Current Capabilities and Gaps in Organic Aerosol Modeling

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In collaboration with
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Florian Couvidat, Victor Lannuque (INERIS) and the CAMAERA Team

Meeting of the EMEP Task Force on Measurement and Modeling
April 28-29, 2026

Norwegian Meteorological Institute

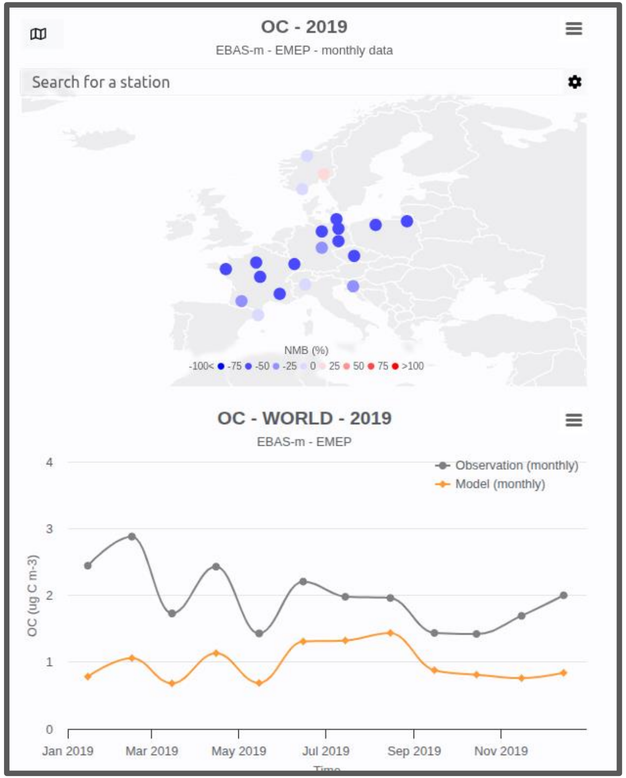
EASVOLEE

CAMAERA

emep

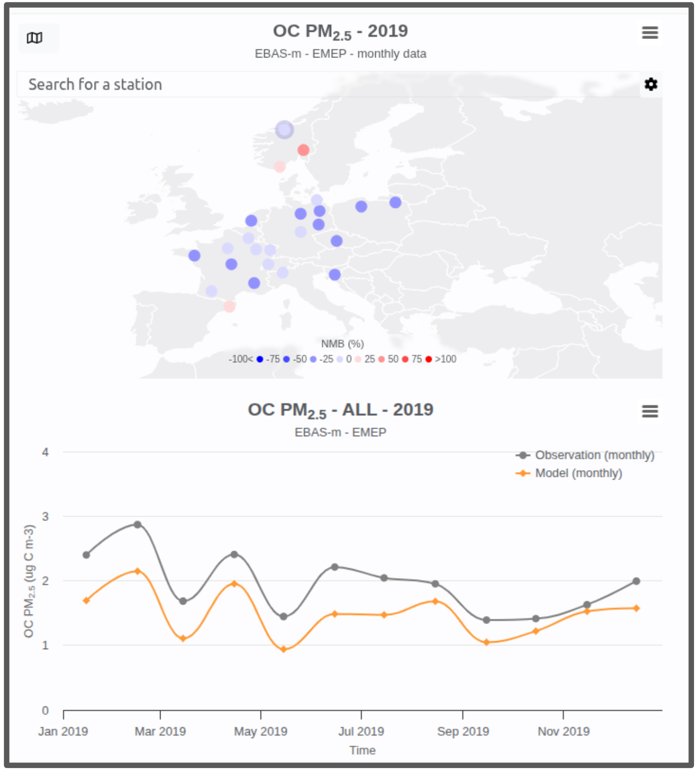
LRTAP Long-range Transboundary Air Pollution

Organic Aerosol Performance for Operational EMEP Model



2019 results from 2021

(before introducing ‘condensables’)



2019 results from 2025

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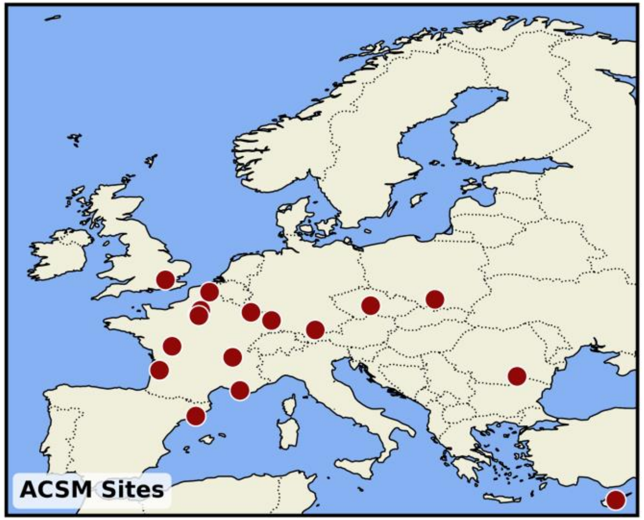
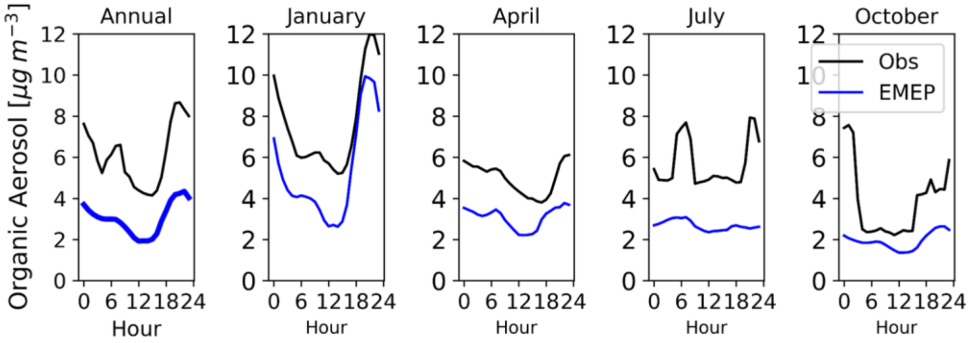
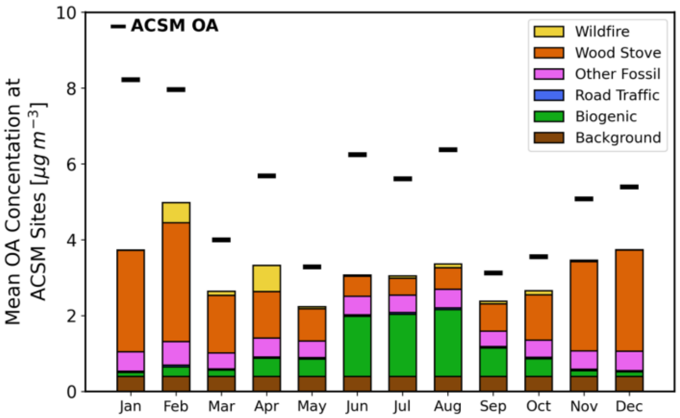




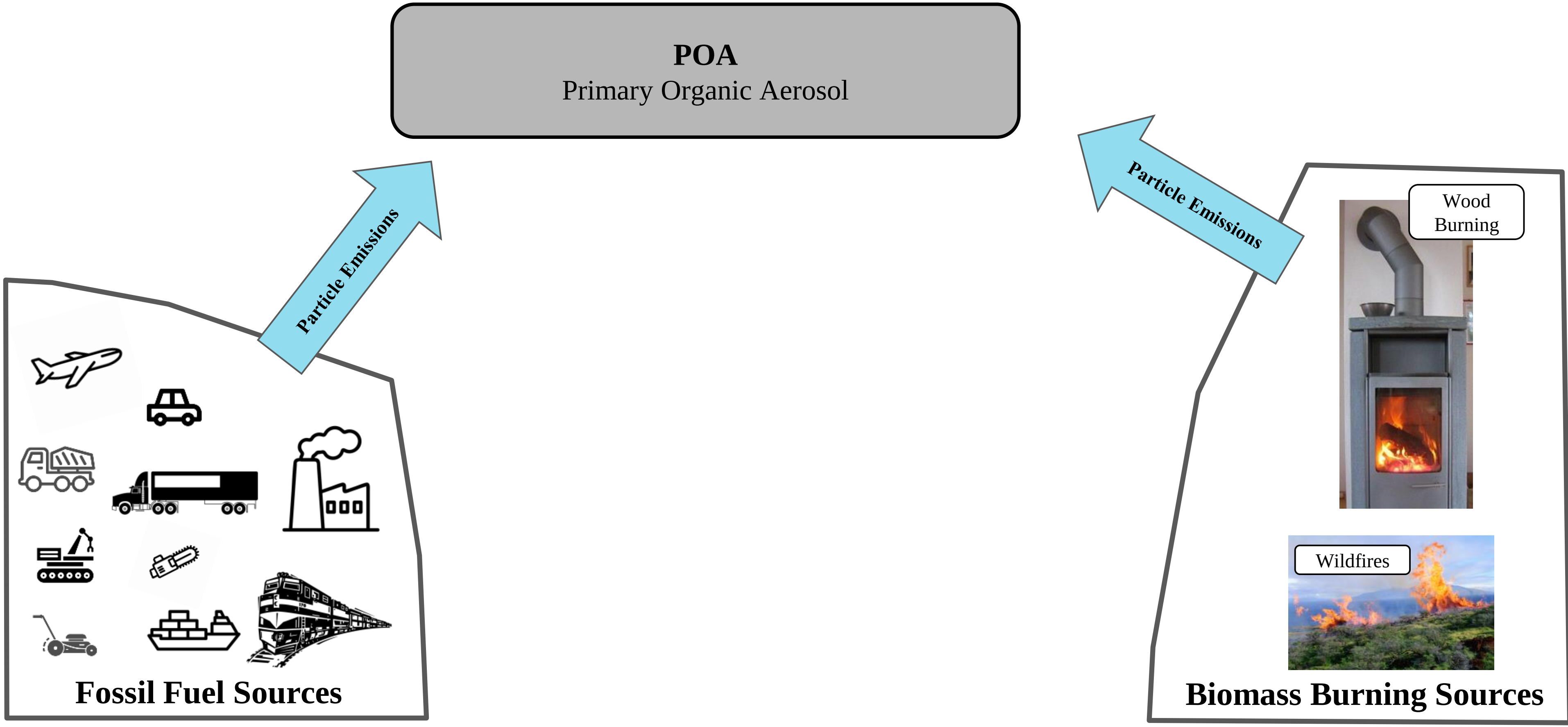


Source	Comp
Wildfire	4.4%
Wood Stove	52.2%
Other Fossil	14.3%
Traffic	1.6%
BSOA	16.8%
Background	11.4%

Aerosol Chemical Speciation Monitor (ACSM) data collection and analysis provided by the:

- Chemical On-Line cOmpoSition and Source Apportionment of fine aerosol (COLOSSAL) project (Chen et al., 2022)
- Chemical Characterization of Particles (CARA) program (Chebaicheb et al., 2024)

Organic Aerosol Formation



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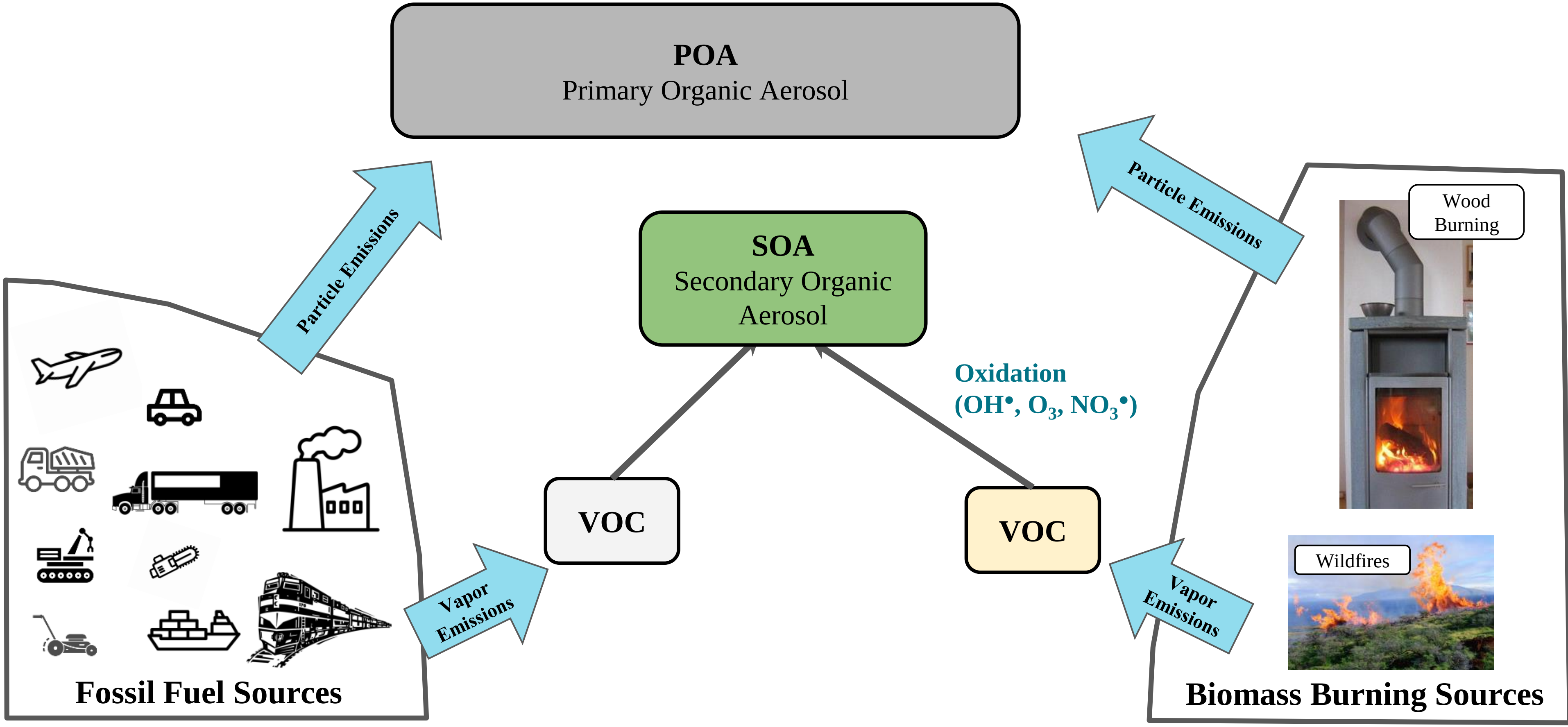

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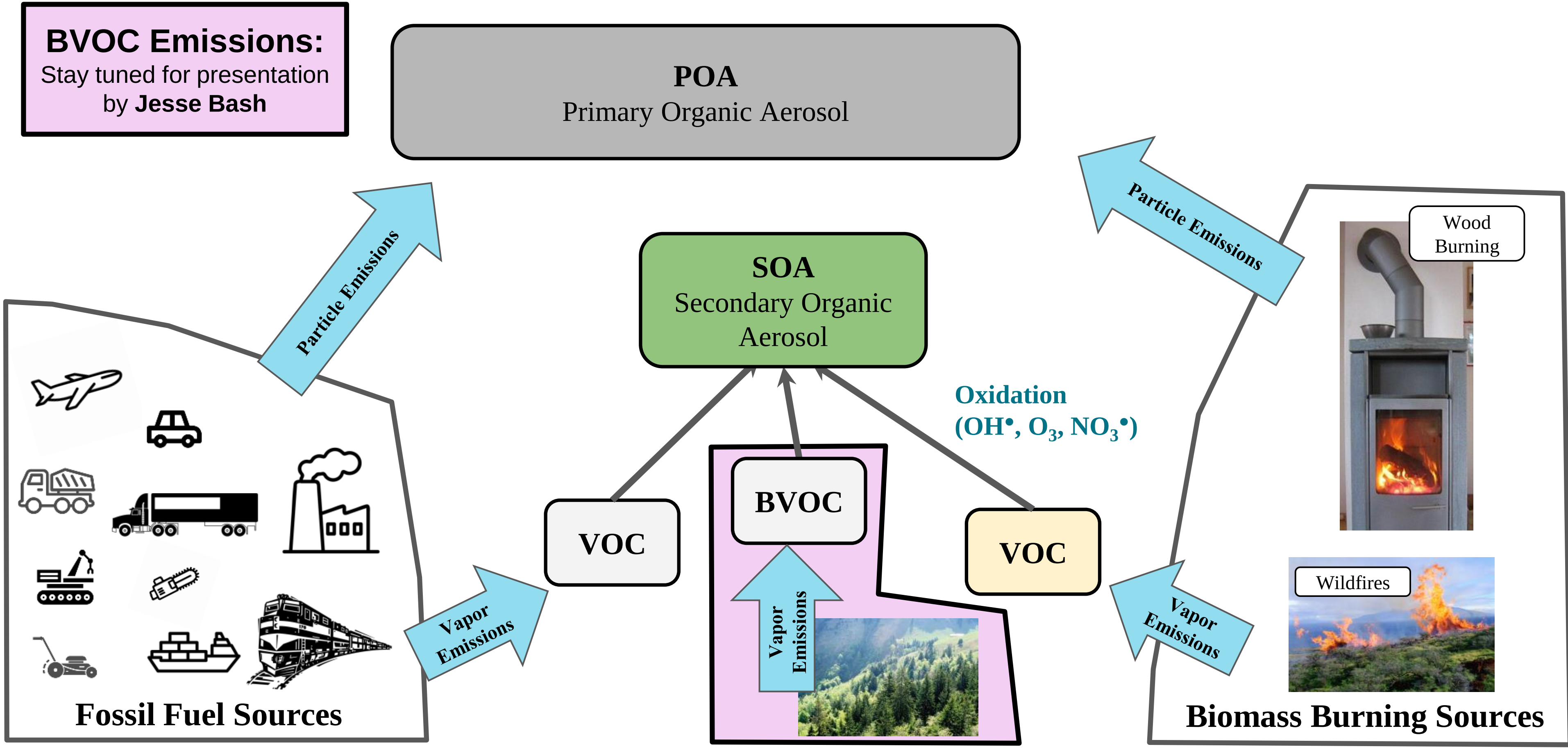

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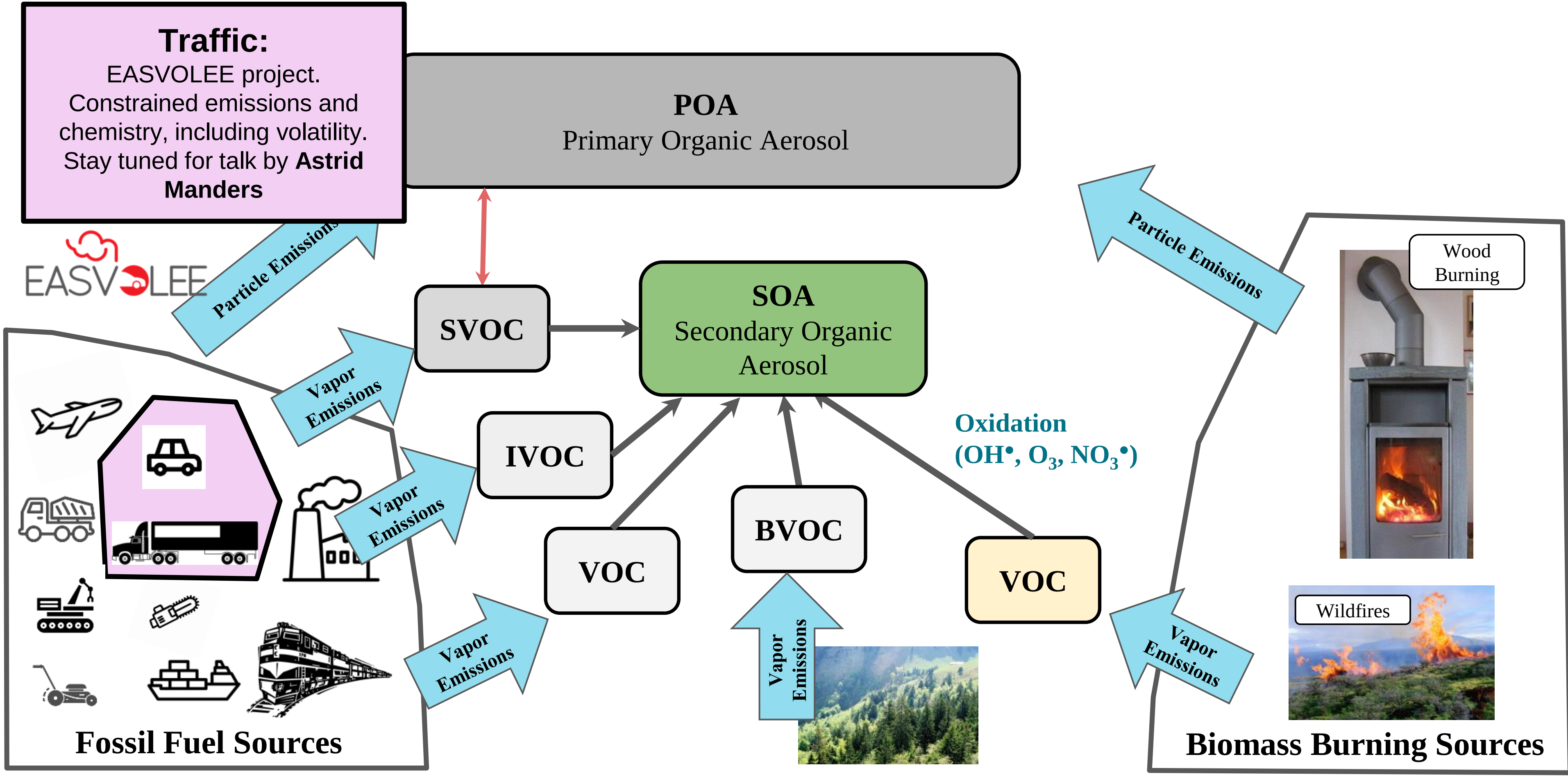

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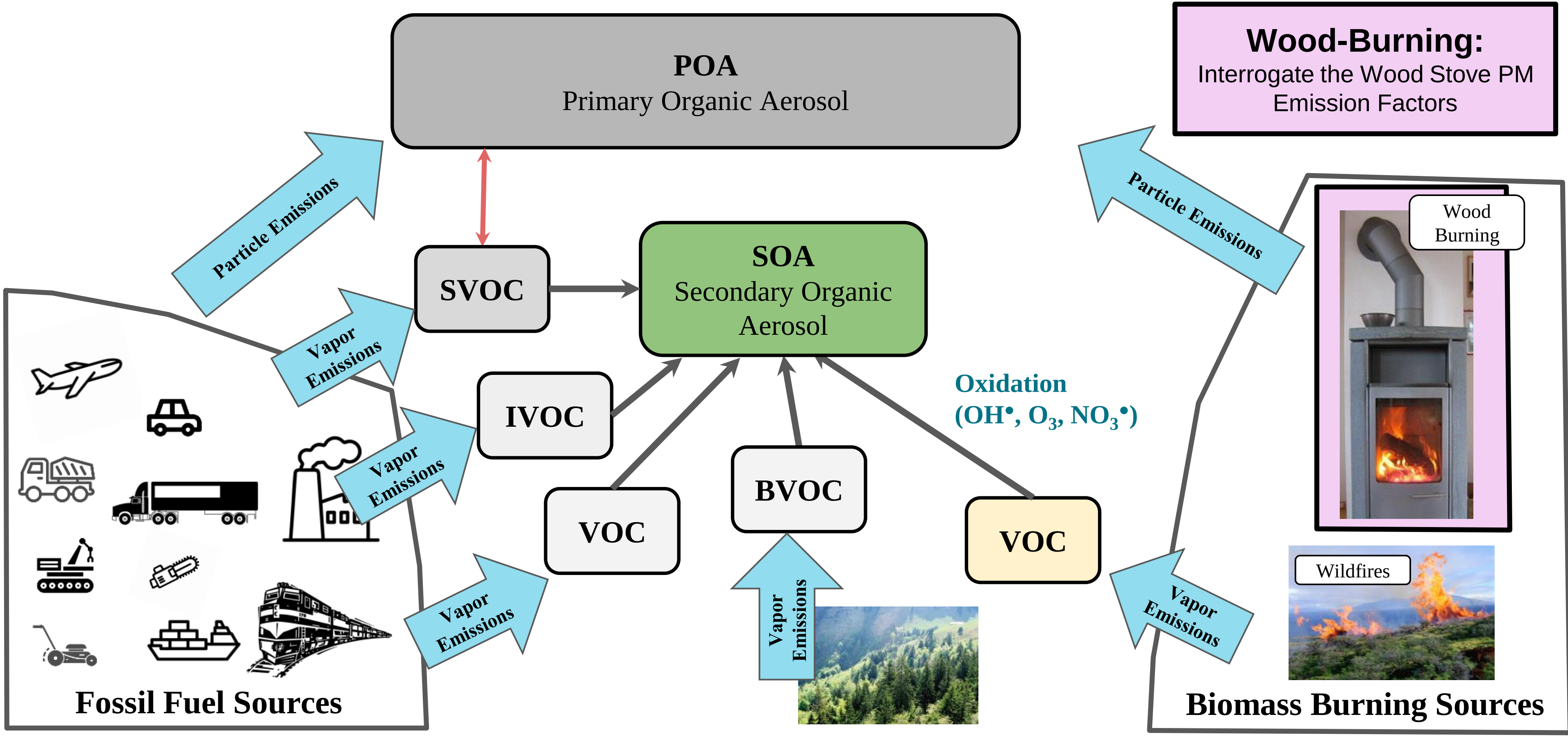
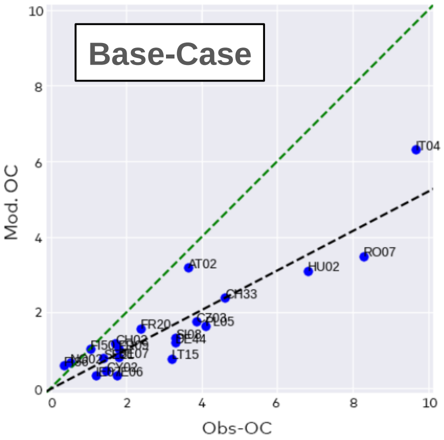
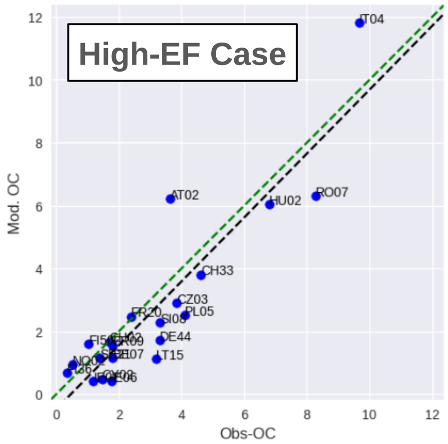


Impact of higher Wood-Burning Emission Factors

- Base-Case: central estimate for wood stove PM emission factors.
- High-EF Case: bottom-up inventory with high emission factor assumptions
- Model OC agreement dramatically improves.





From Simpson, Kuenen, et al., NMR report 2022.

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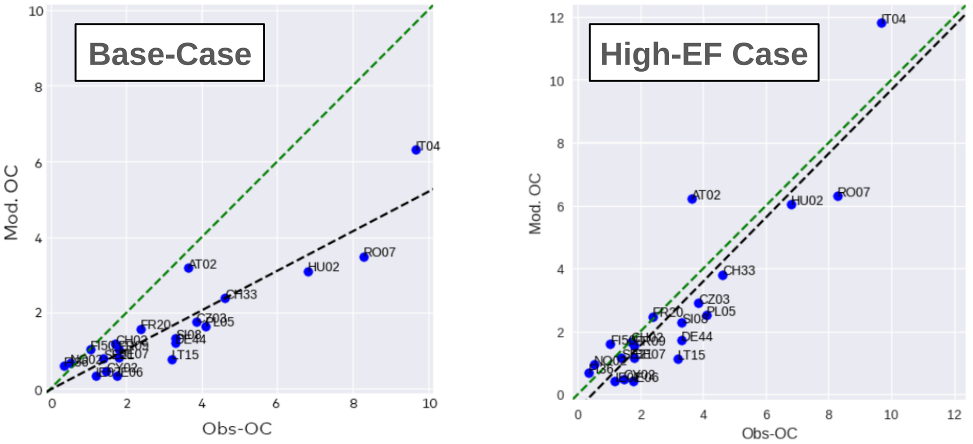

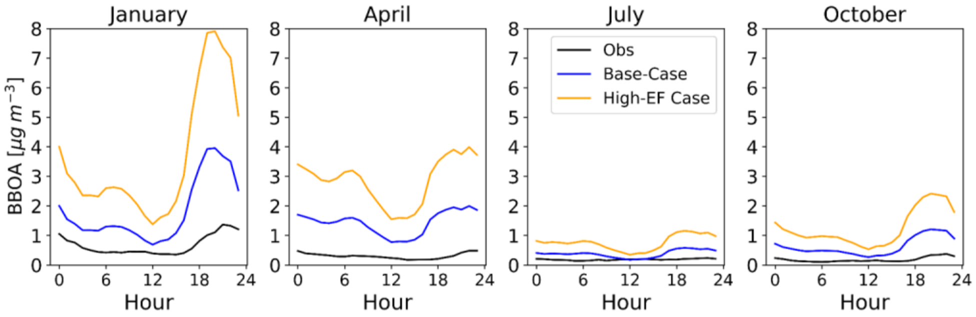

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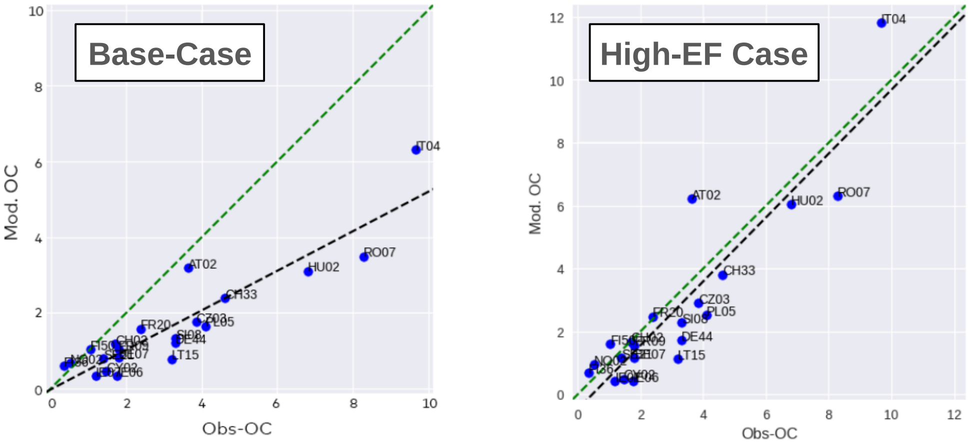
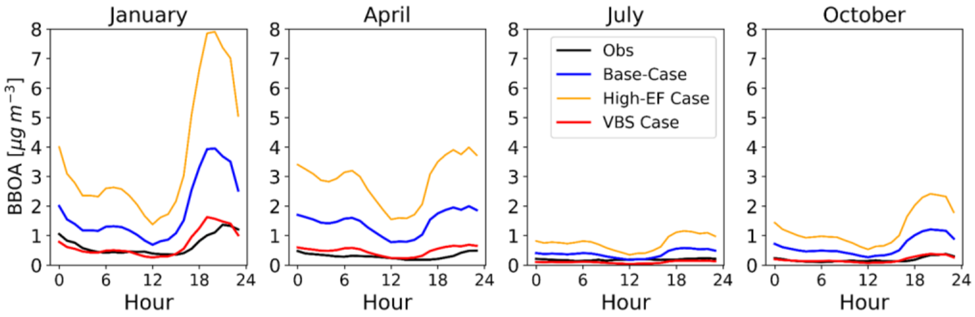
- Including evaporation of Base-Case EFs agrees with PMF





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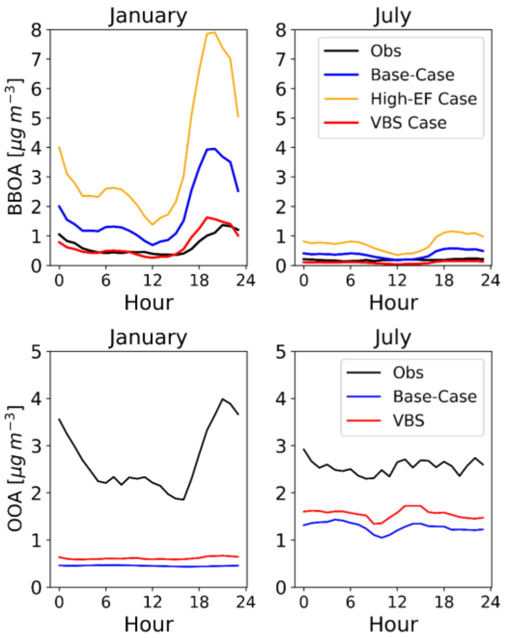
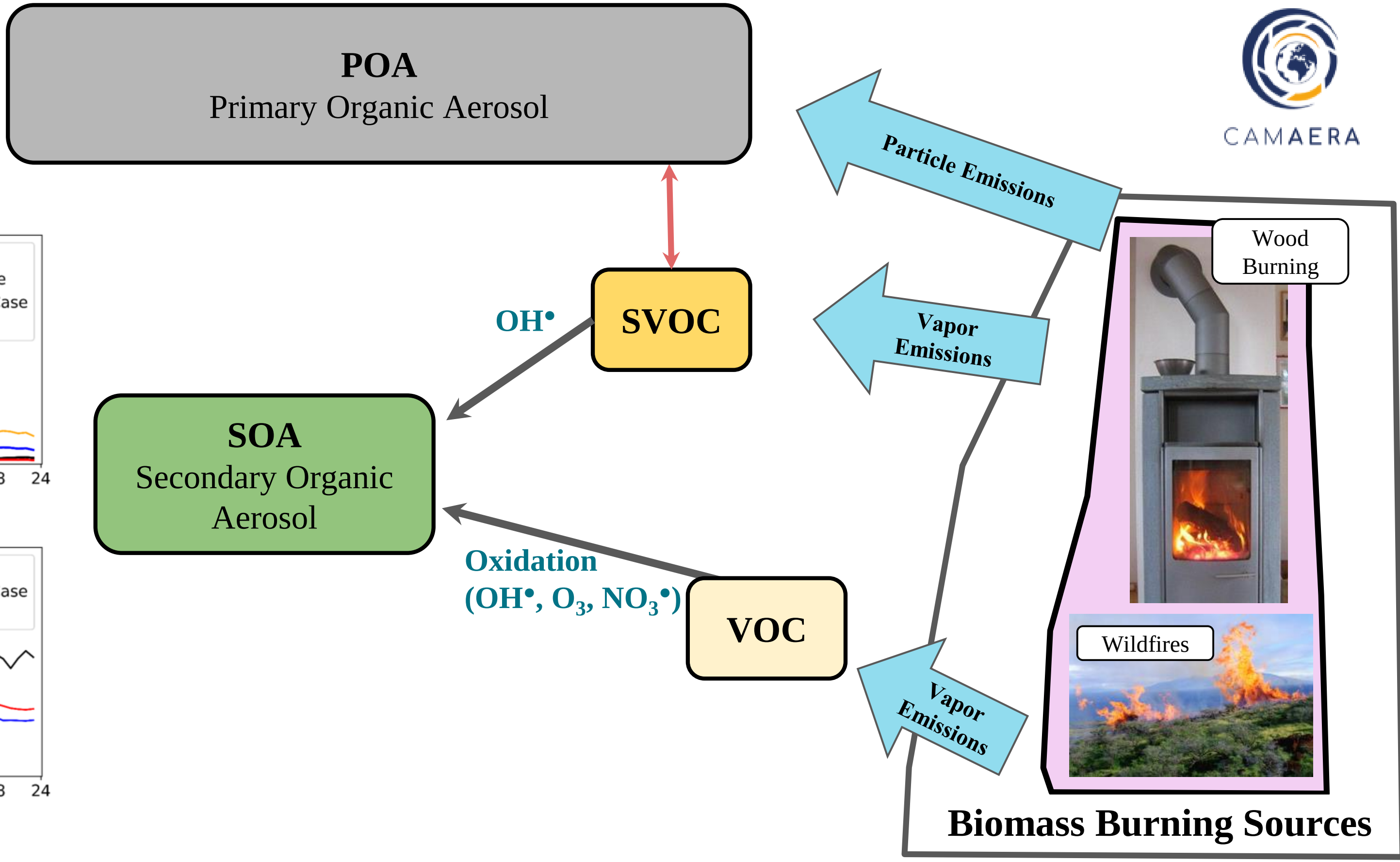
Organic Aerosol Formation

Biomass-Burning:
CAMAERA Project.
Investigate the vapor emissions and chemistry of biomass burning pollutants.





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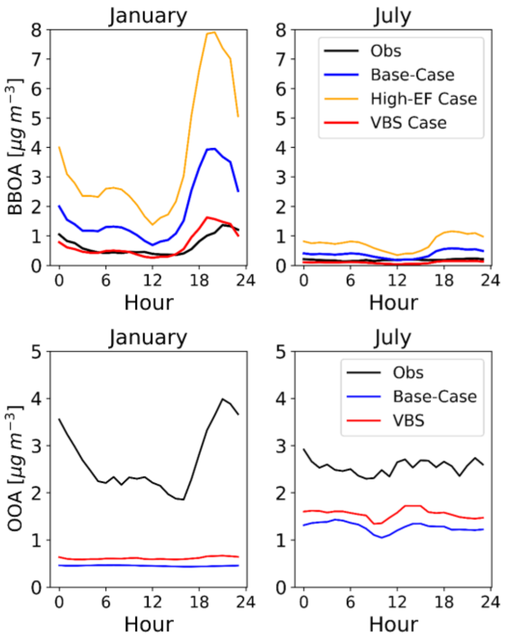
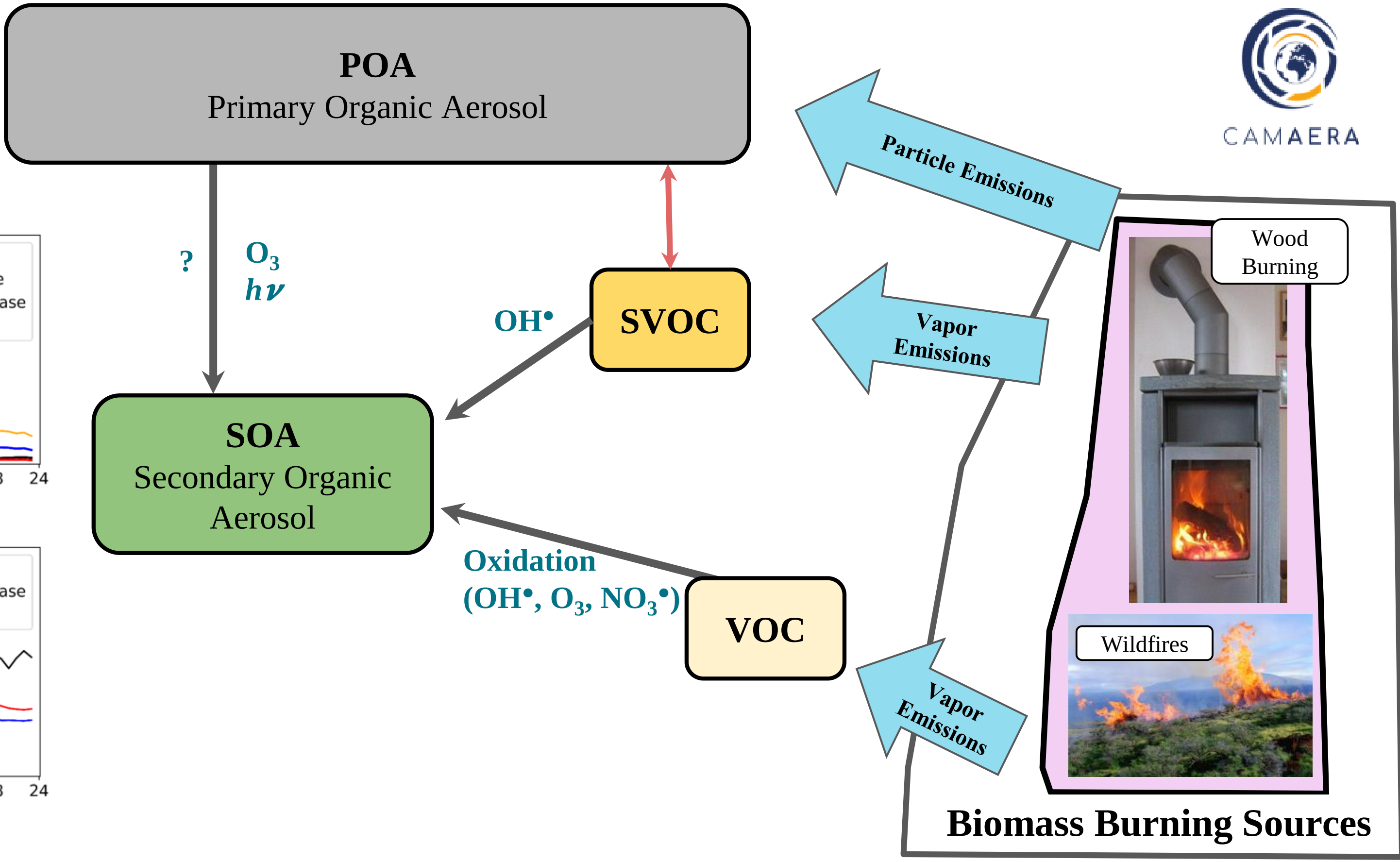


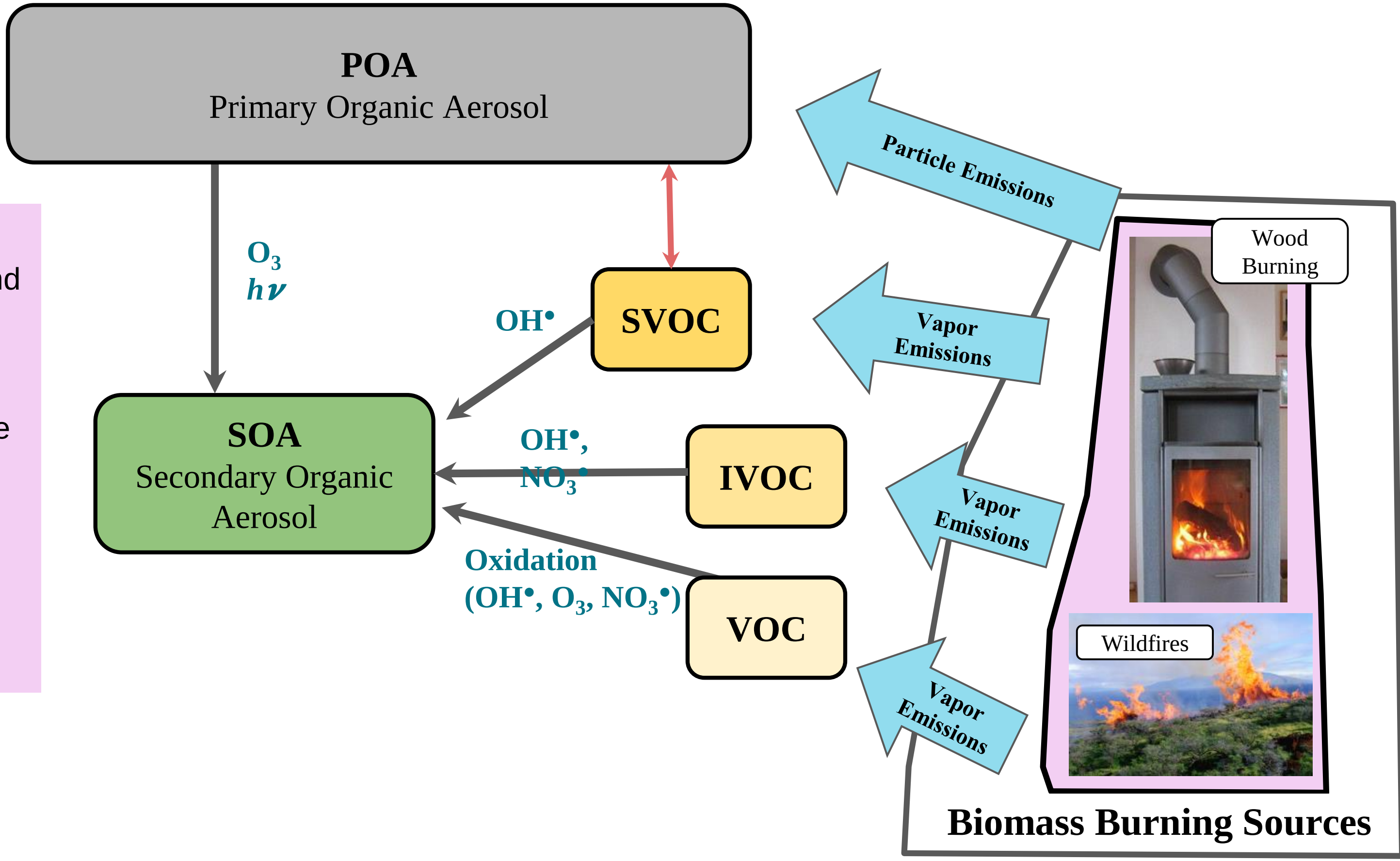


Organic Aerosol Formation

Biomass-Burning:
CAMAERA Project.
Investigate the vapor emissions and chemistry of biomass burning pollutants.

1. Primary emission volatility – relative magnitude of POA and SVOC.
2. Emission of IVOCs and OH reaction to form SOA
3. Novel reactions with nighttime oxidant, $NO_3^\bullet$
4. Heterogenous oxidation or photolysis of PM

Stay tuned for talk by Florian Couvidat



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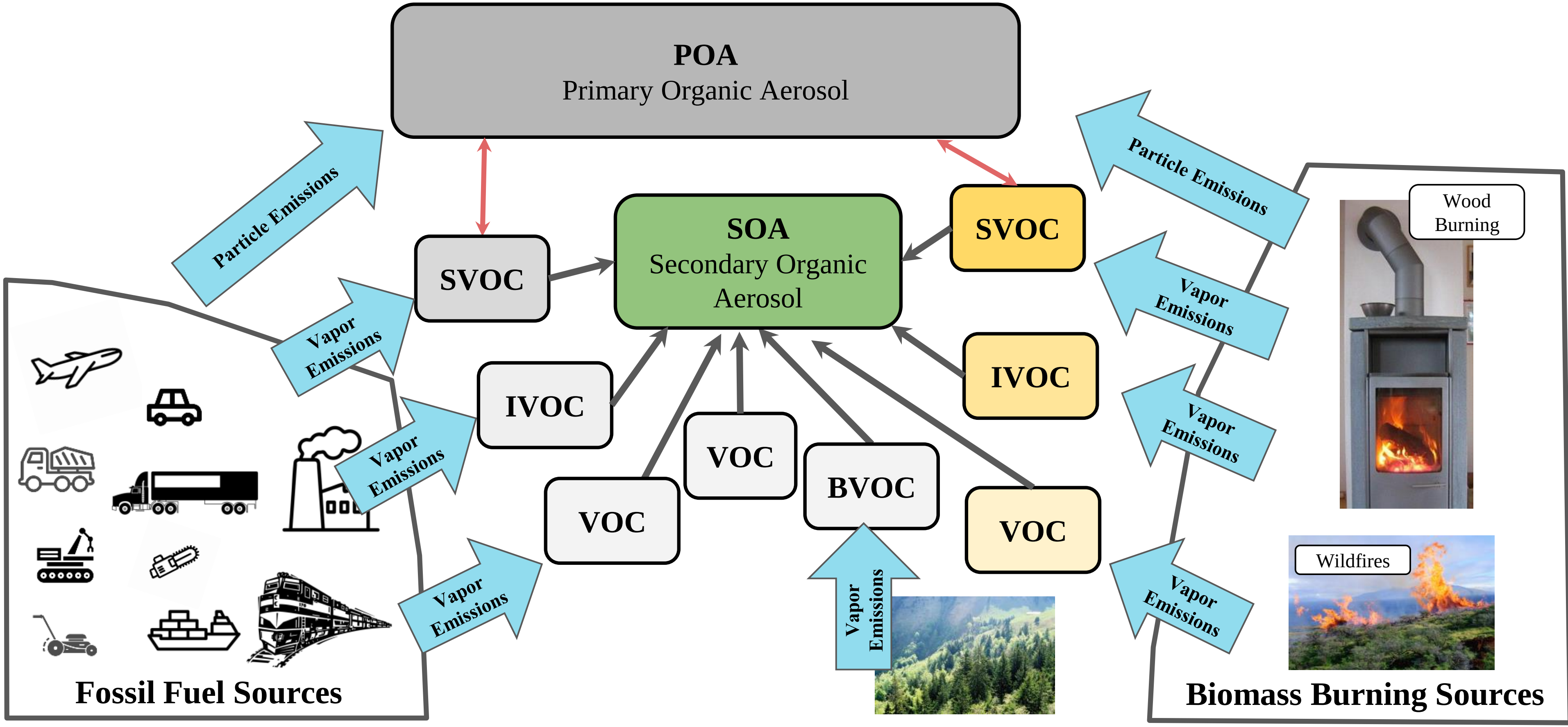

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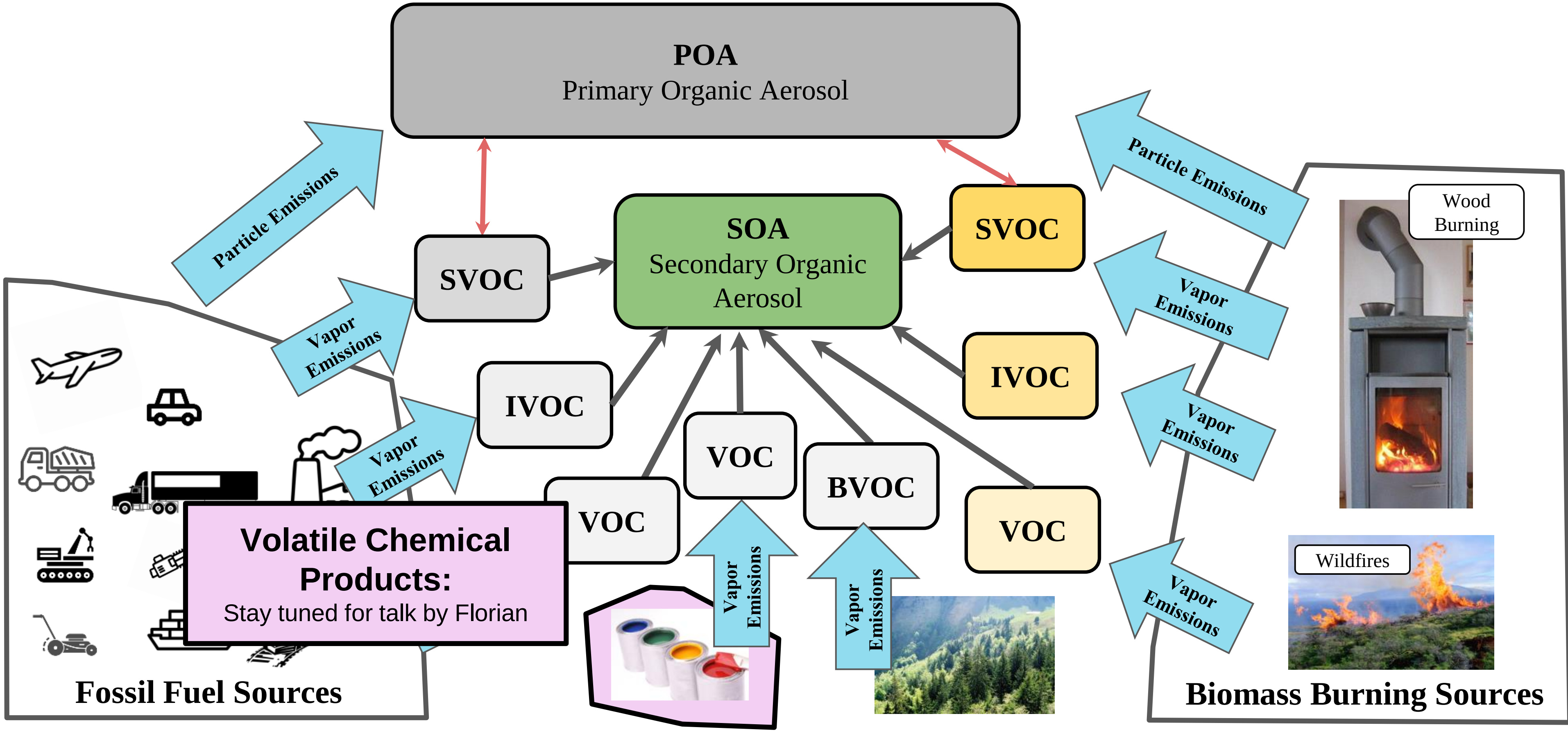

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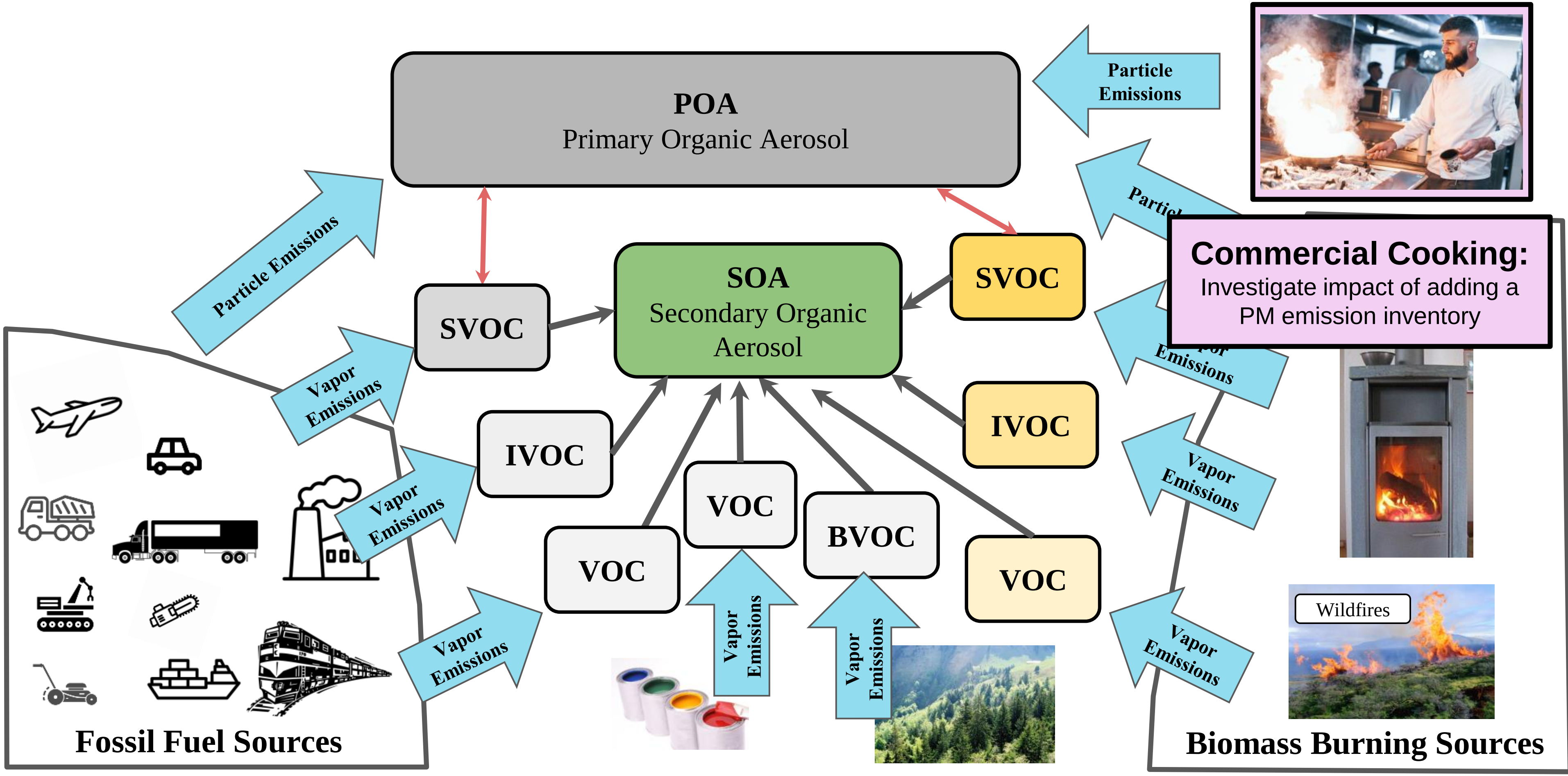
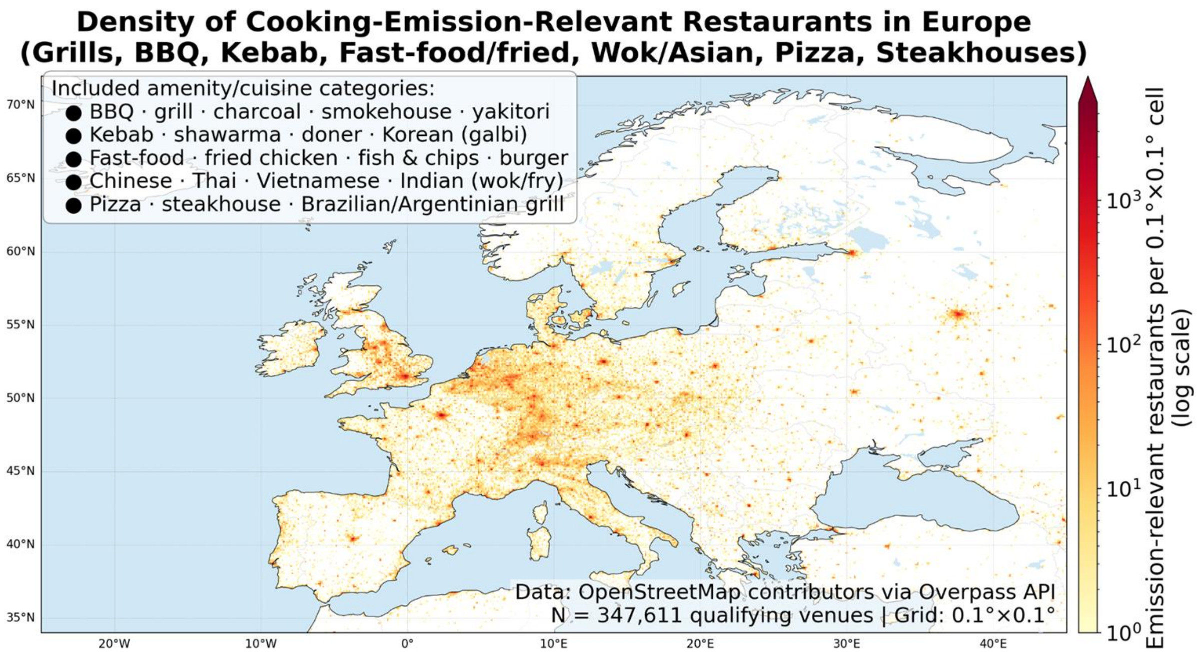


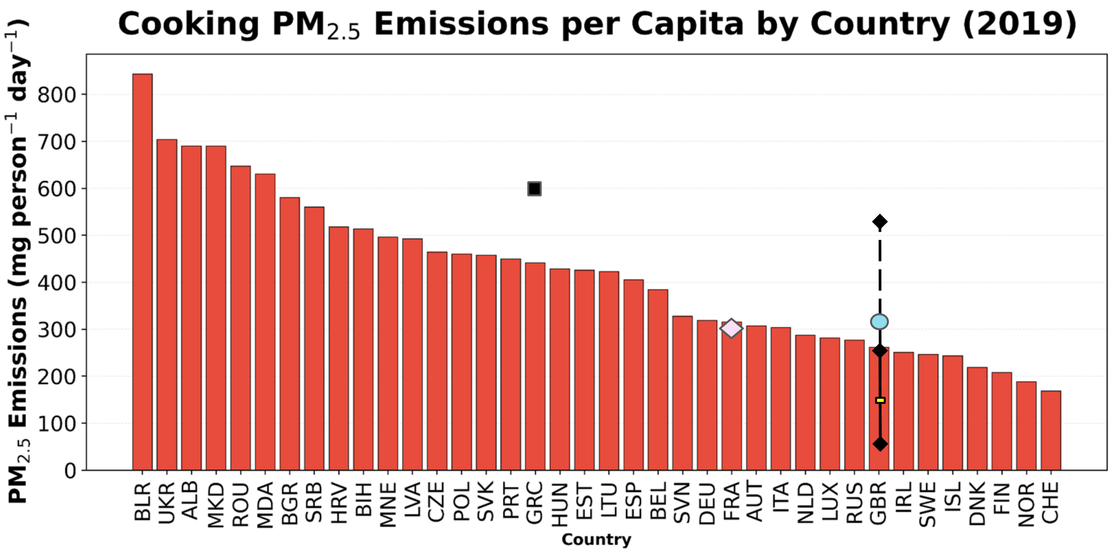
Commercial Cooking Emission Inventory

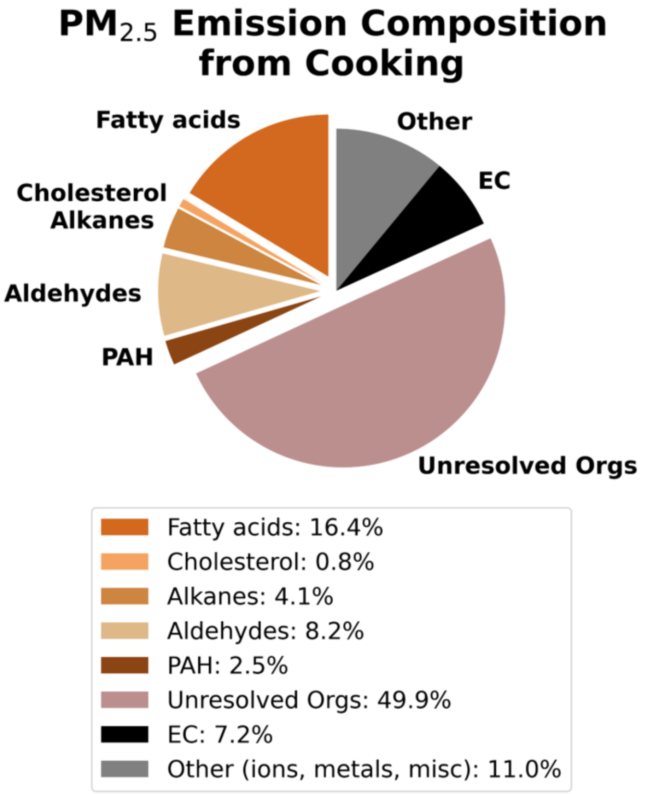
- Restaurant Locations (right)
  - OpenStreetMaps
- Gross Food Supply Data
  - Food and Agricultural Org. of the UN food balances.
- Out of Home Fraction
  - Eurostat, Waste and Resources Action Programme (UK)
- PM Emission Factors
  - Data from 14 literature sources applied at the level of food and cook type.
- Controls
  - Assume conservative controls implemented (~26% reduction).



- PM Composition
  - Data from 9 literature sources including Rogge et al. (1991) and Schauer et al. (1999, 2002).

Commercial Cooking Emission Inventory

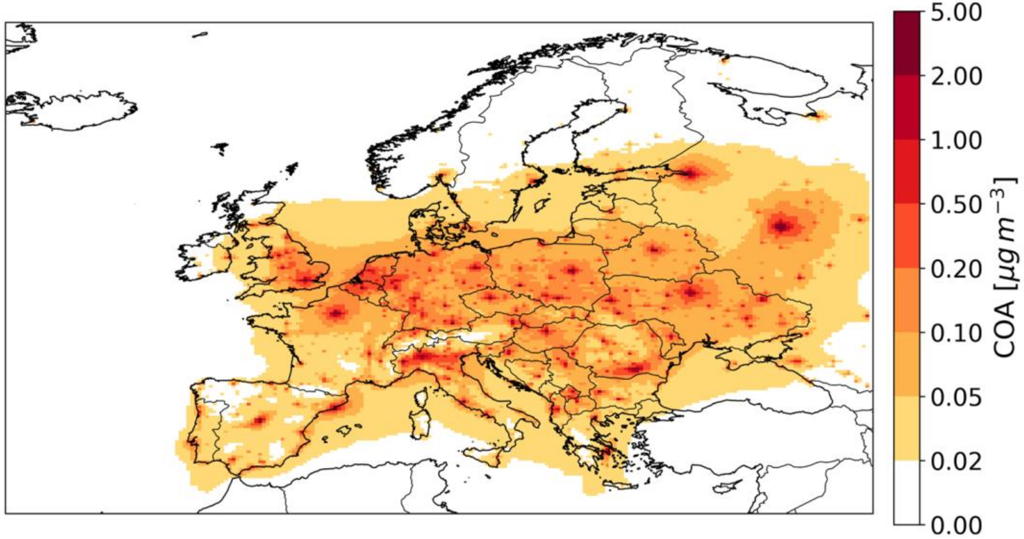


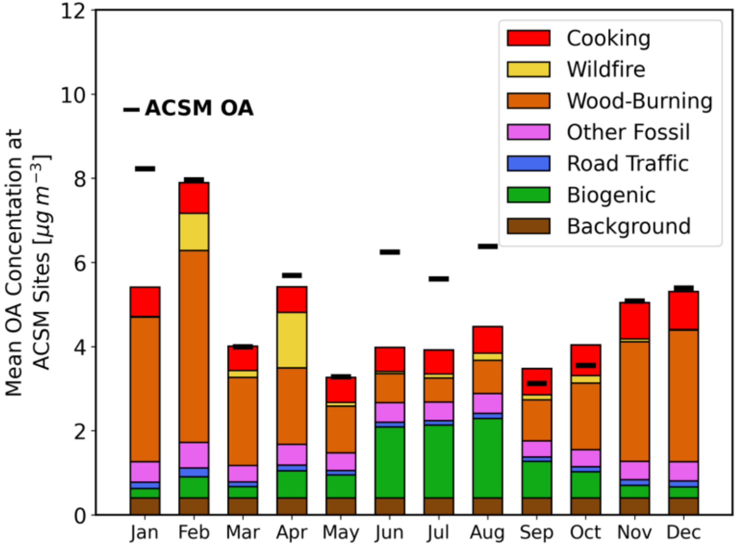


Country	Independent Reference	Estimate (mg/person/day)	Model
Greece (GRC)	Siouti et al., *Atmos. Environ.* 2021.	600	440
UK (GBR)	Murrells et al., TFEIP 2024 Meeting, Dessau 16th May 2024.	168 (76-254) Max: 528	260
UK (GBR)	Ots et al., *Atmos. Chem. Phys.* 2016.	320	260
France (FRA)	Florian Couvidat (CAMAERA)	300	320

Currently assume all OA (~81%) is generic

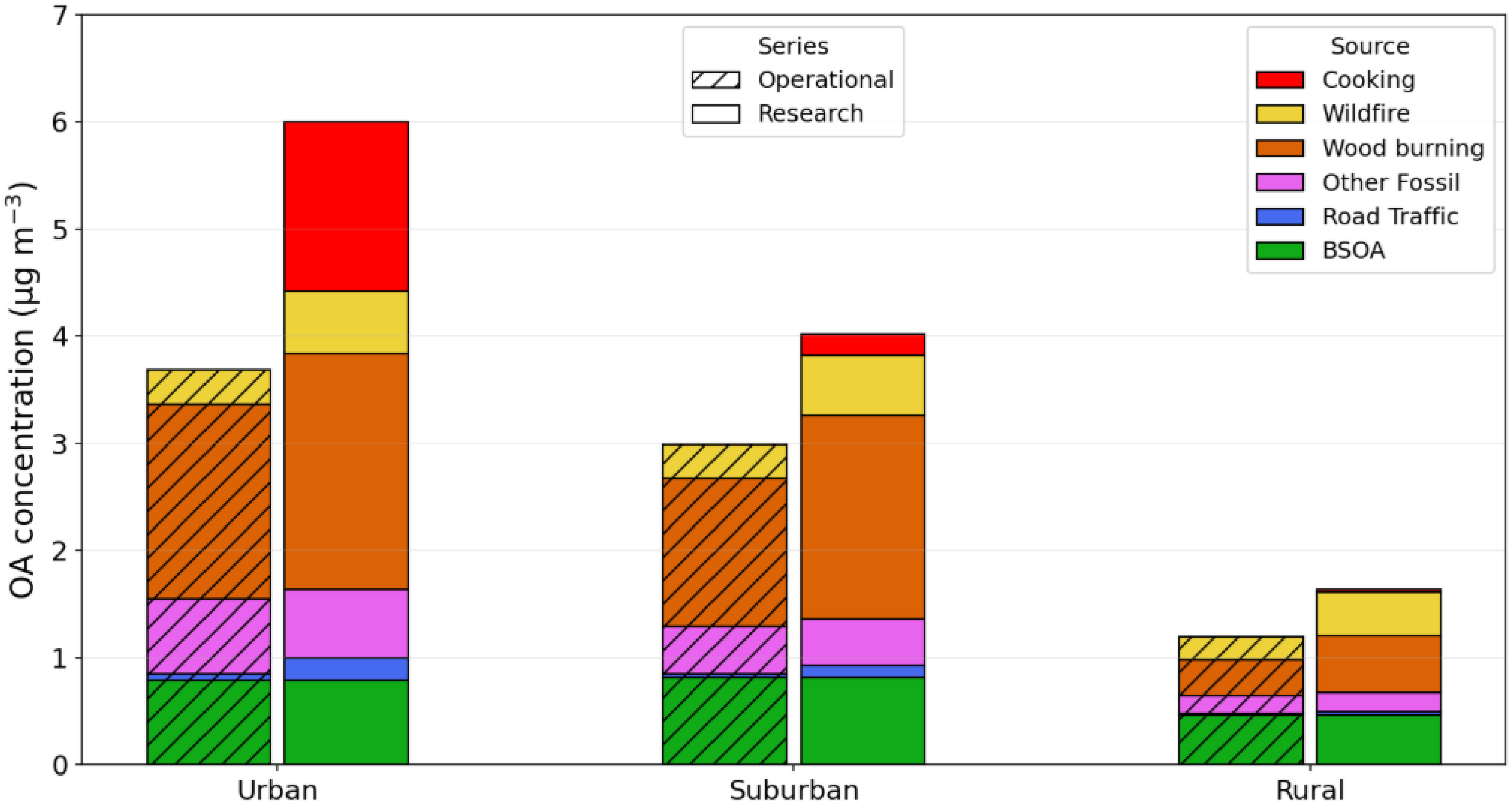
Commercial Cooking Organic Aerosol





- EMEP-predicted annual average 2019 Cooking OA concentrations are highly localized in cities.
- Simulation run at 0.3 x 0.2 deg. Higher resolution model could change substantially.
- ACSM sites dominated by Wood-Burning, BSOA, and Cooking.
- Wood-Burning OA (orange) includes IVOC emissions and uncertain secondary chemistry.

Source Attribution vs. Population



> 1500 people km^{-2} | [1500, 300] people km^{-2} | < 300 people km^{-2}

Conclusions

- Models still underpredict OA, but they are making progress.
    - Understanding of traffic emissions is increasing substantially.
    - Many are taking primary organic aerosol volatility into account.
    - Seasonal and diurnal profiles are better captured.

- Commercial Cooking could contribute PM emissions ~100 kt yr^{-1}. Traffic Exhaust PM emissions for 2019 are ~114 kt yr^{-1}.

- Source attribution results suggest dominant role of Wood-burning and BSOA at regional scales.

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Research Needs

We are still *half-blind*: need <u>major effort</u> to constrain emission inventories and observation-based analysis.

- Conversation should include experimentalists and modelers
- Critical as we move toward generating higher resolution (e.g. cityscale) model outputs.
- Wood-burning is probably the biggest anthropogenic uncertainty remaining for current OM biases. Sensitive to:
    - emission factors
    - speciation (e.g. IVOCs, high-SOA forming structures)
    - activity data
    - fuel condition (wet/dry)
    - spatial distribution
- Other emission sources are both poorly constrained and poorly understood including Volatile Chemical Products (i.e. solvents), Cooking, and Offroad engines
- We need more ACSM measurements and Mass Spectral data in suburban, rural and background sites at low elevation.
- Need to consider more sophisticated chemistry and anthropogenic-biogenic interactions in research-grade models (e.g. organic nitrates, organo sulfates, etc.)

Acknowledgements

- Organic aerosol model developments and evaluation also supported by EU EASVOLEE and CAMAERA projects.
- Comparisons make use of new emissions of SVOC and IVOC from road traffic, compiled by TNO as part of EASVOLEE.
- ACSM data from Chemical On-Line cOmpoSition and Source Apportionment of fine aerosol (COLOSSAL), RI-URBANS, and ACTRIS projects (Chen et al., 2022) as well as French CARA (Chemical Characterization of Particles) program (Chebaicheb et al., 2024).

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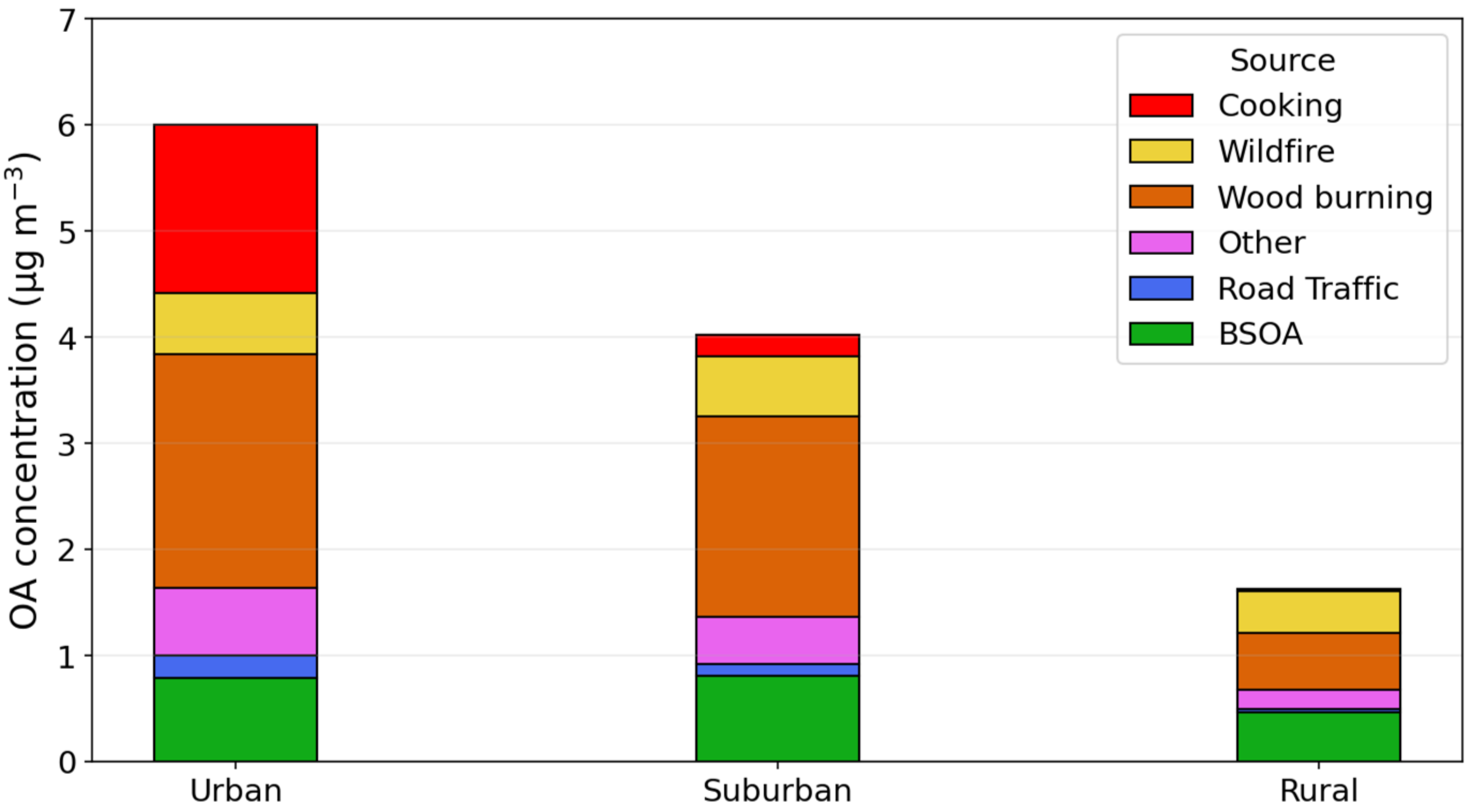
Back-Up Slides

Source Attribution vs. Population



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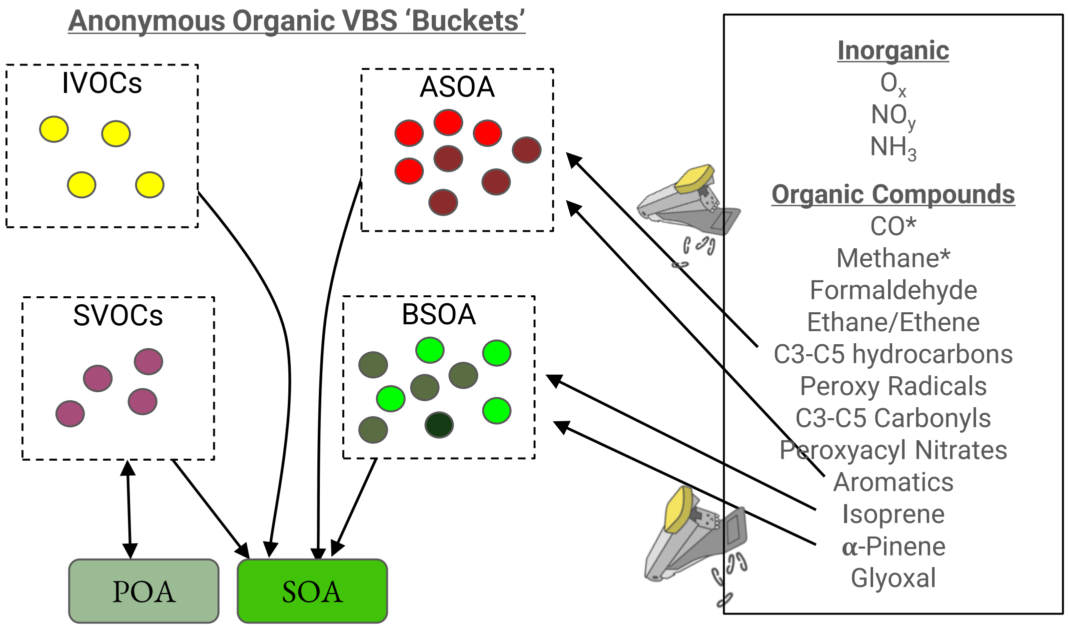
Chemical Mechanisms in Models

There is value in moving from anonymous VBS approaches to mechanisms that:

- include more individual species end-points (direct connections to toxicity)
- better interconnect chemistry among systems
- better represent carbon chain-length and structure (i.e. SOA potential)
- include more reaction intermediates



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Inorganic
O_x
NO_y
NH_3

Organic Compounds
CO*
Methane*
Formaldehyde
Ethane/Ethene
C3-C5 hydrocarbons
Peroxy Radicals
C3-C5 Carbonyls
Peroxyacyl Nitrates
Aromatics
Isoprene
α-Pinene
Glyoxal

Explicitly Integrated Organic Compounds and Families
Limonene
IEPOX
Sesquiterpenes
Polycyclic Aromatics
Furans
Organic Nitrates
Organic Sulfates
Ethylene Glycol
C6-C30 Hydrocarbons
C6-C27 Alkylperoxy and Peroxy Radicals
C9-C20 Carbonyls

Species Partition based on their volatility and solubility

Organic Aerosol

Modeled Total OA: SVOC, IVOC, and VOC Contributions

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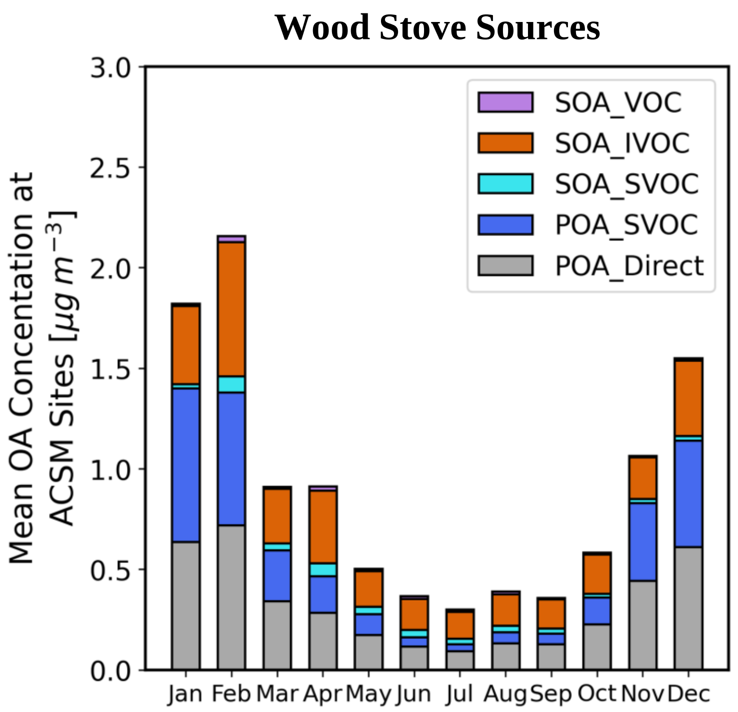
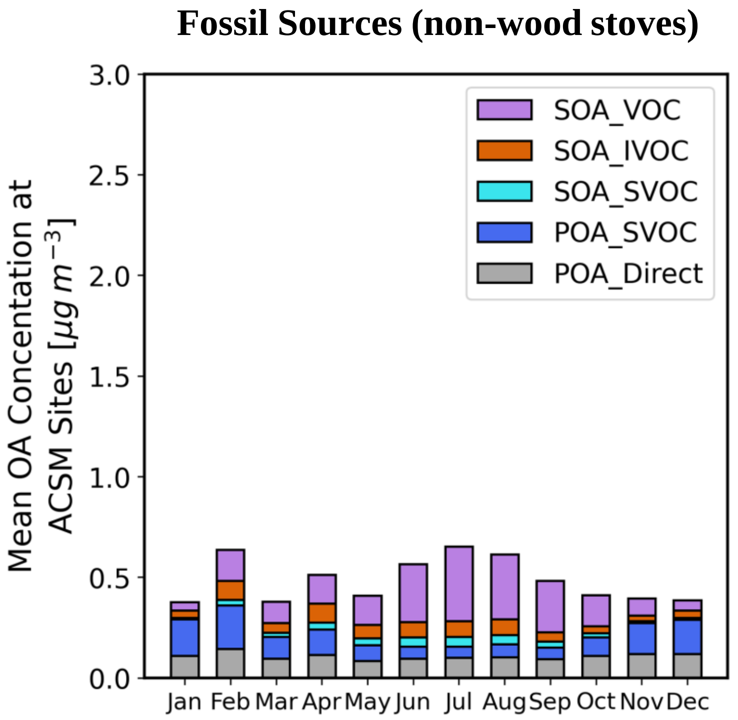
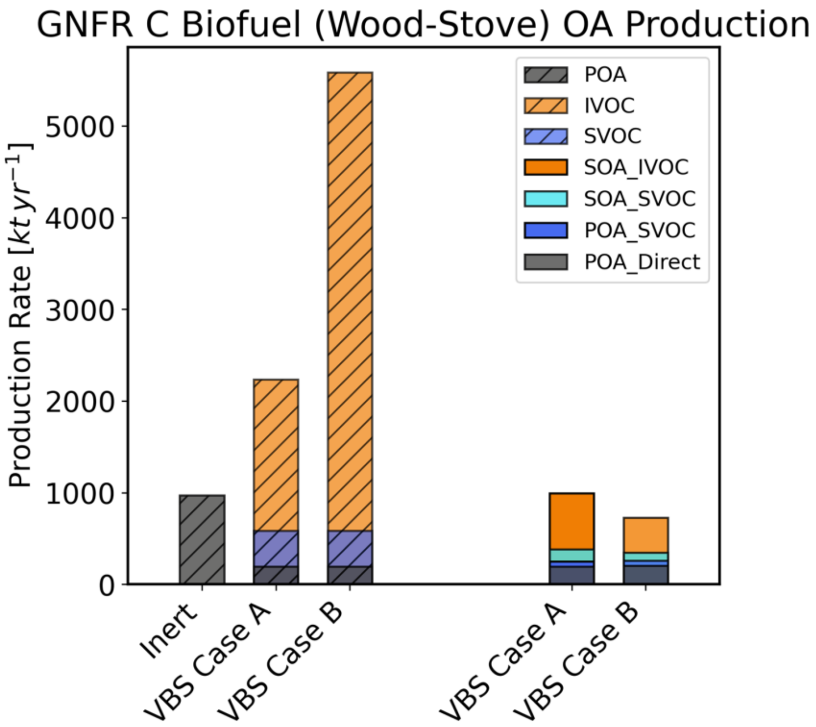




SVOCs, IVOCs, and VOCs are important contributors to OA throughout the year.

Is SOA from wood-burning VOCs underrepresented in the model (speciation, magnitude)?

Wood-Burning IVOC is Uncertain



- Production Rate predicted by EMEP for 2019 Annual Simulation
- LVOC - low volatility organic carbon (aka. primary particle emissions)
- VBS Case A -
  - Koo et al. (2014)
  - IVOC Emission = 1.5 * [Inert POA]
  - IVOC SOA Yield ~ 0.15
- VBS Case B -
  - Ciarelli et al. (2017)
  - IVOC Emission = 4.75 * [Inert POA]
  - IVOC SOA Yield ~ 0.08

How will we further improve OC performance?

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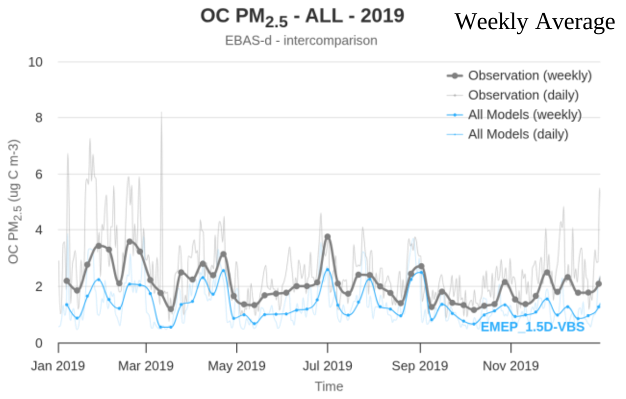
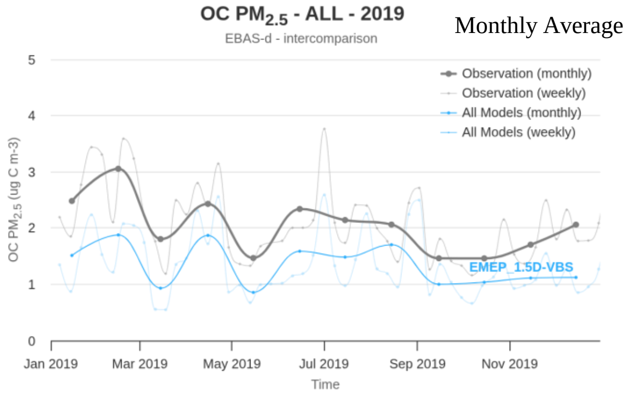
- Underrepresented Emissions
  - IVOCs from wood stoves are uncertain. To what extent are they included in inventories already?
  - Is the speciation of wood burning and fossil VOC correct?
  - Commercial cooking PM and VOC emissions?
- New Chemical Pathways
  - Wood burning VOCs and IVOCs may react with NO_3 radicals at night
  - Heterogeneous processing of BBOA may drive more SVOC into the particle phase.
  - Isoprene oxidation produces epoxides that may form SOA through complex aerosol processes.
  - Monoterpenes form extremely low volatility organic compounds, depending on the NO_X regime.





What is the Role of Cooking OA (COA) Emissions?

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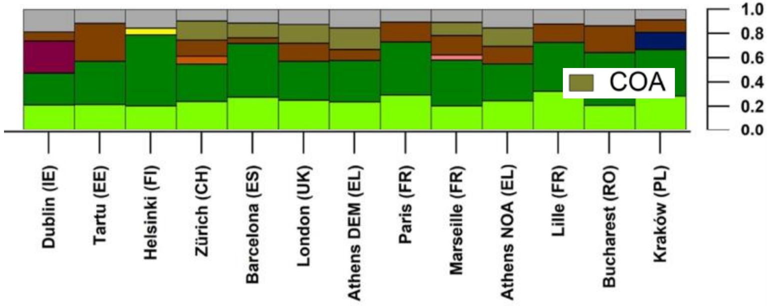
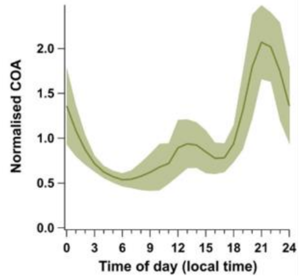
- From PMF Measurements (Chen et al., 2022)
  - Fresh COA relative contribution ~15% of total OA with little seasonal dependence.
  - Fresh COA is mainly detected in cities. The fingerprint is less detectable at rural sites.
  - Shows distinctive noon and evening peaks.
- Reactivity
  - High composition of fatty acids
  - May react quickly with ozone on the order of minutes to hours
  - Could be detected as OOA at regional scales?

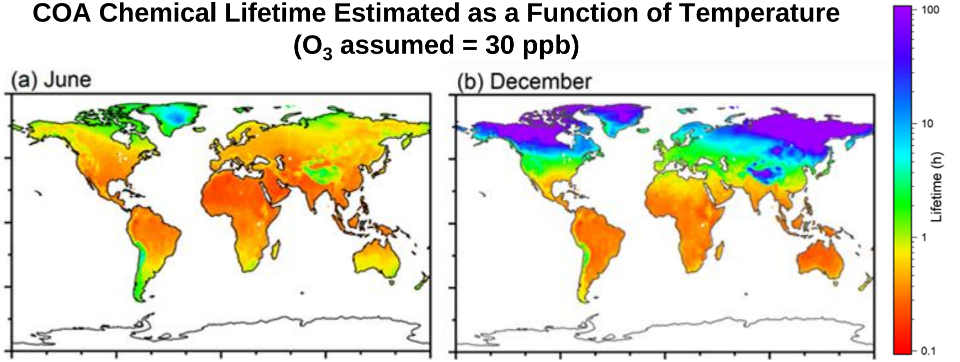
RI-URBANS observations from 2016-2019.





Chen et al., 2022

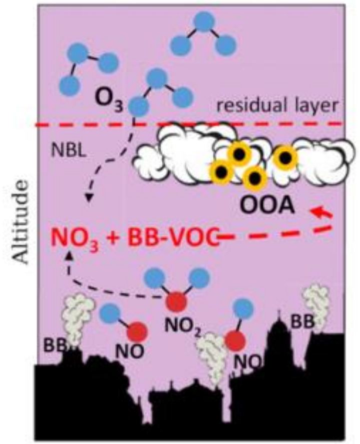
COA Chemical Lifetime Estimated as a Function of Temperature (O_3 assumed = 30 ppb)



Liu et al., 2024: Measured k_{O3} in the lab for a series of cooking oils over a range of temperatures.

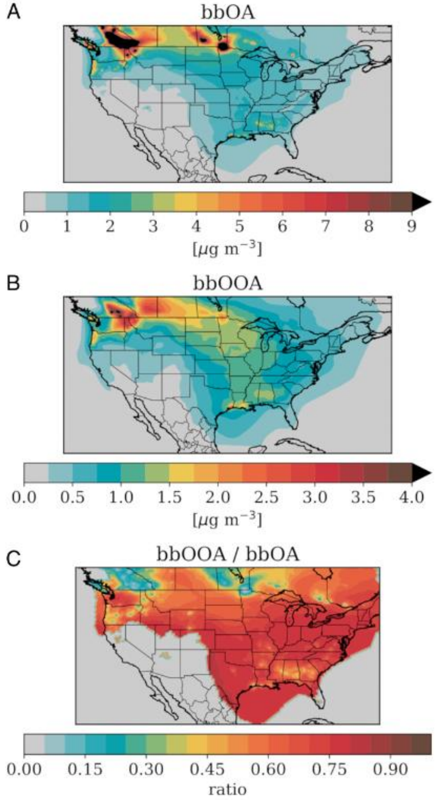
What is the Role of Nighttime Oxidation of BBOA

- Biomass Burning organic vapors may react readily at night with Nitrate (NO_3) radicals that stabilize under dark conditions
- This could enhance the signal predicted for OOA in Winter at night.
- Other Heterogenous BBOA reactions will be investigated:
  - could further enhance the OOA signal
  - but would potentially reduce the Fresh BBOA signal
  - unless more Fresh BB organic vapors partition to the aerosol after the existing BBOA reacts.



PMCAMx Model (Nov, 2011)

Kodros et al., 2020



Fine OA from natural sources (BVOC)

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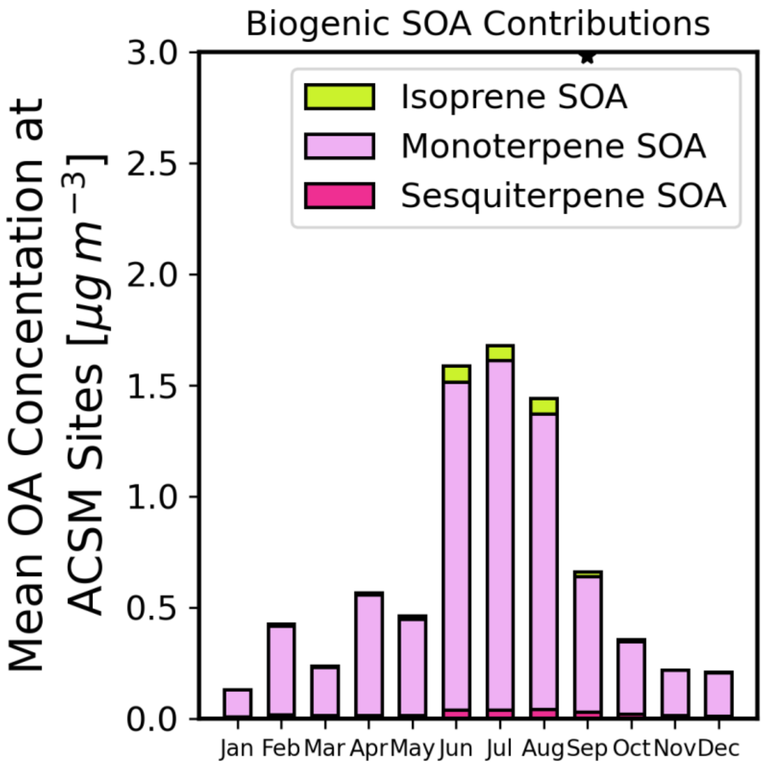
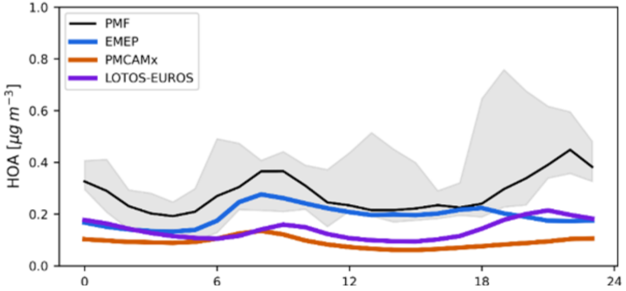
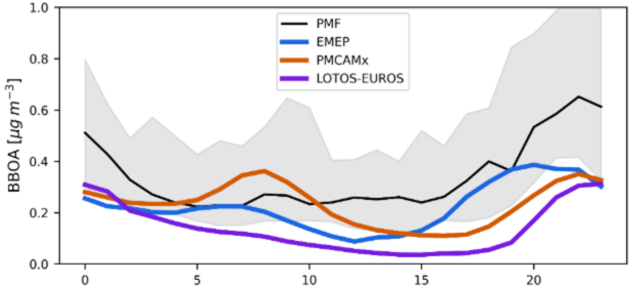


- Isoprene and Sesquiterpenes form minimal SOA at ACSM sites (Chen et al., 2022; Chebaicheb et al., 2024).
- Investigate missing SOA pathways (e.g. autooxidation, isoprene epoxide heterogeneous reaction).
- Need to apply most recently updated BVOC emissions.

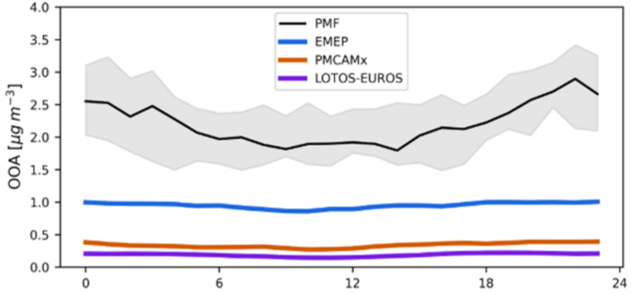
Hints that RWC emissions might be too low 2.



HOA



BBOA



OOA

- Evidence from EASVOLEE PMF comparison:
  - HOA from EMEP not too bad
  - BBOA also looks okay-ish
  - OOA underpredicted strongly.
- Seasonal/diurnal comparisons suggest that a combination of more RWC emission + conversion of BBOA to OOA might help solve this puzzle.
- Still, many other possibilities for OOA from multiple sources…..

From Ben Murphy, EASVOLEE presentation

What are IVOC, SVOC, VOC?

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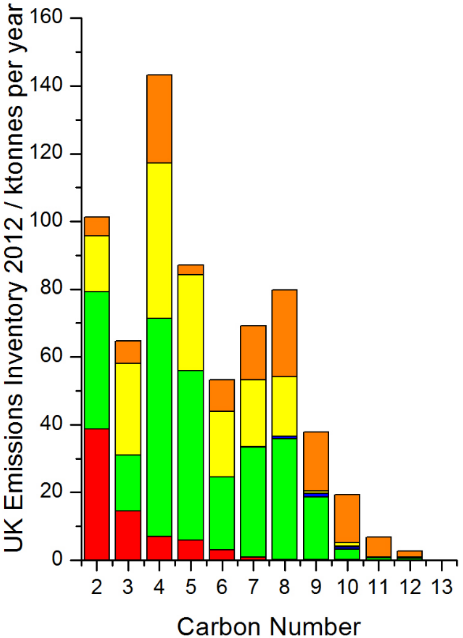
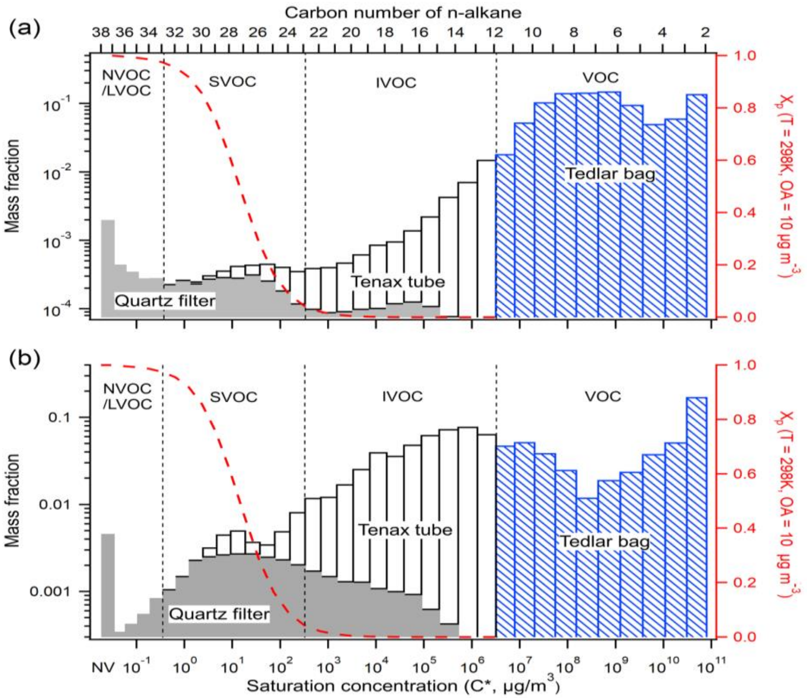
UK NMVOC emissions by C-number



(Dunmore et al, ACP, 2015)

Measured emissions from (a) gasoline, (b) diesel



(Lu et al., ACP, 2018)

Note:
UN-ECE defn VOC:

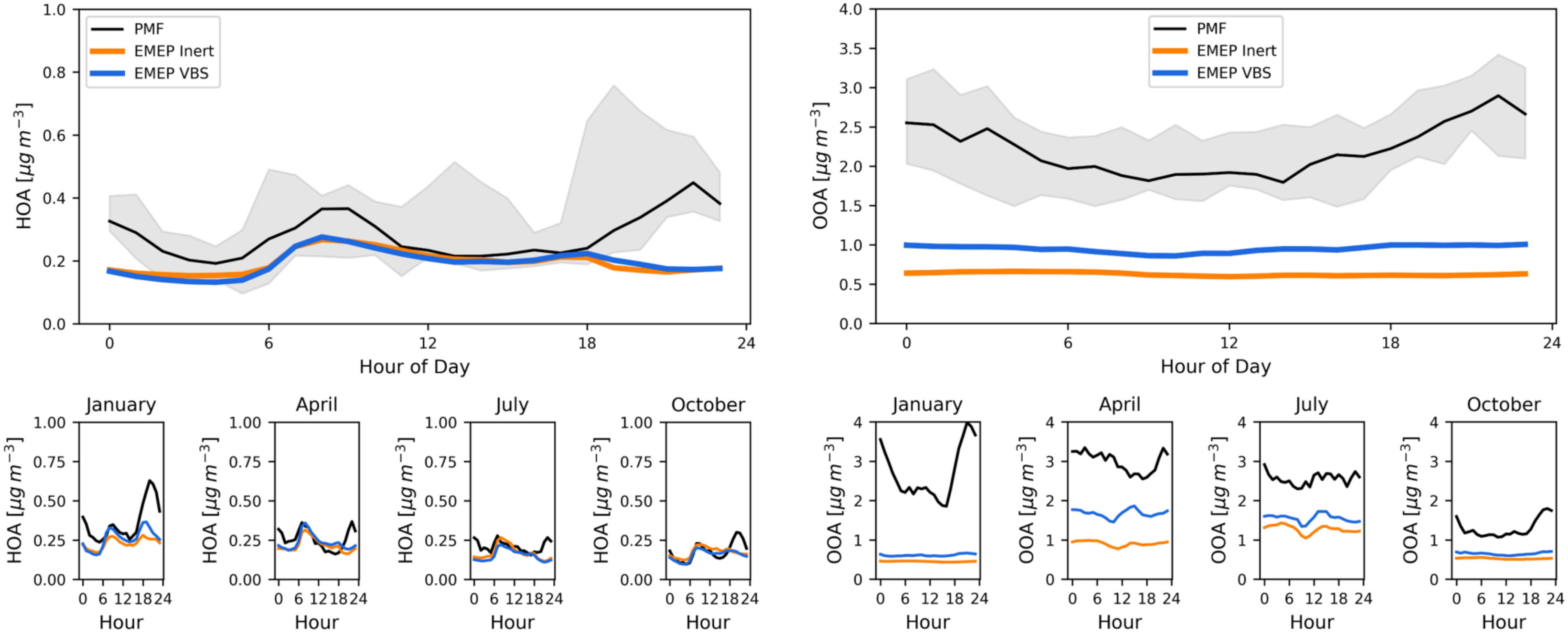
Volatile organic compound" (VOC) means any organic compound as well as the fraction of creosote, having at 293.15 K a vapour pressure of 0.01 kPa or more, or having a corresponding volatility under the particular conditions of use.

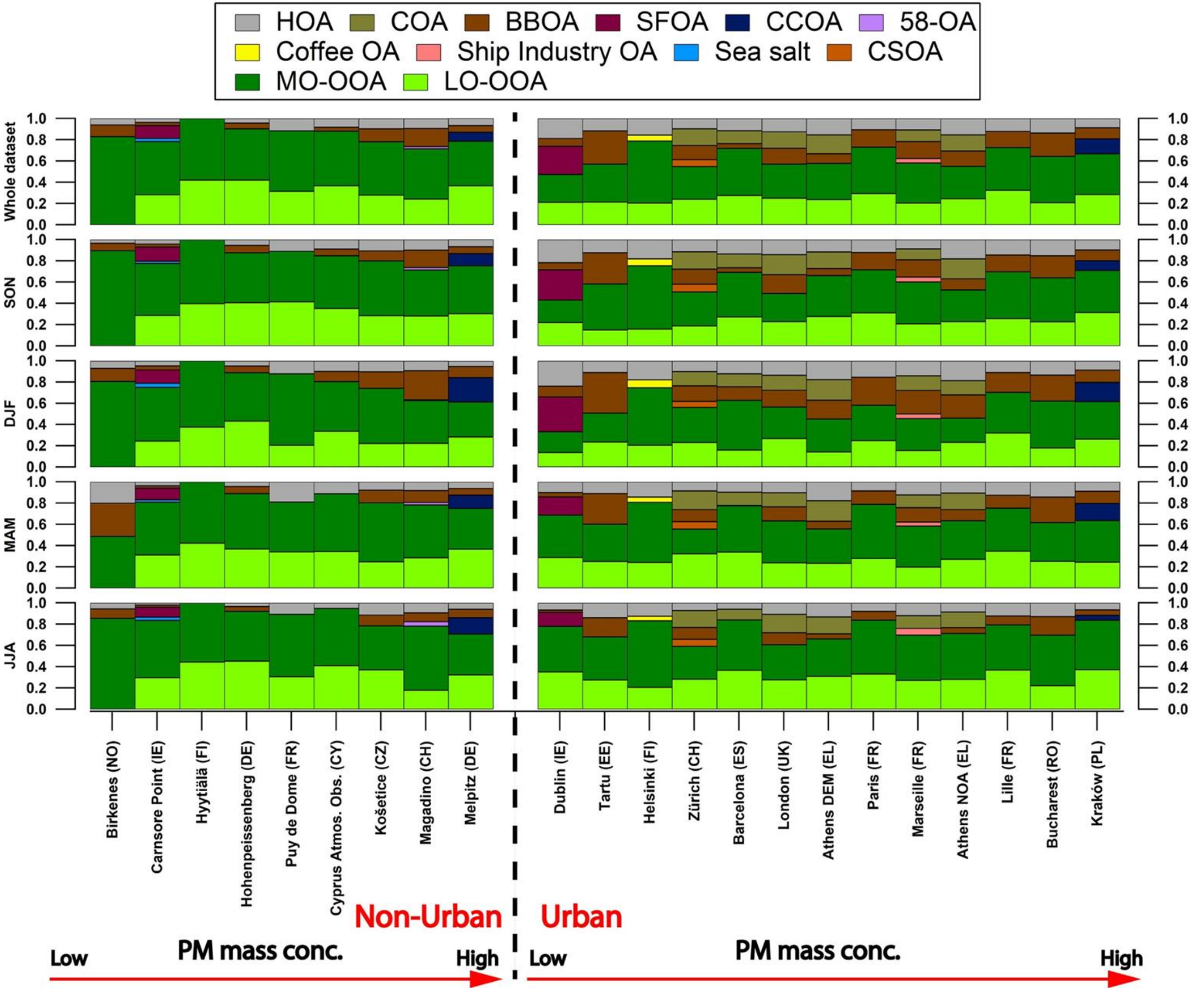
Official UK inventory stops at ca. C12. Misses a lot of SVOC, IVOC.

Road transport emissions low, but what about off-road?

Do most inventories include SVOC, IVOC, or miss them?

HOA and OOA PMF Evaluation





HOA: Hydrocarbon-like OA

COA: Cooking

BBOA: Biomass Burning

SFOA: Solid Fuel

CCOA: Coal Combustion

58-OA:

Coffee OA

Ship Industry OA

Sea Salt

CSOA: Cigarette Smoke

MO-OOA: More Oxidized Oxygenated OA

LO-OOA: Less Oxidized Oxygenated OA

Chen et al. (2022)

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[Liu et al. (2024)]

Suppressed atmospheric chemical aging of cooking organic aerosol particles in wintertime conditions

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- Injected COA from heated cooking oils into a flowtube
- Ozone was produced with a UV lamp and injected as well.
- Found significant impact of temperature on reaction rate of acids within the particles and ozone.

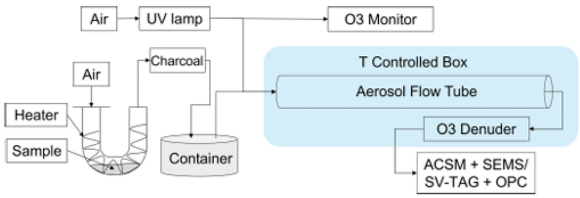


Figure 1. Schematic diagram of the experimental setup. Particles were measured using the SV-TAG and OPC for Exp. no. 8, 15 and 22. The ACSM and SEMS were employed for other experiments.

$$\frac{[f_{\mathrm{unsaturated\_fatty\_acid}}]}{[f_{\mathrm{stearic\_acid}}]} = \frac{[f_{\mathrm{unsaturated\_fatty\_acid\_0}}]}{[f_{\mathrm{stearic\_acid\_0}}]} \exp\left(-k_2 P_{O_3} t\right)$$

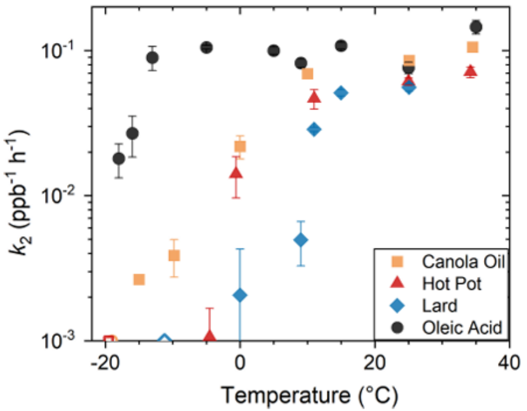
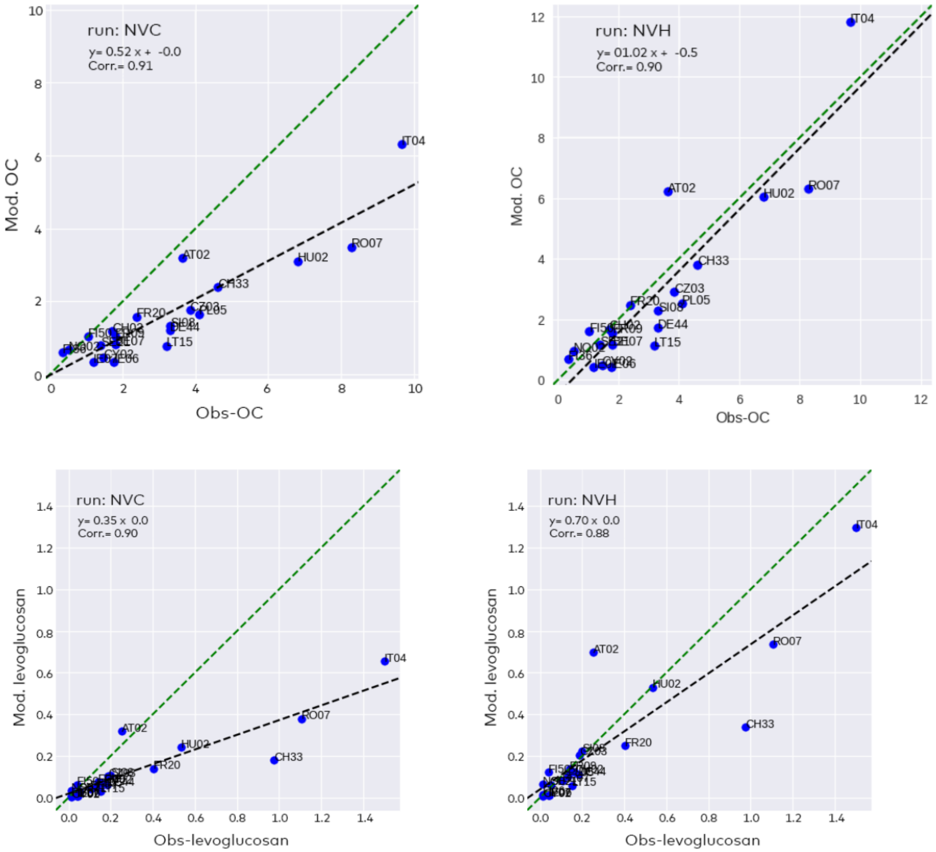


Figure 6. Dependence of k_2 on temperature. Data for oleic acid were retrieved from our previous study (Liu et al., 2023). The values of k_2 for Exp. no. 7, 10 and 21 were unmeasurably slow. Thus, the corresponding data are shown as open symbols at the bottom.

Larger Wood-Burning Emission Factors Solve Bias

Funded by the European Union



- Plots show:
  - OC (top row), for central (left) and high (right) RWC emission scenarios. (Note: inert BBOA assumed!)
  - Same, for Levoglucosan (bottom).
- High emissions seems to work better, though not for all sites.
- OC bias could also be due to chemical issues, but levoglucosan more convincing evidence of low emissions?
- Also, levoglucosan should decay, so results may be evidence of even more RWC in real life!
- Caveat: the assumed OM/levo ratio can also be inaccurate. This ain't simple!

From Simpson, Kuenen, et al., NMR report 2022.