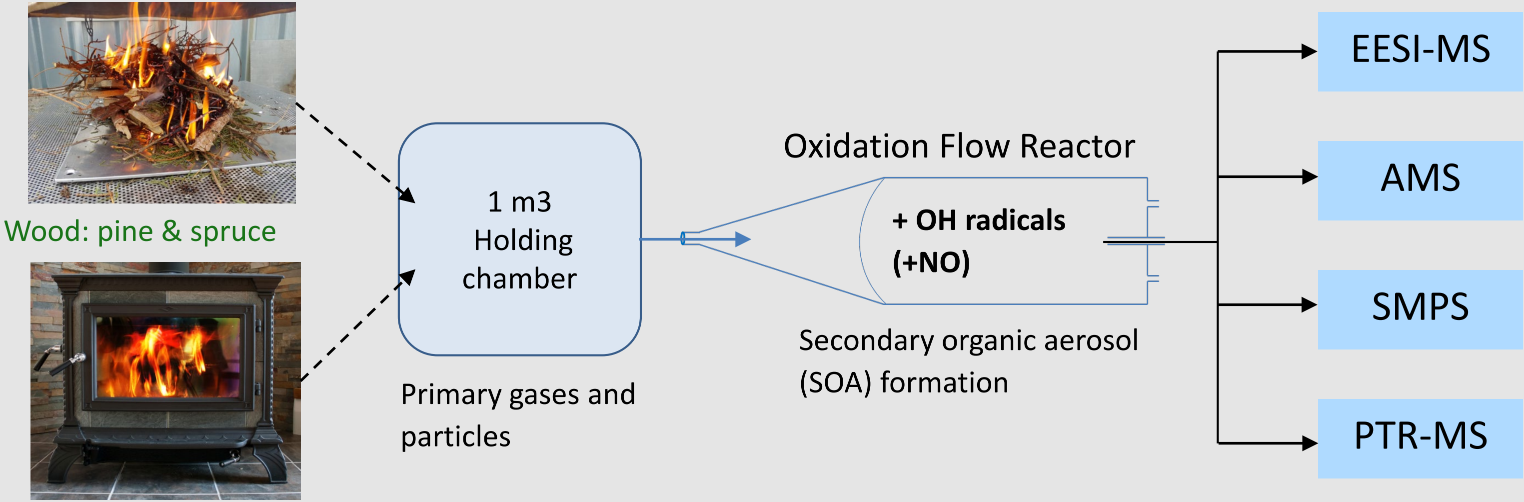
- Forest fire (wildfire) has increased in frequency, intensity, and area in the past few decades, and is predicted to continue to do so.
- Residential wood combustion is one of the major sources of organic aerosol in Europe during winter (15-60%).



McClure & Jaffe, PNAS, 2018, 7901.

Methods – Overview

Open burning (Forest fire) - twigs, barks and needles



Wood: pine & spruce

Residential wood burning - wood logs

1 m3 Holding chamber

Primary gases and particles

Oxidation Flow Reactor

+ OH radicals (+NO)

Secondary organic aerosol (SOA) formation

EESI-MS

AMS

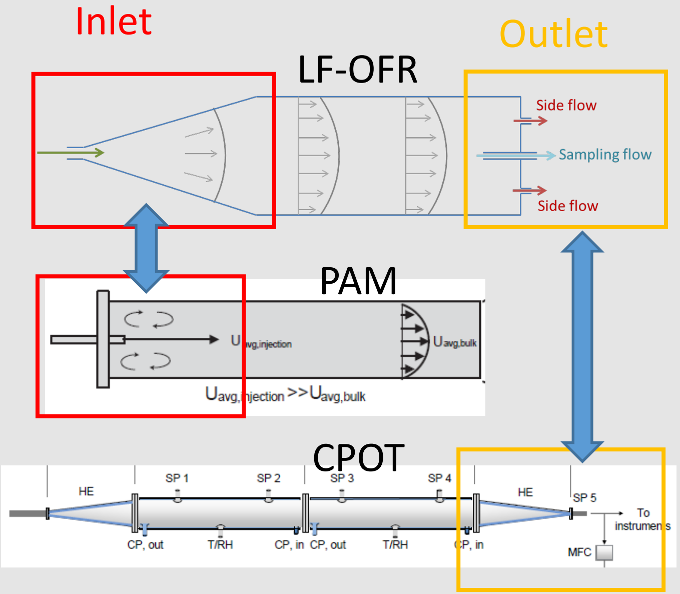
SMPS

PTR-MS

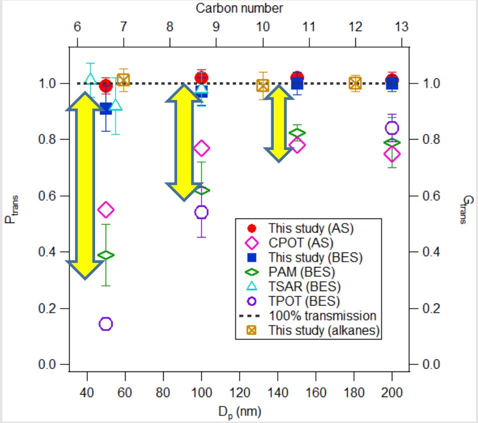
Objectives:

- Quantify the SOA formation at a wide range of photochemical ages.
- Molecular-level SOA composition and its evolution over oxidation.

Oxidation Flow Reactor (OFR)

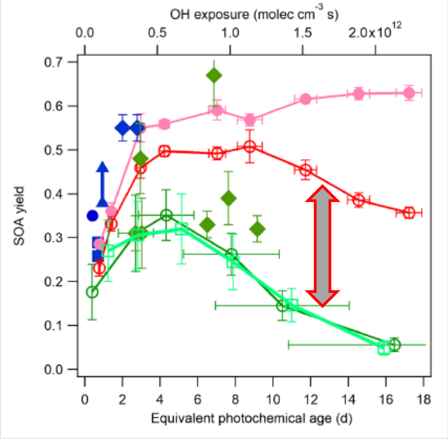


Wall loss



Li et al., Atmos Chem Phys, 2019, 9715.

SOA yield



Blue: smog chambers; Red: LF-OFR; Green: other OFRs.

Laminar-flow OFR (formerly ECCC-OFR):

- Improved design can reduce jetting and recirculation in the reactor.
- Reduced wall loss and enhanced SOA yields.

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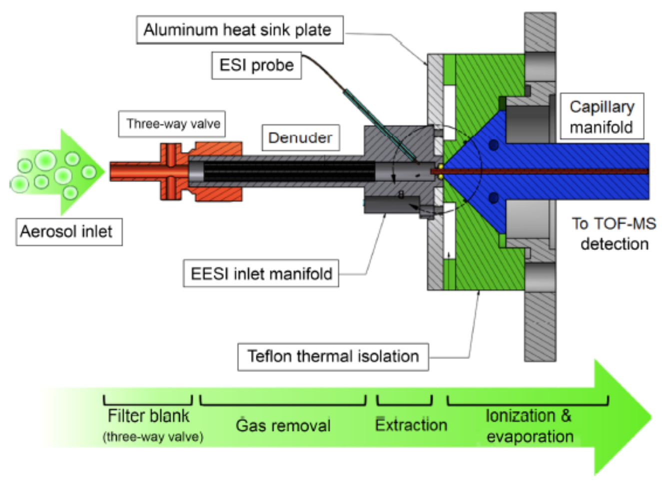
EESI-MS

- Extractive electrospray ionization mass spectrometry (**EESI-MS**) is the cutting-edge technique to characterize molecular-level composition of aerosols.



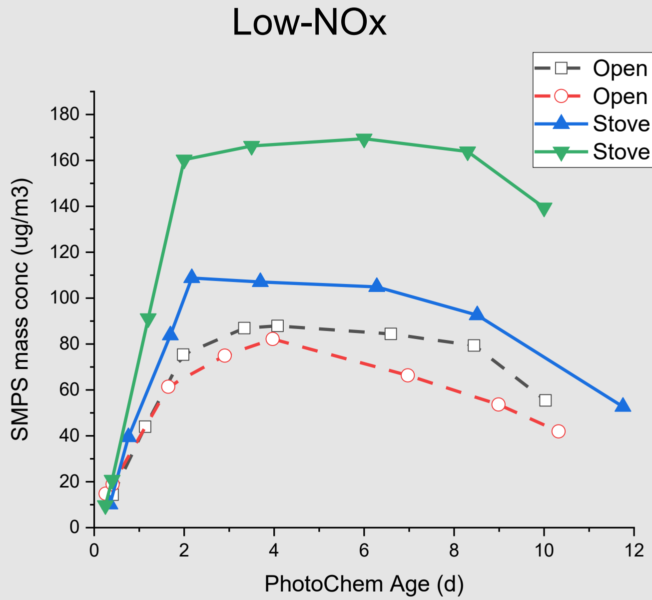
Lopez-Hilfiker et al., Atmos Meas Tech, 2019, 4867.

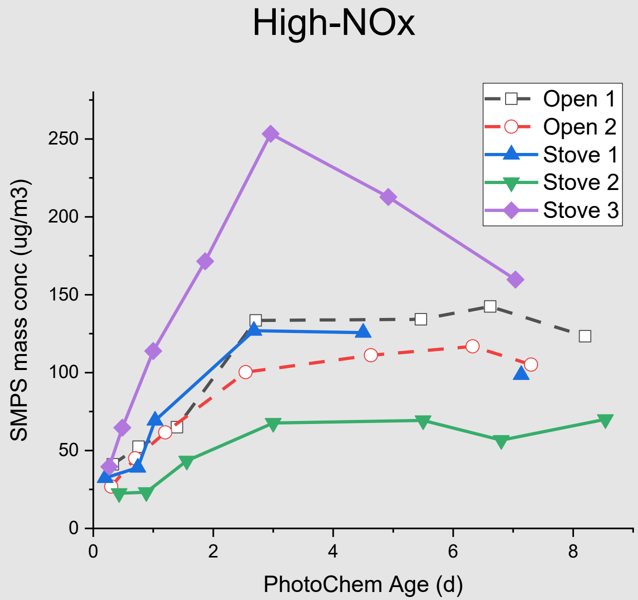
Technique	Time resolution	Molecular resolution	Fragmentation
ESI-MS*	🟥 Hours (off-line)	🟩 High	🟩 Minimal
AMS*	🟩 Seconds to minutes	🟥 Low	🟥 High due to EI
TD-CIMS*	🟨 Minutes to hours	🟩 High	🟨 Medium (high temperature)
EESI-MS	🟩 **Seconds**	🟩 **High**	🟩 **Minimal**

Red: Bad; Yellow: Neutral; Green: Good

*ESI-MS: Electrospray ionization MS; AMS: Aerosol MS; TD-CIMS: Thermo-desorption chemical ionization MS.

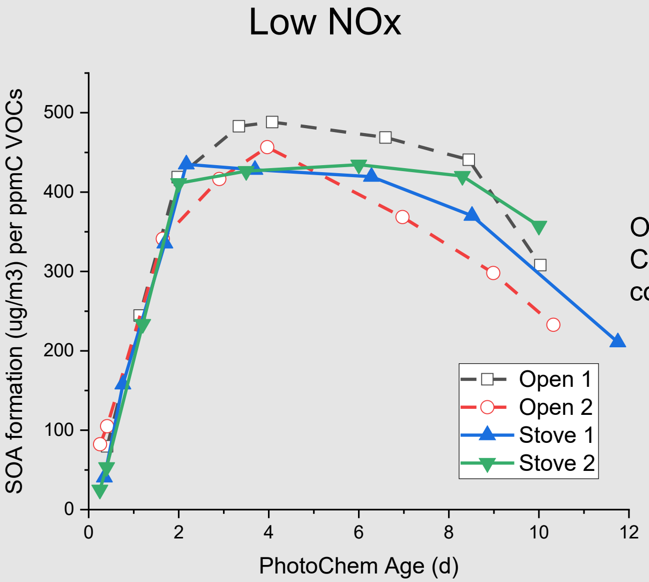
SOA formation under different NOx conditions



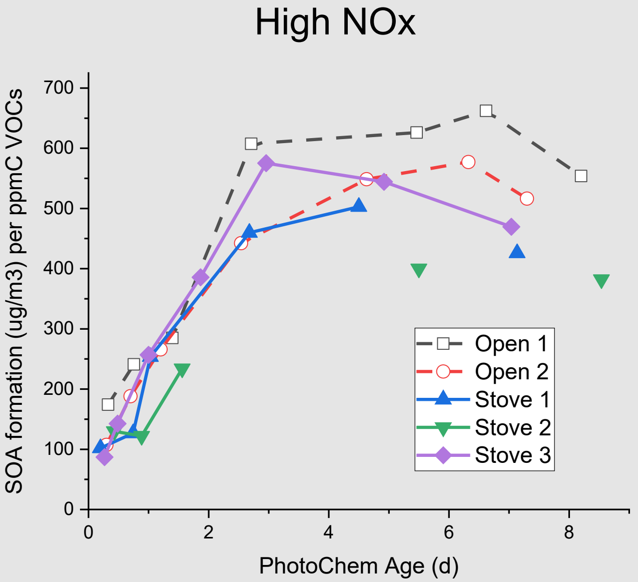


- SOA mass first increases and then decreases with increasing photochemical age.
- SOA production varies between experiments.

SOA formation normalized by VOCs

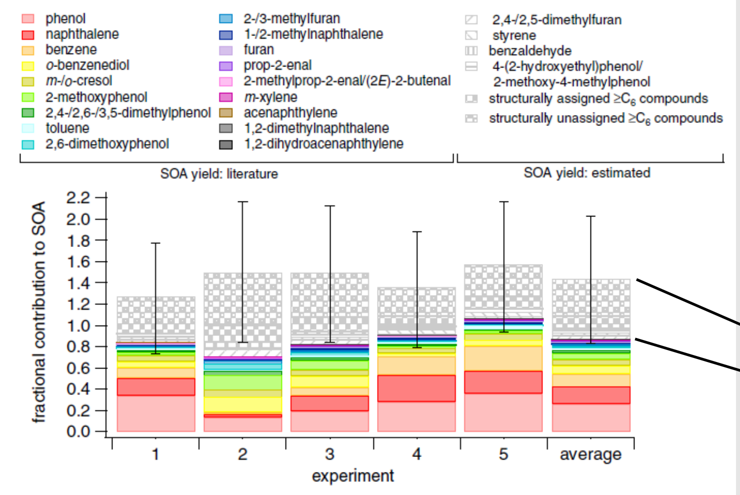


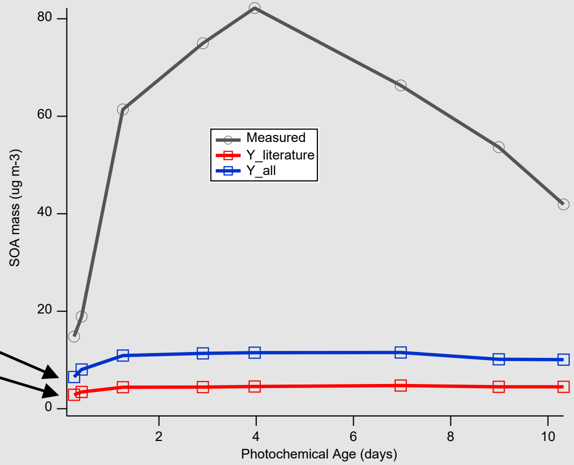
Only consider C# ≥ 4 compounds



- Open and stove burning are very similar after normalization.
- High-NOx condition slightly increases the SOA formation.

Using measured VOCs to calculate SOA

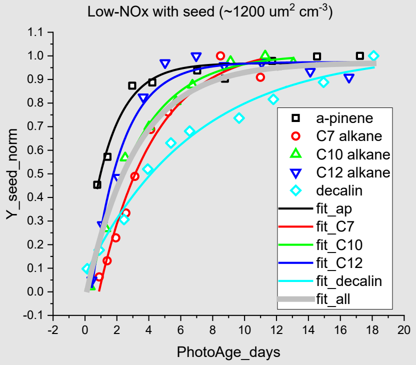


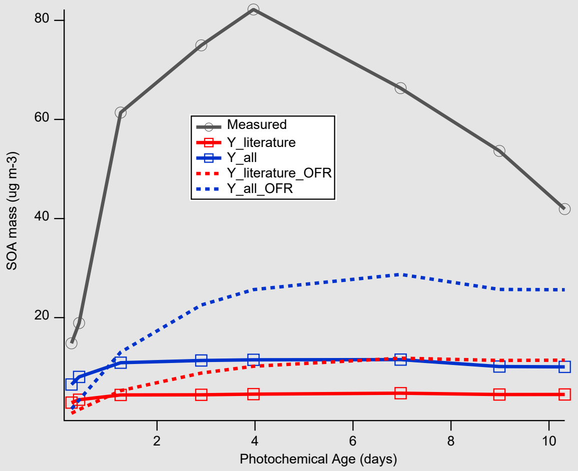


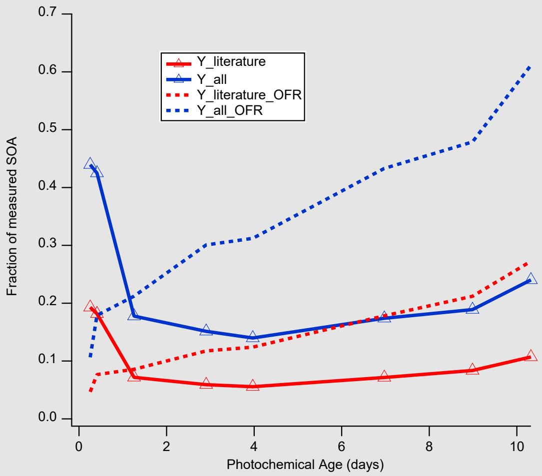
Bruns et al., Sci Rep, 2016, 27881.

Measured VOCs can only explain 10-20% of the formed SOA in the OFR, very different from previous smog chamber study.

Using measured VOCs to calculate SOA







Possible reasons:

- OFRs and smog chambers have different photochemical ages.
- The contribution of unmeasured low-volatility compounds (e.g., IVOCs).

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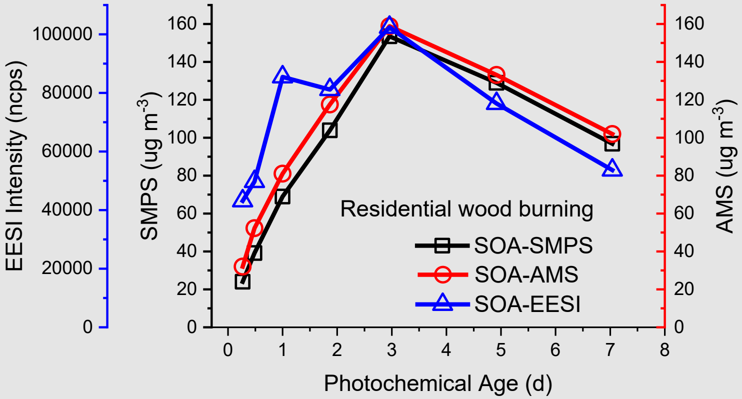
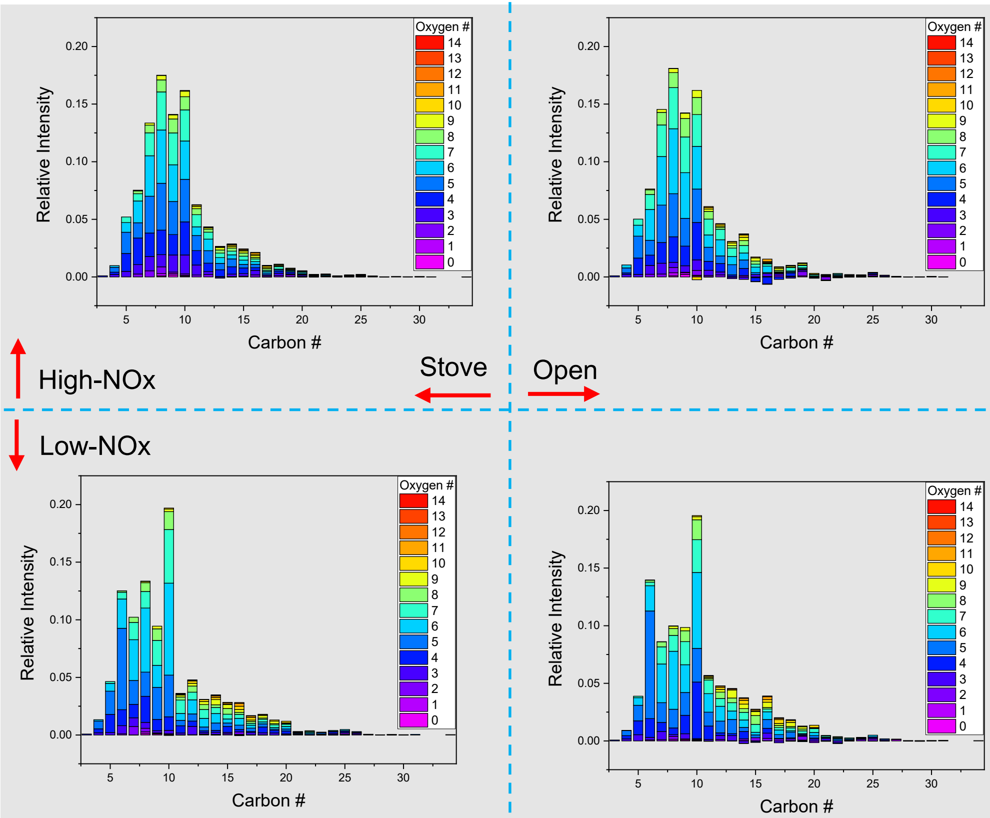
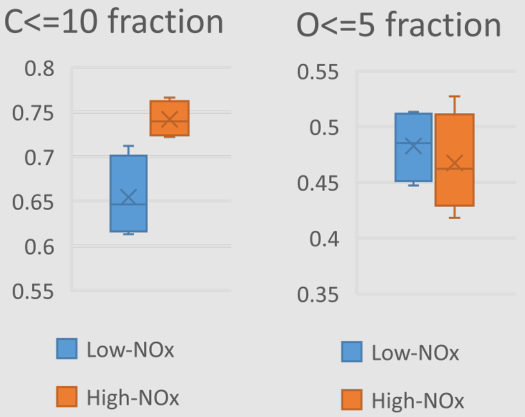
SMPS vs AMS vs EESI



- AMS and SMPS have very similar trend in SOA concentration.
- EESI is slightly different, likely due to different sensitivities of different category of products.

Carbon and Oxygen distribution





Snapshots at ~1-2 photochemical days.

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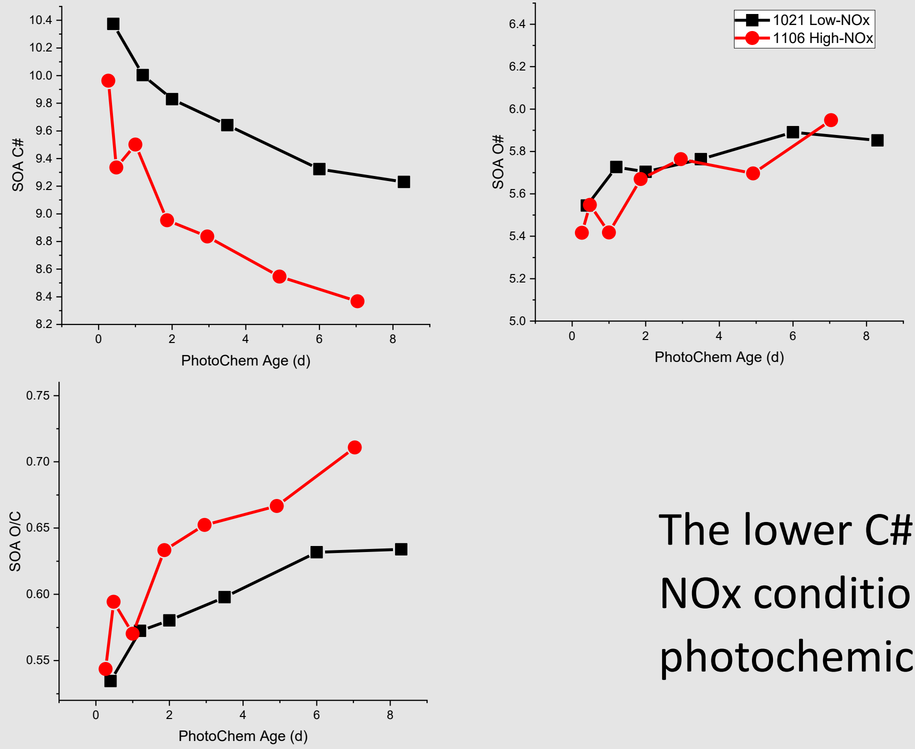
Average C#, O#, and O/C



The lower C# and higher O/C under high-NOx condition is observed for all photochemical ages.

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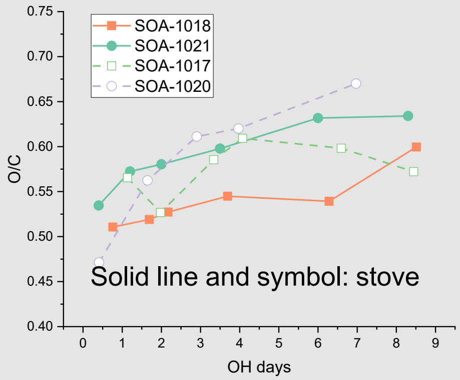
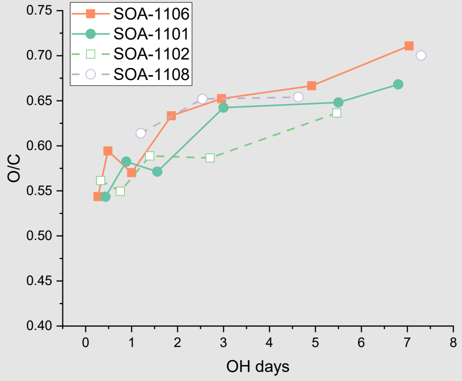
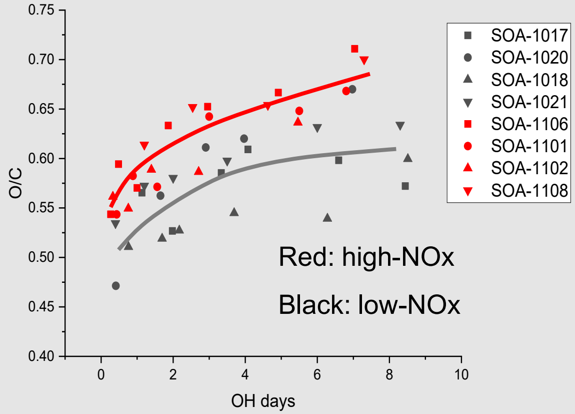
Average O/C


Low-NOx

Solid line and symbol: stove


High-NOx


Red: high-NOx

Black: low-NOx

- No significant difference between open and stove burning.
- Higher O/C under high-NOx condition.

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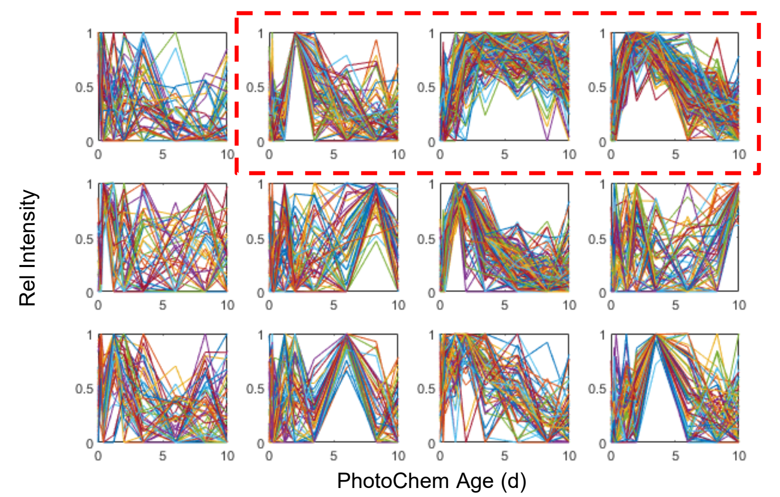
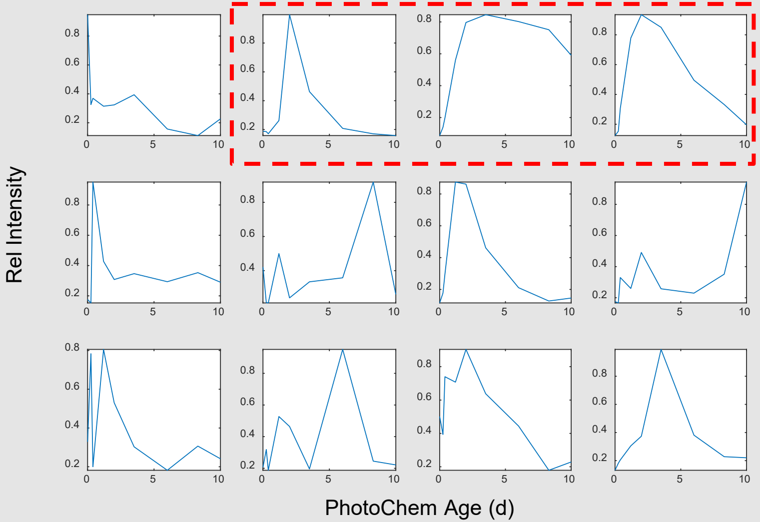
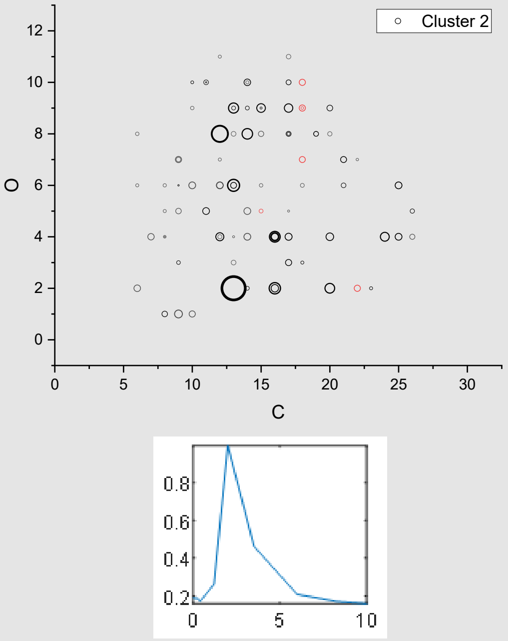
Clustering analysis

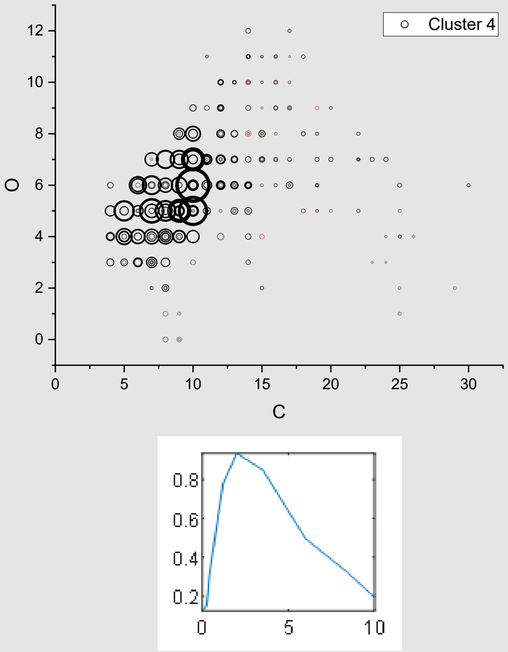


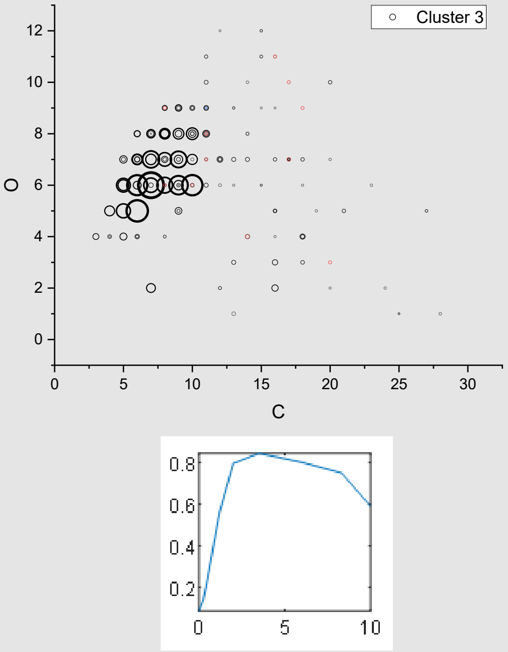


- 12 clusters based on k-means clustering analysis.
- They evolve differently with increasing photochemical age.

Clustering analysis – O# vs C#

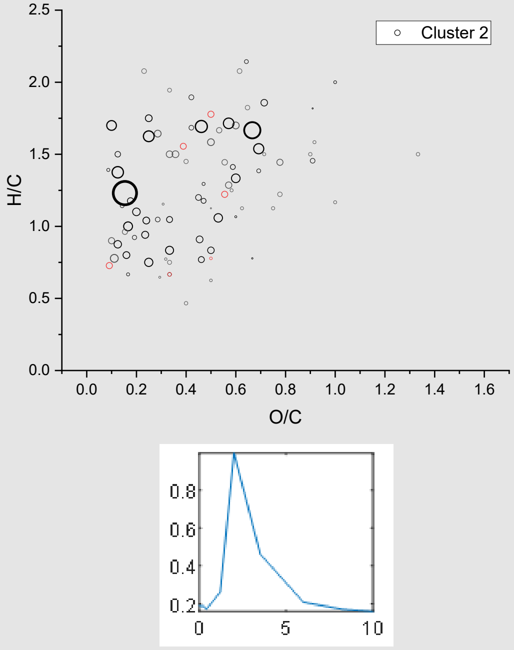


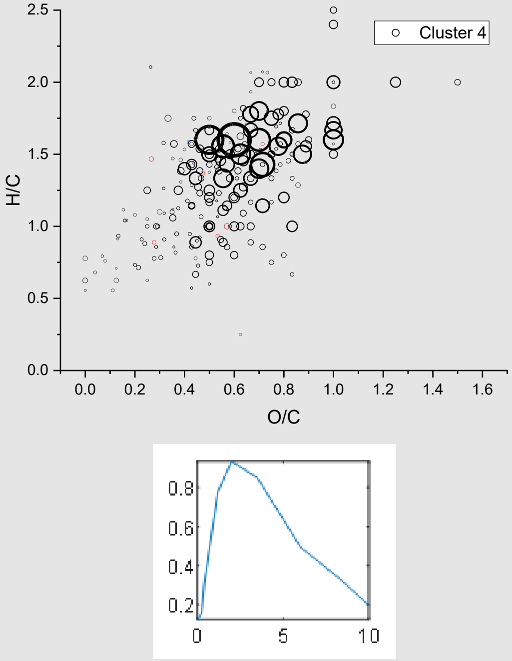


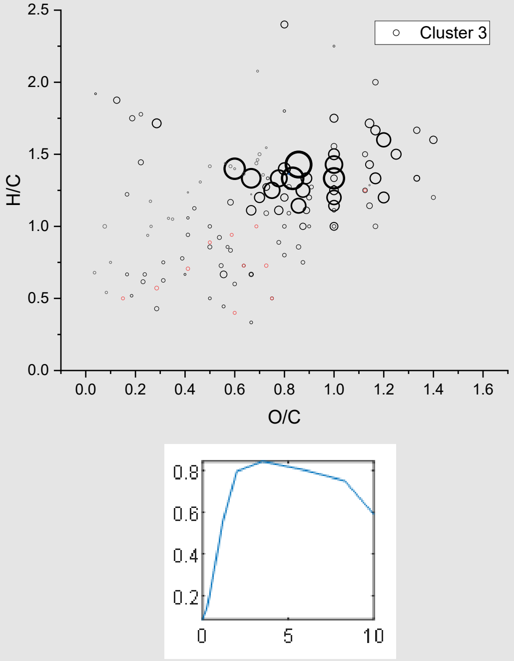


Later-generation products have lower C# and higher O#.

Clustering analysis – H/C vs O/C







Later-generation products have lower H/C and higher O/C.

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Conclusion and outlook

- Measured VOCs cannot explain the observed SOA production in OFR. Likely due to the contribution of IVOCs.
- High-NOx condition enhances O/C of SOA by reducing carbon number (molecular size).
- Using EESI-MS, we can track the molecular information of different generation of products in the oxidation of biomass burning emissions.

Outlook:

- Using other techniques (e.g. VOCUS PTR-MS) to quantify IVOCs and their contributions to SOA production.
- Performing PMF analysis with EESI and AMS data, and compare with the clustering analysis results.

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Acknowledgments

- Andre Prevot
- Imad El Haddad
- David Bell
- Jun Zhang
- Jay Slowik
- Tiantian Wang
- John Liggio

- Laboratory of Atmospheric Chemistry (LAC, PSI)
- Swiss National Science Foundation (SNSF)
- Marie Sklodowska-Curie PSI-Fellow

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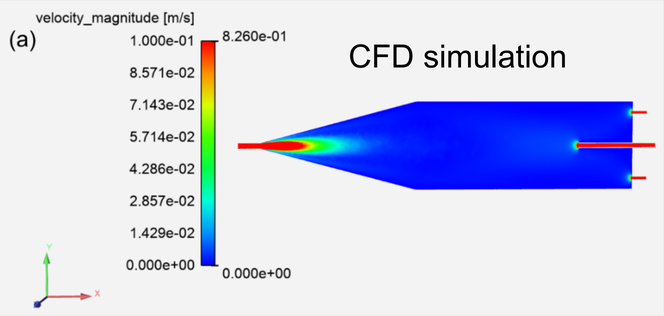
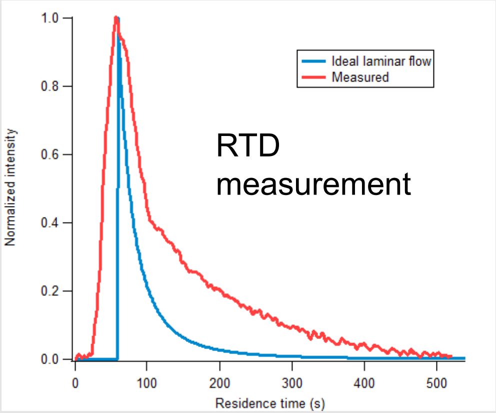
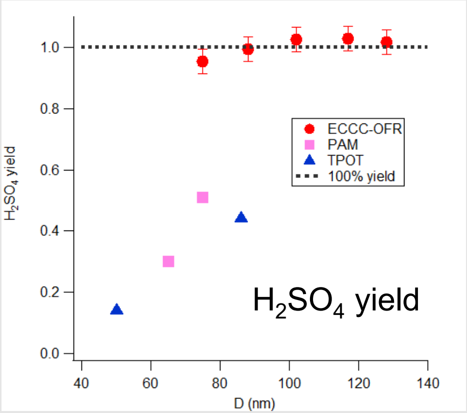
Supplementary Slides

LF-OFR



Li et al., Atmos Chem Phys, 2019, 9715.





- CFD simulation indicates that the velocity in LF-OFR is generally uniform.
- The residence time distribution (RTD) is similar to that of ideal laminar flow.
- The H_2SO_4 yield from SO_2 oxidation is ~100%, significantly higher than other OFRs.

Number of clusters