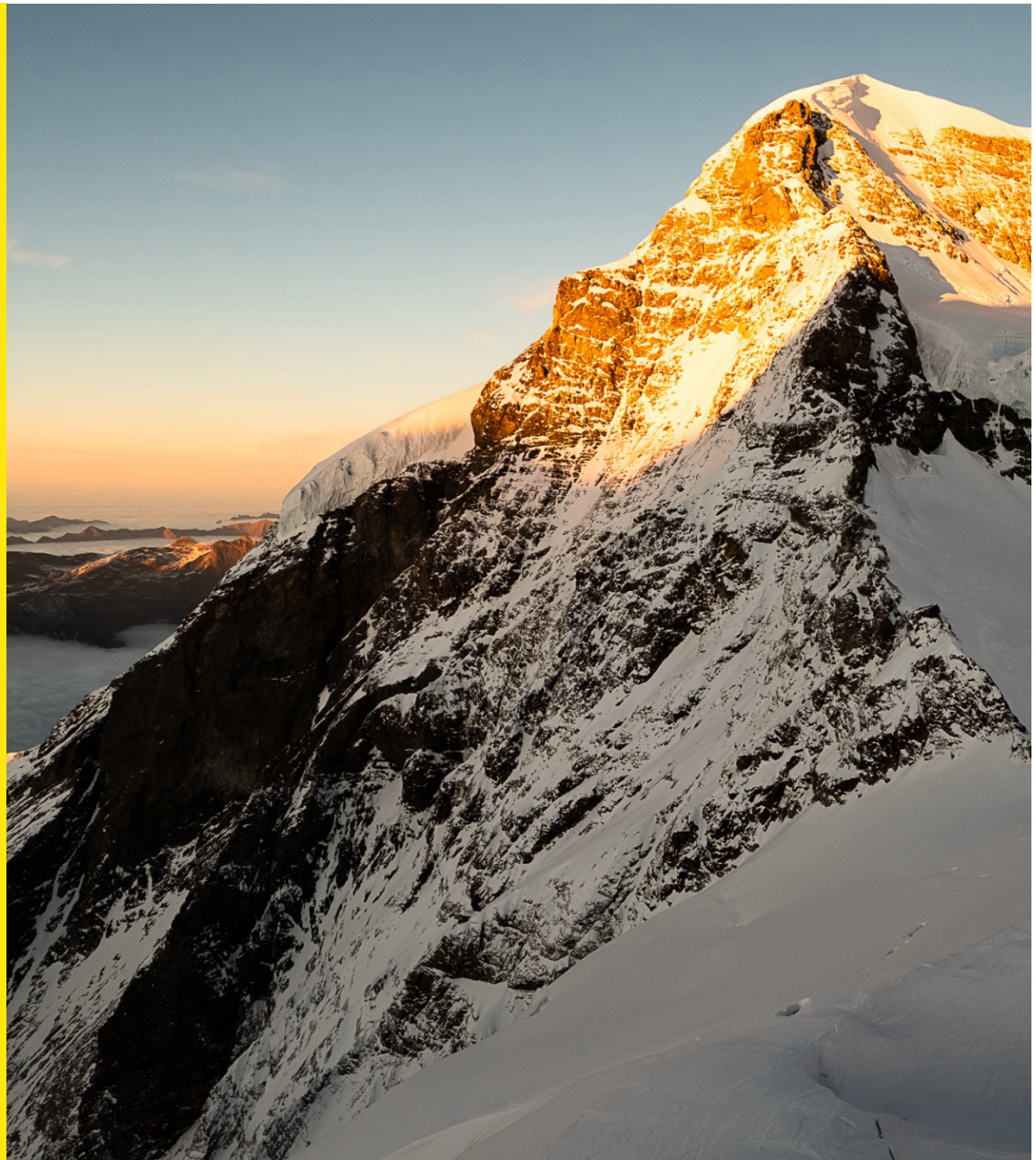
University of Applied Sciences and Arts Northwestern Switzerland
School of Engineering

Enhancing total carbon quantification using fast-thermograms



Alejandro Keller, P. Specht, P. Steigmeier, E, Weingartner

13 June 2024

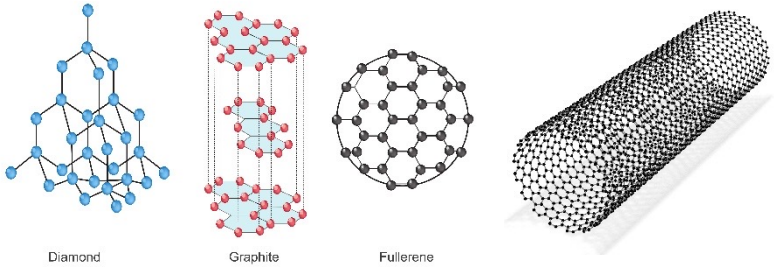
Importance of Carbonaceous Aerosols

- Between 20% and 50% of the total aerosol mass (Contini, 2018)
- 10 million premature deaths from combustion particles annually
- Complex effect on climate (composition, mixing state, location, deposition…)
- Global carbonaceous aerosol budgets and trends remain poorly characterized due to limited observations (IPCC AR6; 2023)

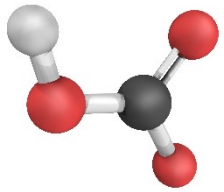
Complexity of Carbonaceous Aerosols

6 12.0102
C
CARBON

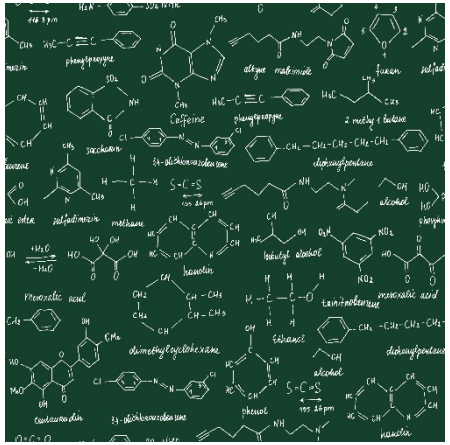
Allotropes (and amorphous carbon)



Carbonates
(and inorganic carbon)



Organic compounds



	Elemental composition, % wt				
	C	H	O	N	Other
Engine Soot (after removing oil components)					
A	85.7	3.0		0.4	0.9
B	80.3	2.6		0.6	0.3
C	91.4	2.9	4.1	0.4	1.4
D (H:C = 0.34)	92.6	2.6	3.3	0.4	1.1
Exhaust Soot					
E (H:C = 0.19)	43.4	0.7		0.3	14.3

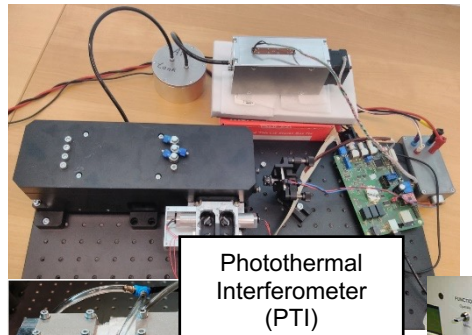
Clague, 1999, doi: 10.1016/S0008-6223(99)00035-4

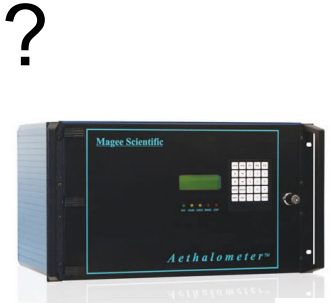
How are Carbonaceous Aerosols Measured?

- Light Absorption Methods
  - Units 1/Mm (inverse megameters)
  - Material and wavelength dependent
  - Using Mass Absorption Coefficient:

    Equivalent black carbon (eBC)
    Refractory black carbon (rBC)
    Equivalent brown carbon (eBrC)

- Thermal-Optical Methods
  - (regional discrete) Heating protocol
  - Separates total carbon (TC) into organic carbon (OC) and elemental carbon (EC)
  - Separation prone to artifacts





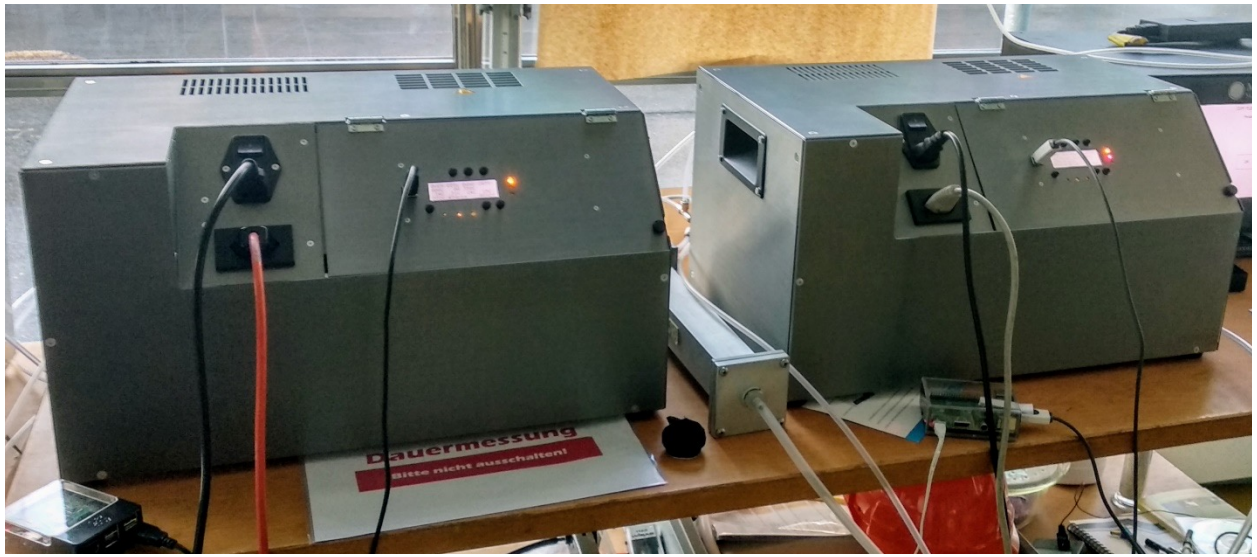
TC = all C atoms (=OC+EC)

Subfractions are defined by sample behavior:
OC ≠ C in organic compounds
EC ≠ eBC ≠ rBC
eBrC?

Novel Approach: FATCAT

FAst Thermal CArbon Totalizer
Keller, 2023; https://doi.org/10.5194/ar-1-65-2023

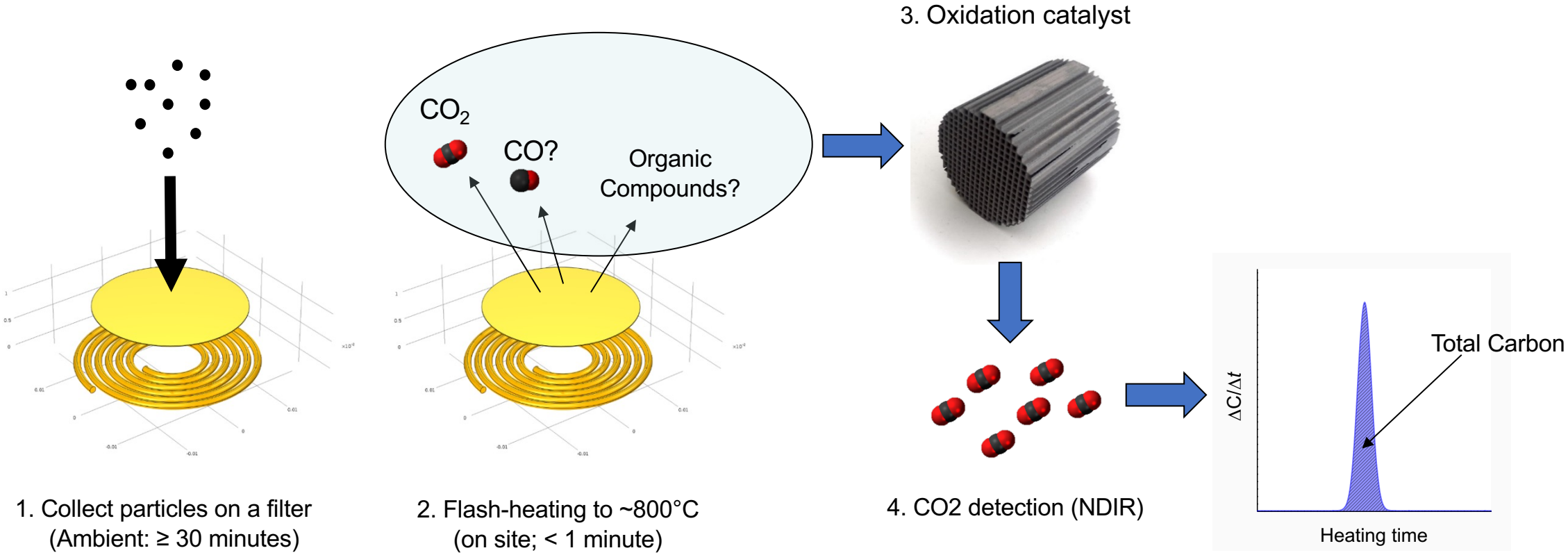
- Goal: Provide an accurate, standalone method for measuring aerosol-bound total carbon (i.e. TC).
- Applications: Atmospheric observations and emission monitoring.



Novel Approach: FATCAT

FAst Thermal CArbon Totalizer
Keller, 2023; https://doi.org/10.5194/ar-1-65-2023

- **Unique:** Rigid metal filter (no filter displacements or leaks)
- **Unique:** Direct and homogeneous heating of the filter (instead of using a heating filament or a furnace)
- **Unique:** Calibration performed using CO_2 (other devices use a sugar solution) and through calibration of a mass flow controller
- Most precise instrument: Limit of detection LoD=0.1 µg-C

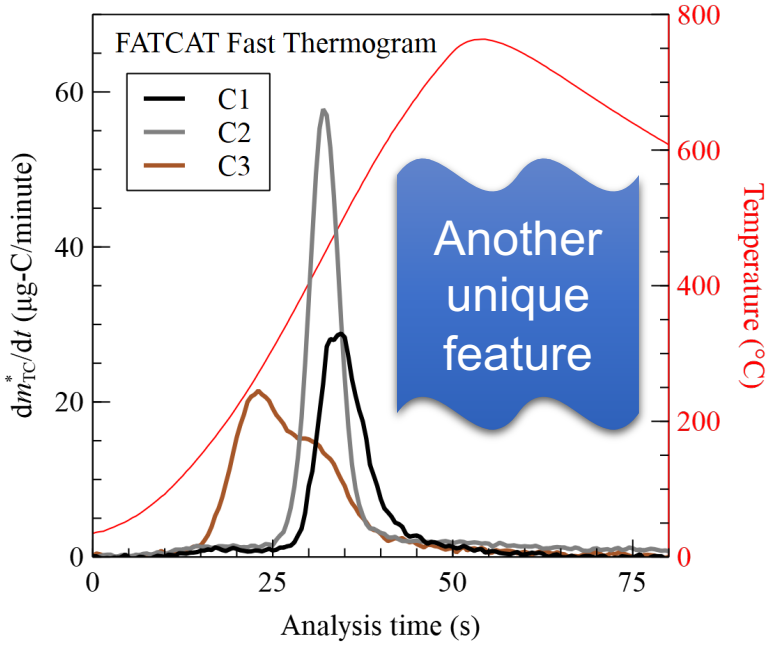


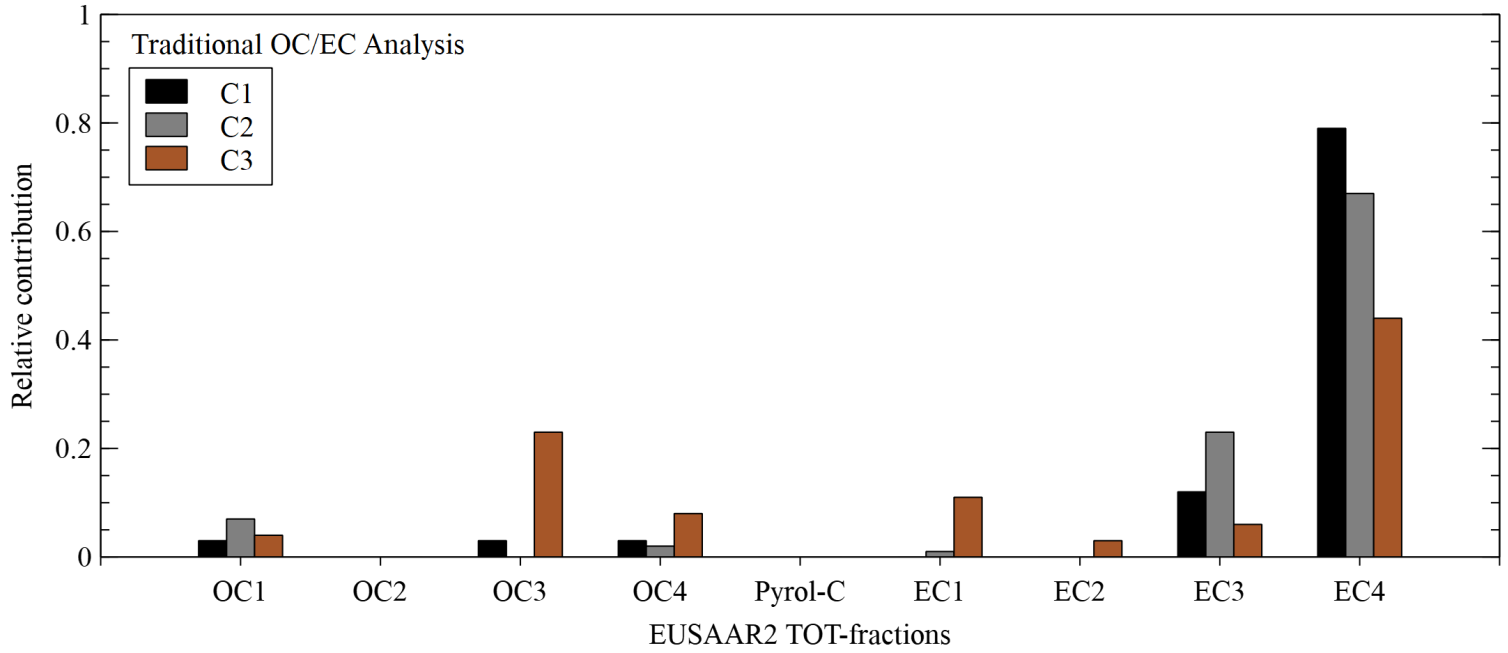
1. Collect particles on a filter (Ambient: ≥ 30 minutes)

2. Flash-heating to ~800°C (on site; < 1 minute)

4. CO2 detection (NDIR)

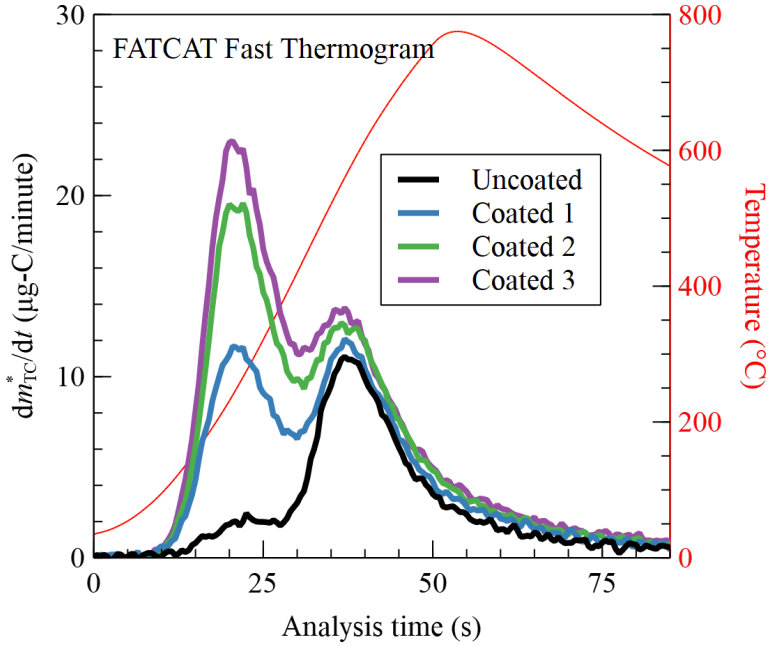
Features of Fast Thermograms (diffusion flame soot)

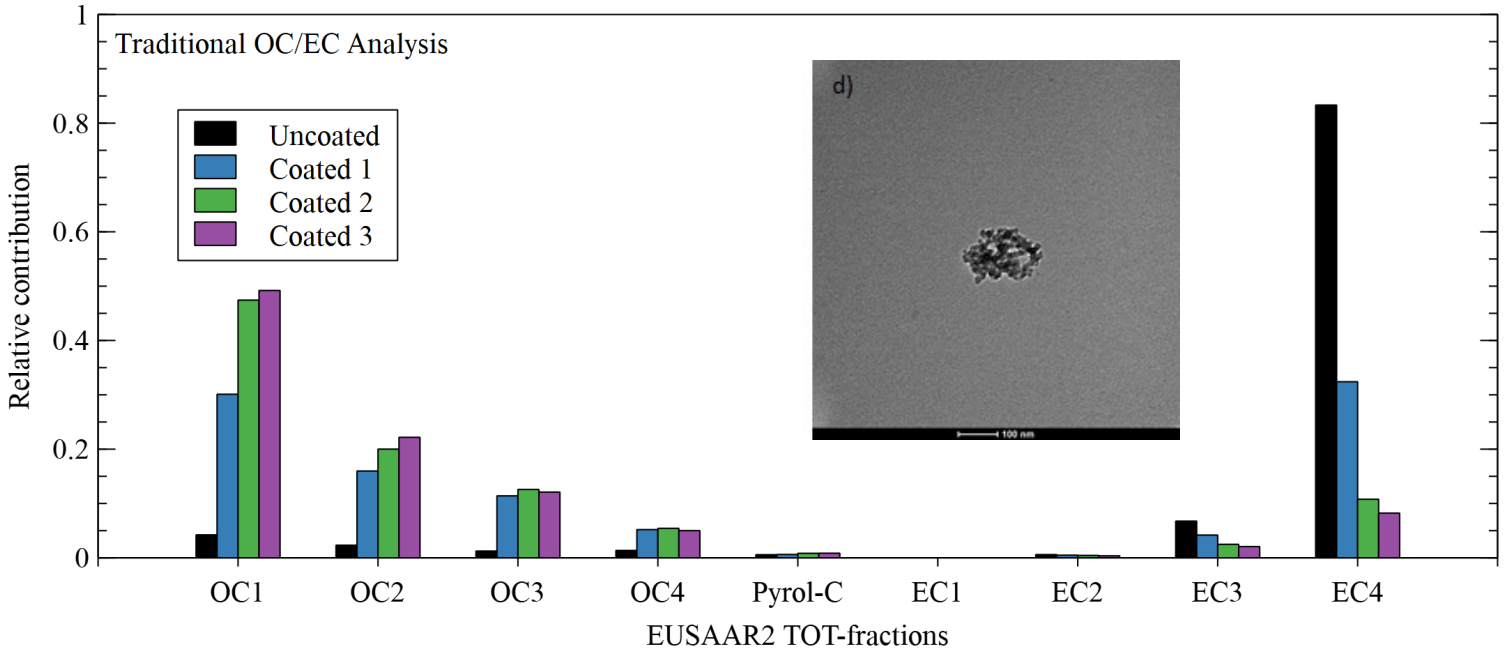


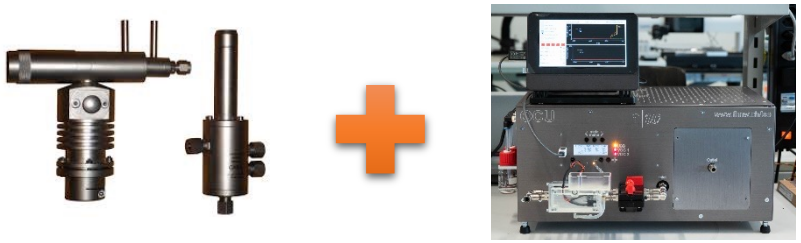


	Type	Fuel C/O	TC (µg-C)	EC/TC	Diameter, d_{pg} (nm)
C1	Black Soot	0.26	5.2	0.91	72
C2	Black Soot	0.25	4.4	0.91	150
C3	Brown Soot	0.41	5.6	0.65	29

Fast Thermograms (Soot + Secondary Organic Coating)







Experiment	TC (µg-C)	EC/TC	Diameter, d_{pg} (nm)
Uncoated	3.9	0.84	88
Coated 1	6.4	0.37	90
Coated 2	8.3	0.13	111
Coated 3	9.4	0.10	126

Coating Unit: Keller, 2022; doi:10.1080/02786826.2022.2110448

Carbon Balance Campaign 2022 at JFJ

Slide: **Nora Nowak (PSI)**
ACTRIS Science Conference 2024 - May 16, 2024

Aim: Achieve closure for gaseous and particulate carbonaceous species

Gas Chromatographs:

Total Carbon Analyzers:

Proton Transfer- and Aerosol Mass Spectrometers:

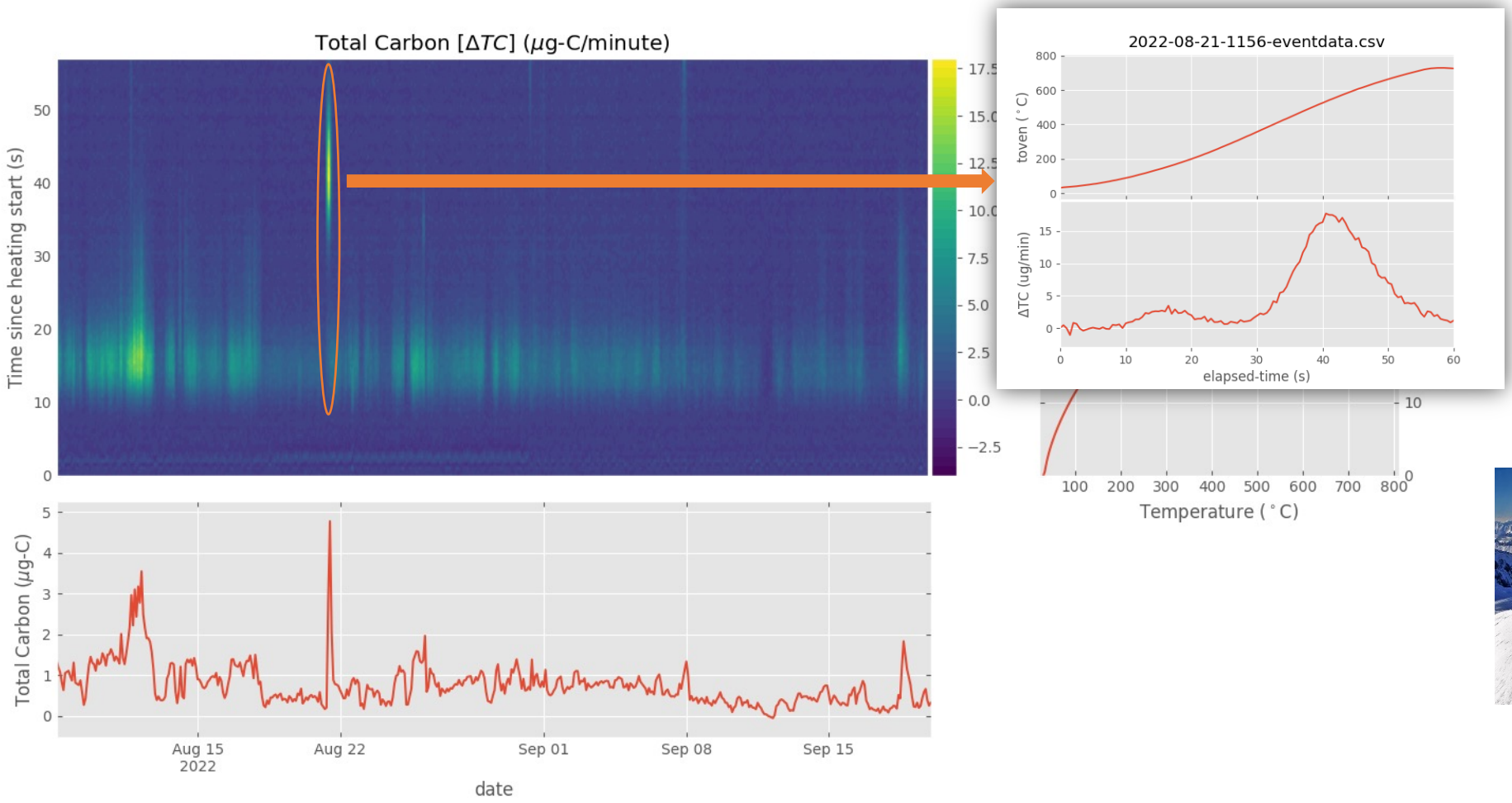
Infrastructures and Trans-National Access:

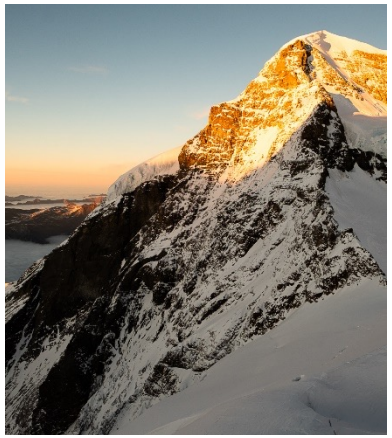
Jungfraujoch – Sphinx Observatory – Site Characteristics

Slide: **Nora Nowak (PSI)**
ACTRIS Science Conference 2024 - May 16, 2024

- Lower free troposphere
- Vertical transport from planetary boundary layer during summer
- Long-range transported aerosol plumes: Saharan dust, volcanic ash, wildfire emissions
- Local emissions from tourist activities

Ambient Measurements (Jungfraujoch CBC)





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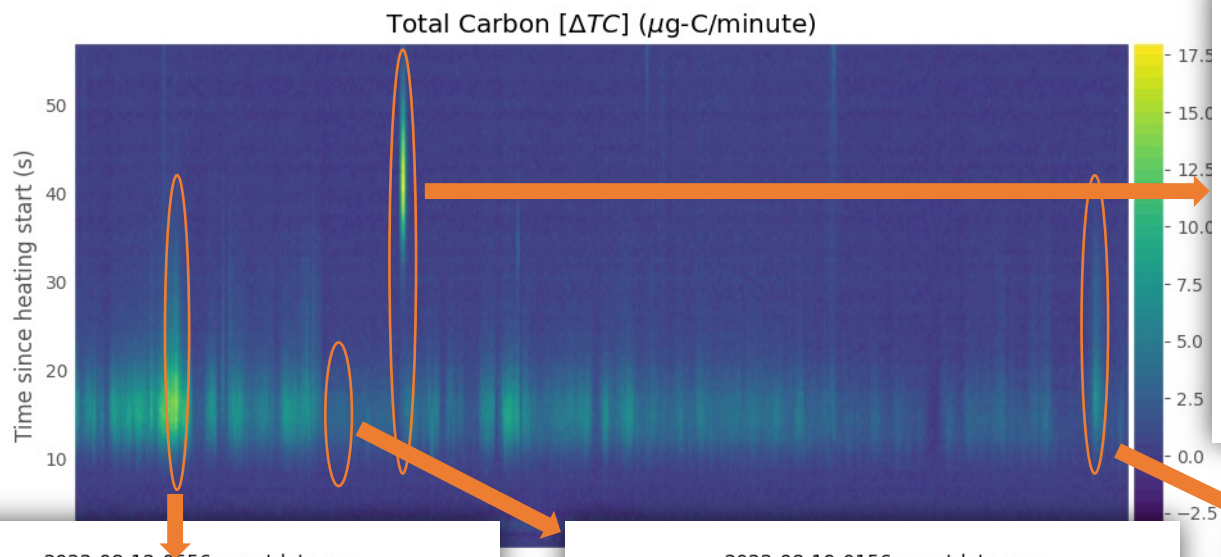
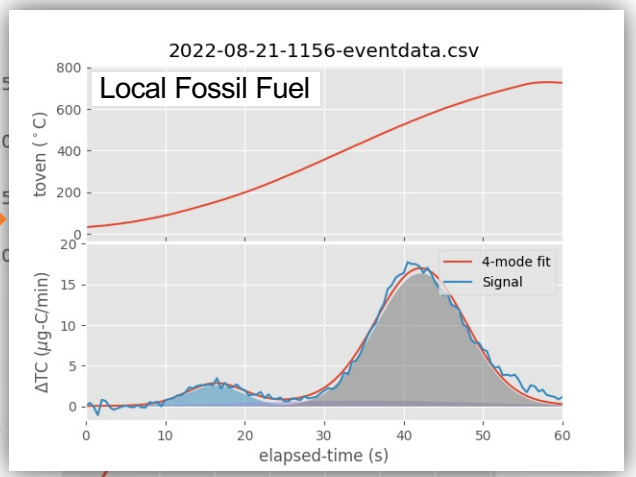
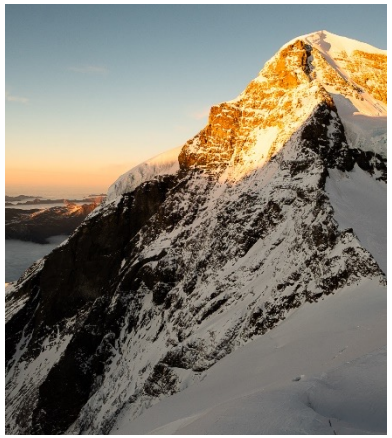
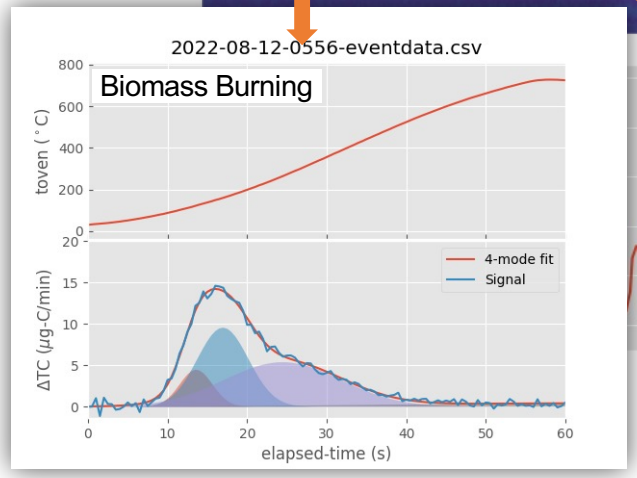
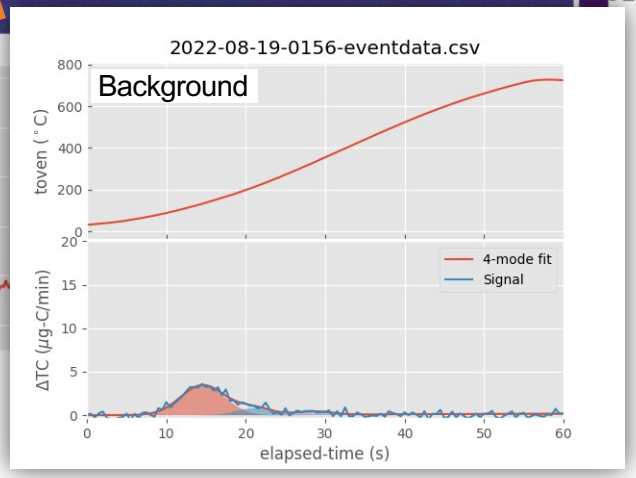
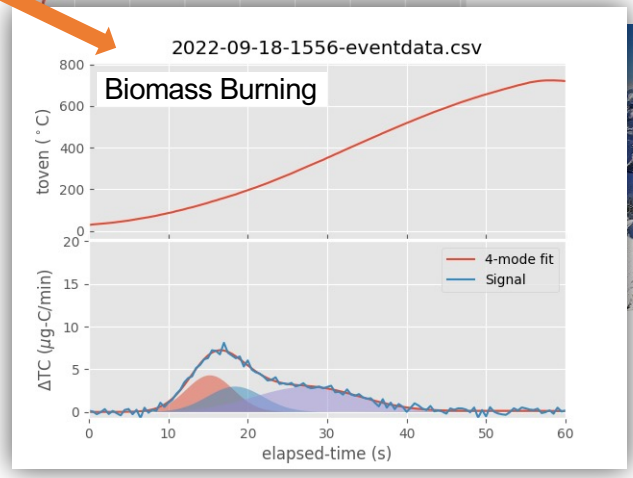






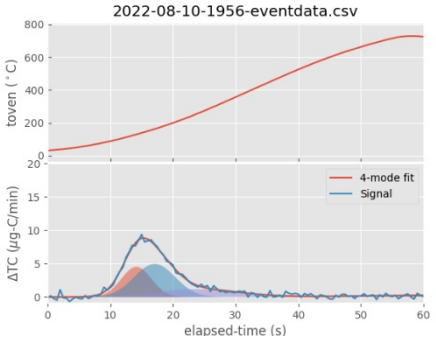


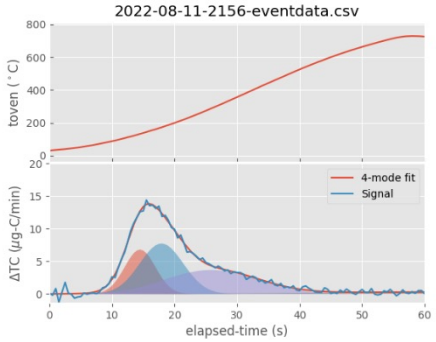


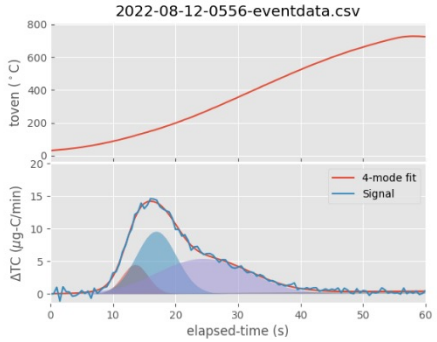


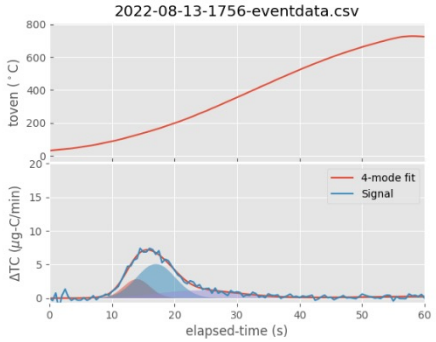
Biomass Burning Candidates

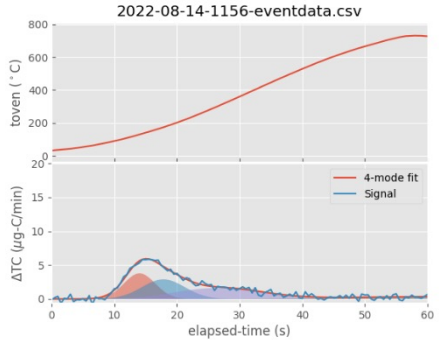
- Criteria includes goodness of the fit, area of the component and wide of the distribution.
- 38 samples (11 days)

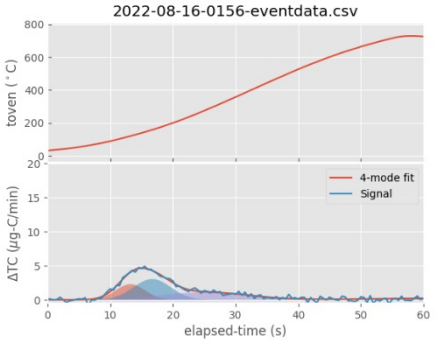


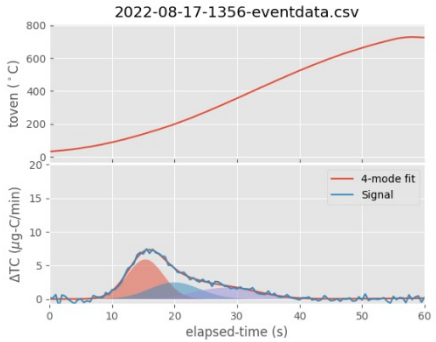


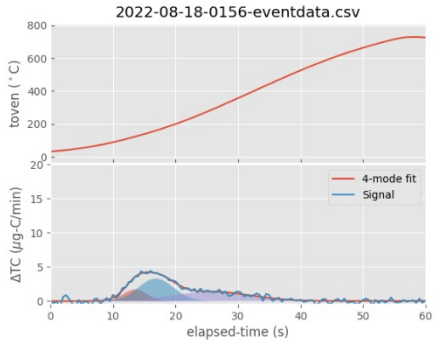


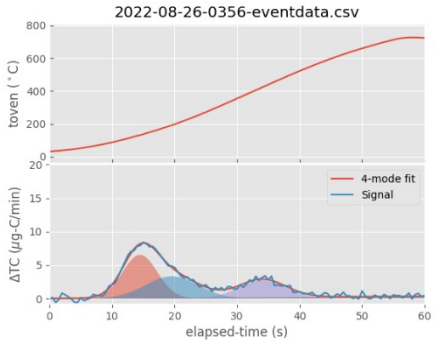


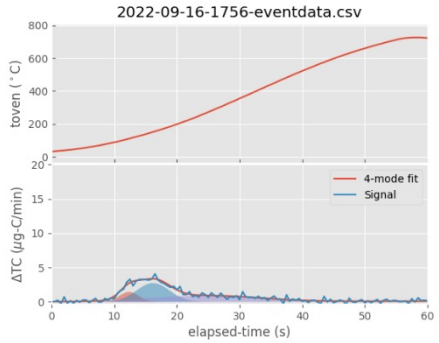


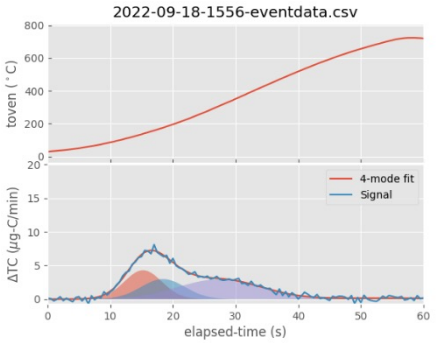






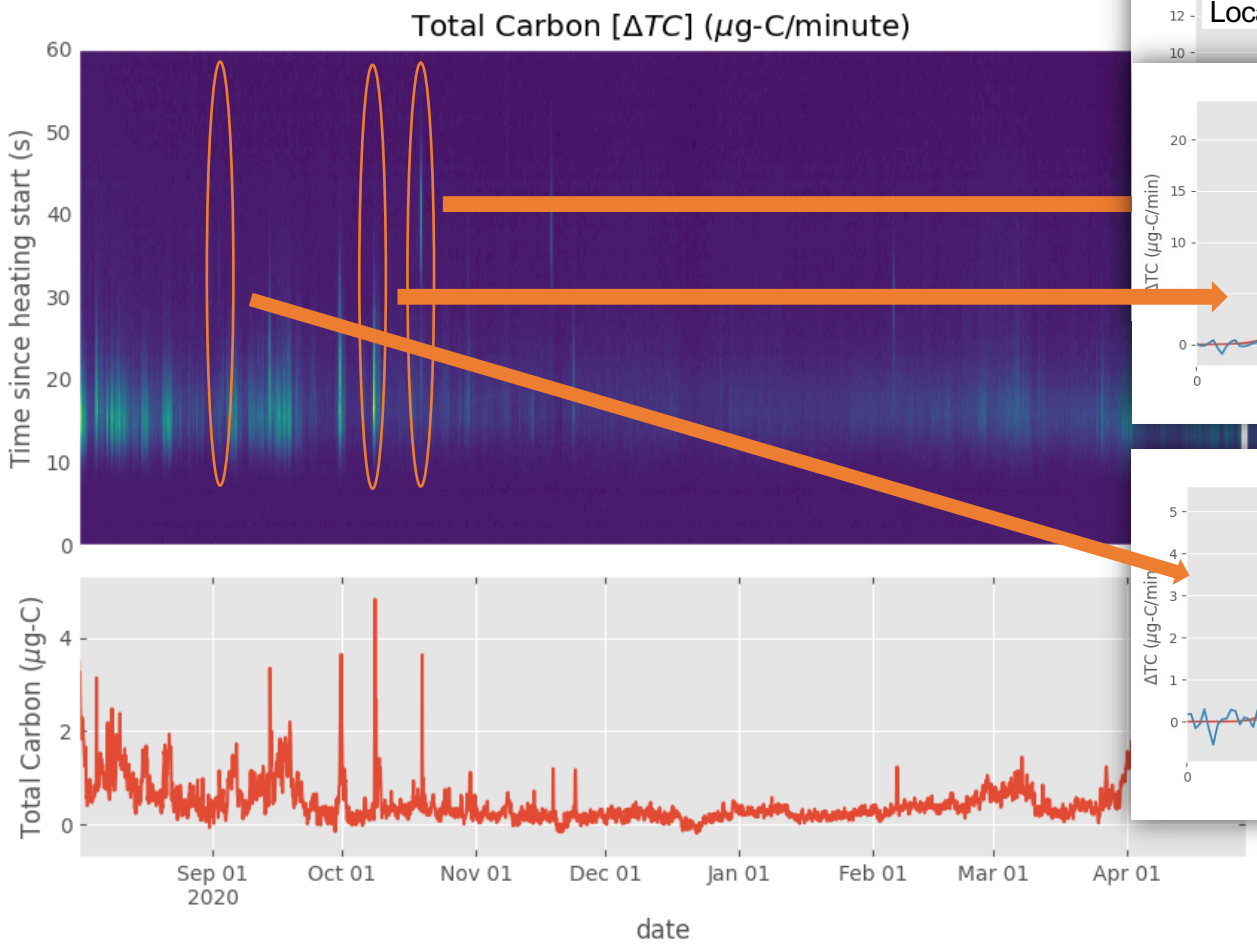


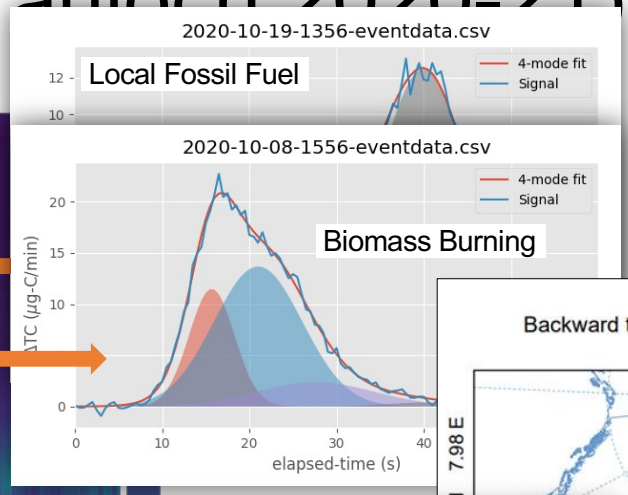


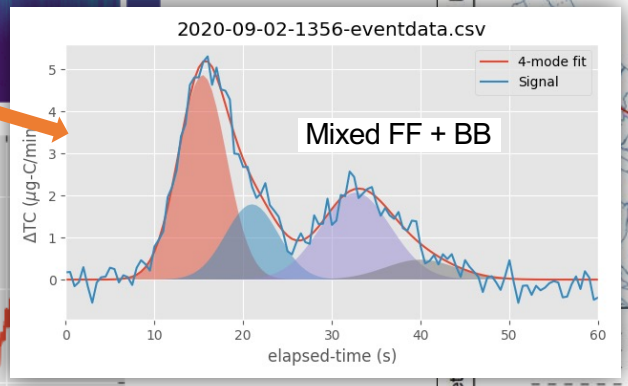


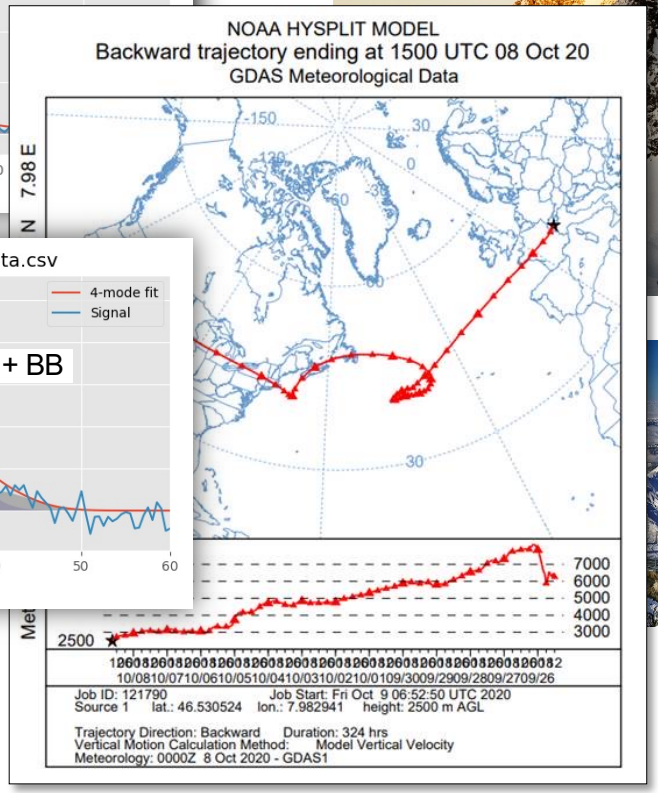
Ambient Measurements (Jungfraujoch 2020-21)

GAW-CH Science Projects 2018-2021









To do

- Collect more ambient data… and analyze collected data:
  - Jungfrauchjoch (Lower troposphere)
  - Zurich (Urban, busy street)
  - Payerne (Suburban, background)
  - Windisch (urban background)

- Evaluate different tools for data analysis

- Laboratory tests with model substances

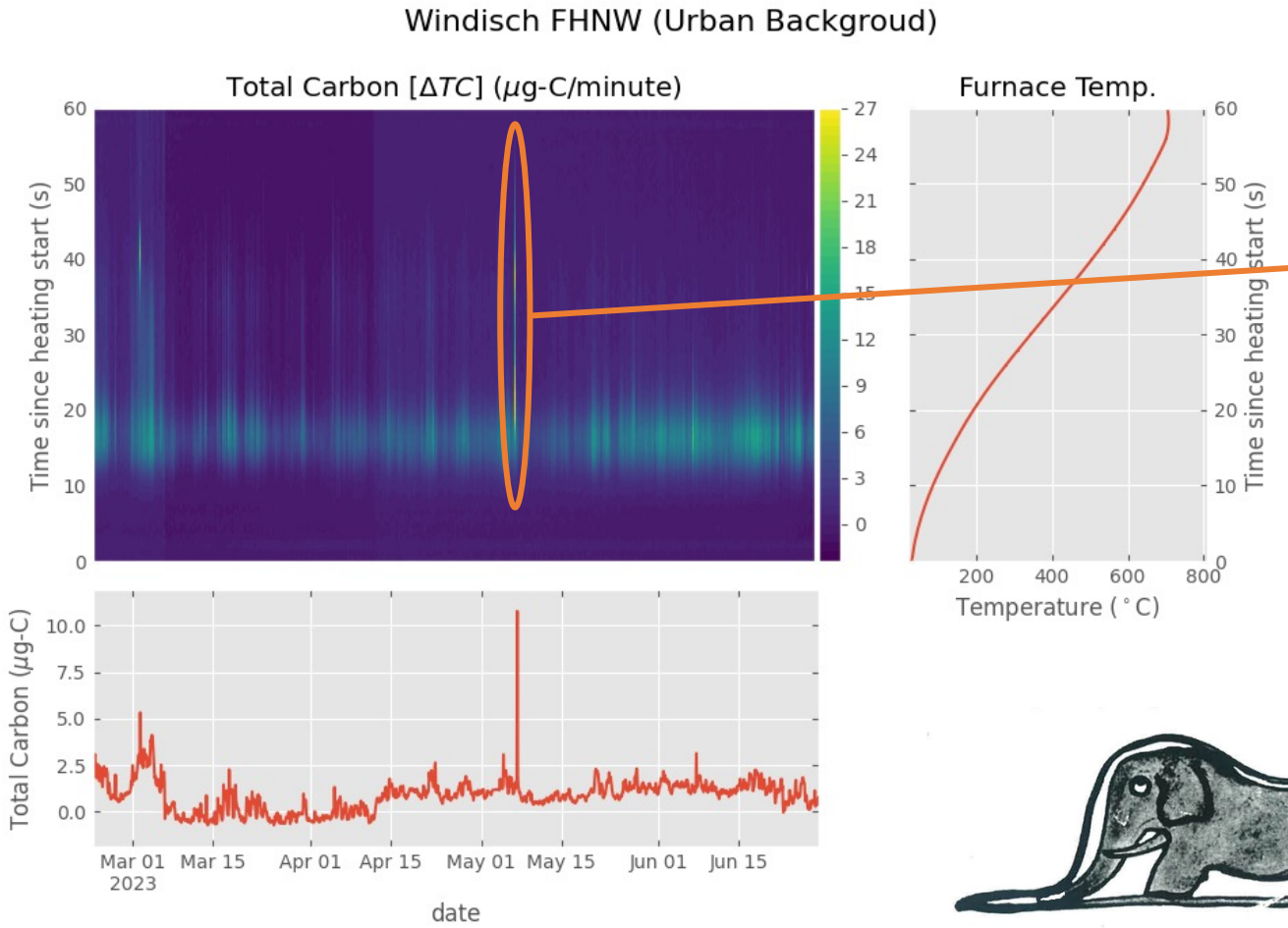
- Hardware improvements (e.g., direct measurement of filter temperature)

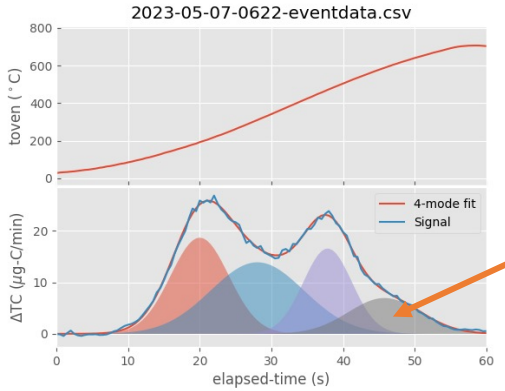
Take home

- Carbonaceous aerosols (CA) comprise a wide range of substances with a continuum of properties (thermal, optical, etc.)
- This complexity cannot be capture by a few discrete categories

- FATCAT provides a unique way of looking at CA:
  - Based on the continuous analysis of thermal-refractivity
  - Fast generated data and thermograms can be analyzed on the spot
  - Validated through laboratory studies and ambient measurements
  - Potential to be better suited for source apportionment
  - Unsupervised and low maintenance

Questions?

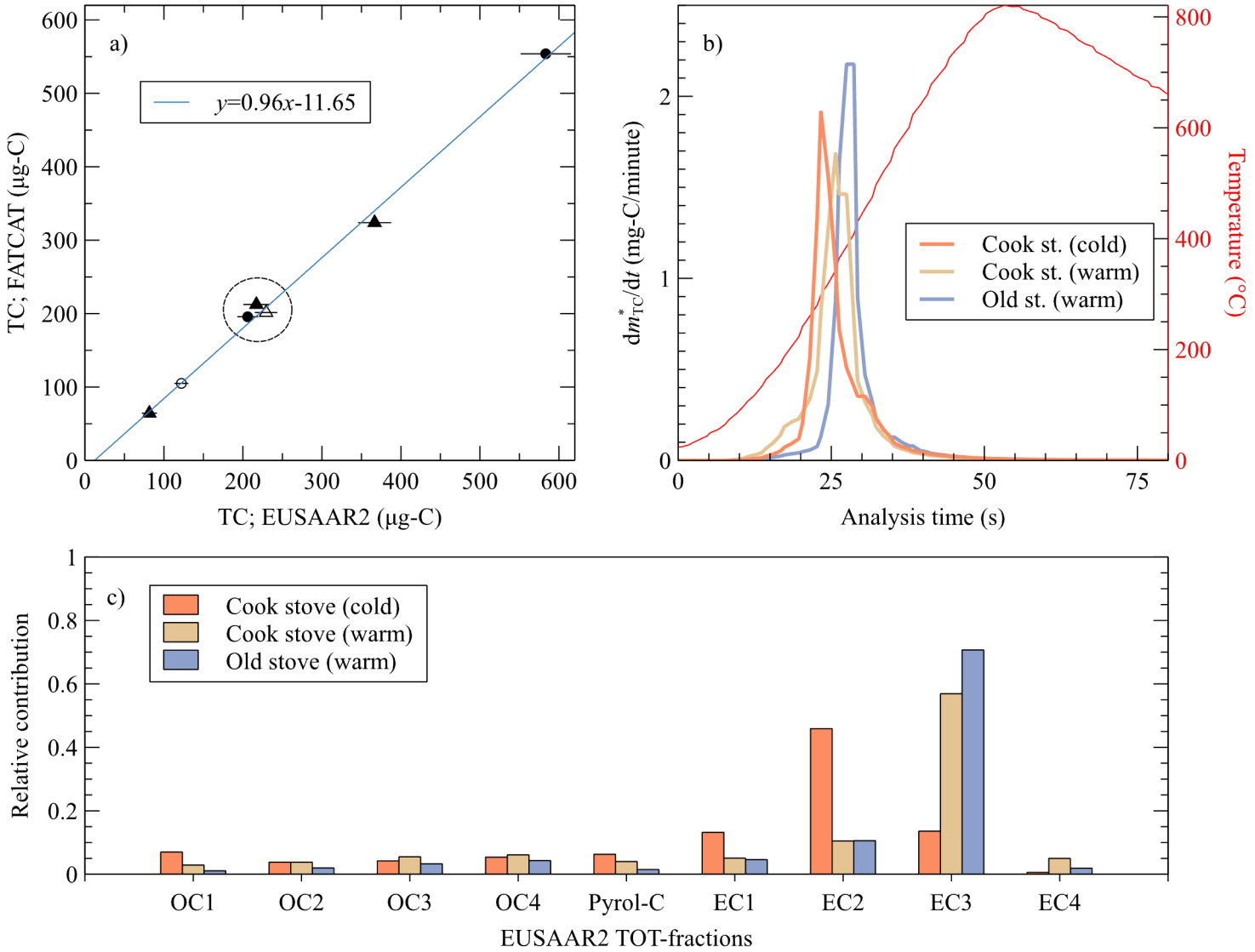




More refractory than diesel soot!

Fast Thermograms (Wood Burning)

Keller, 2023; https://doi.org/10.5194/ar-1-65-2023



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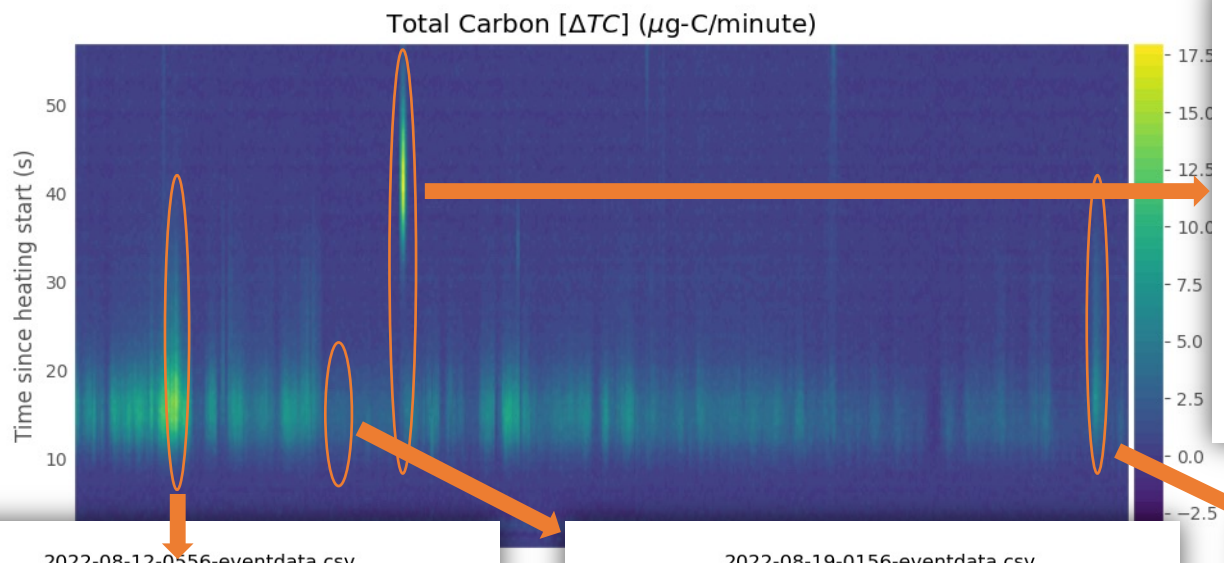
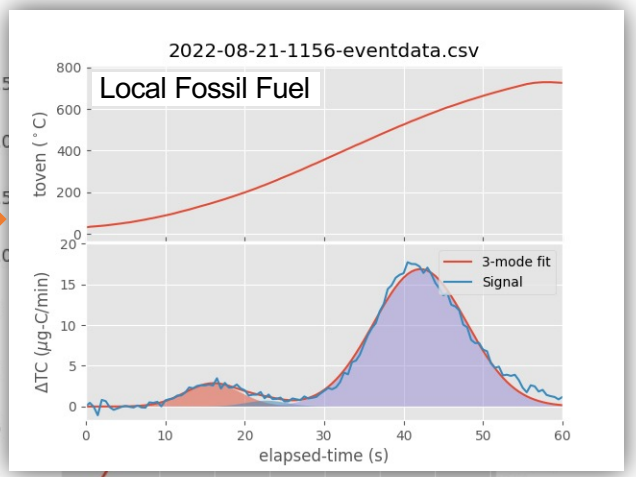
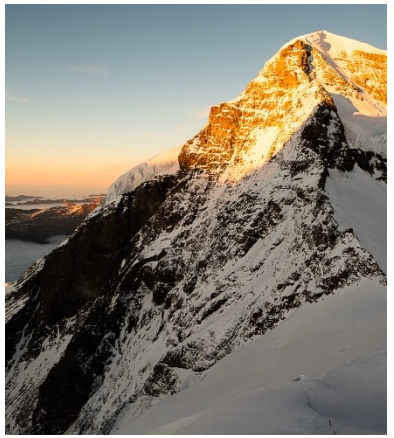
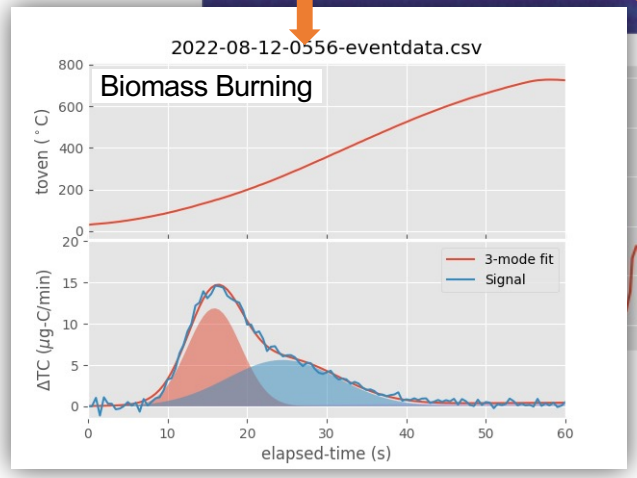
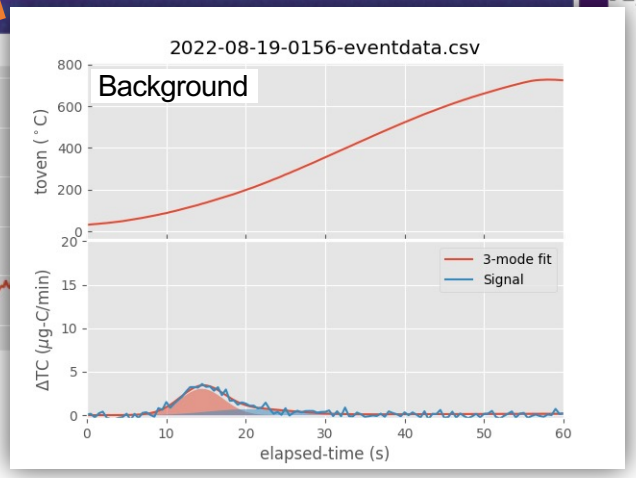
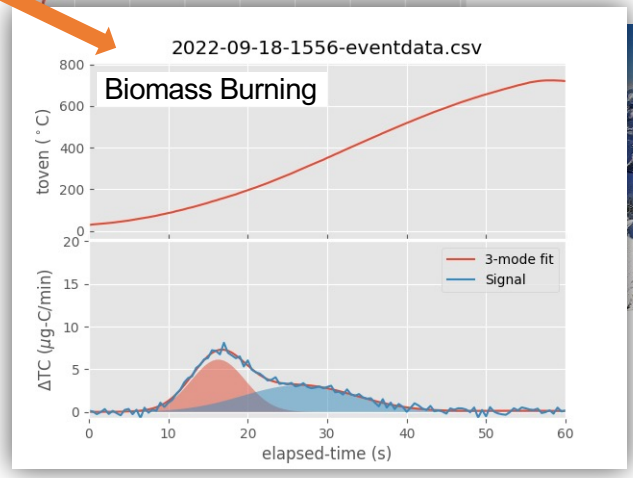




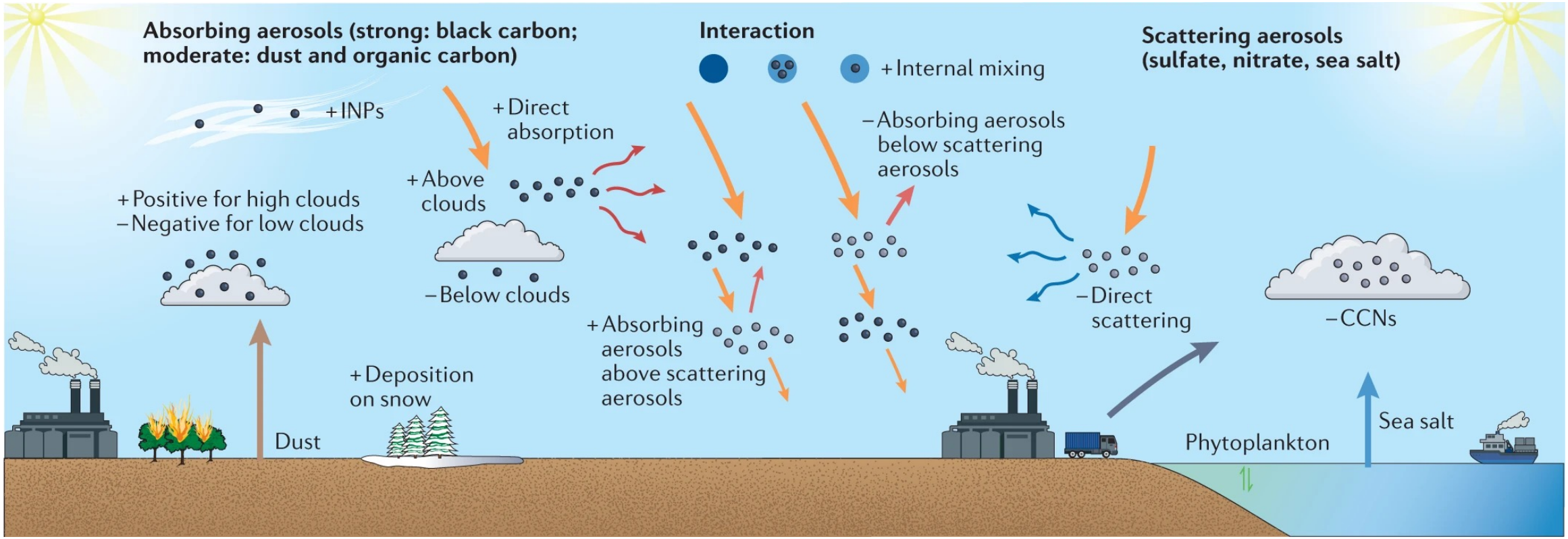








The radiative effects of aerosols



Li, J., Carlson, B.E., Yung, Y.L. et al. Scattering and absorbing aerosols in the climate system. Nat Rev Earth Environ 3, 363–379 (2022). https://doi.org/10.1038/s43017-022-00296-7

Sources of Atmospheric Aerosols