Organic aerosol components across Europe using 24 ACSM/AMS yearlong datasets and a harmonized source apportionment protocol

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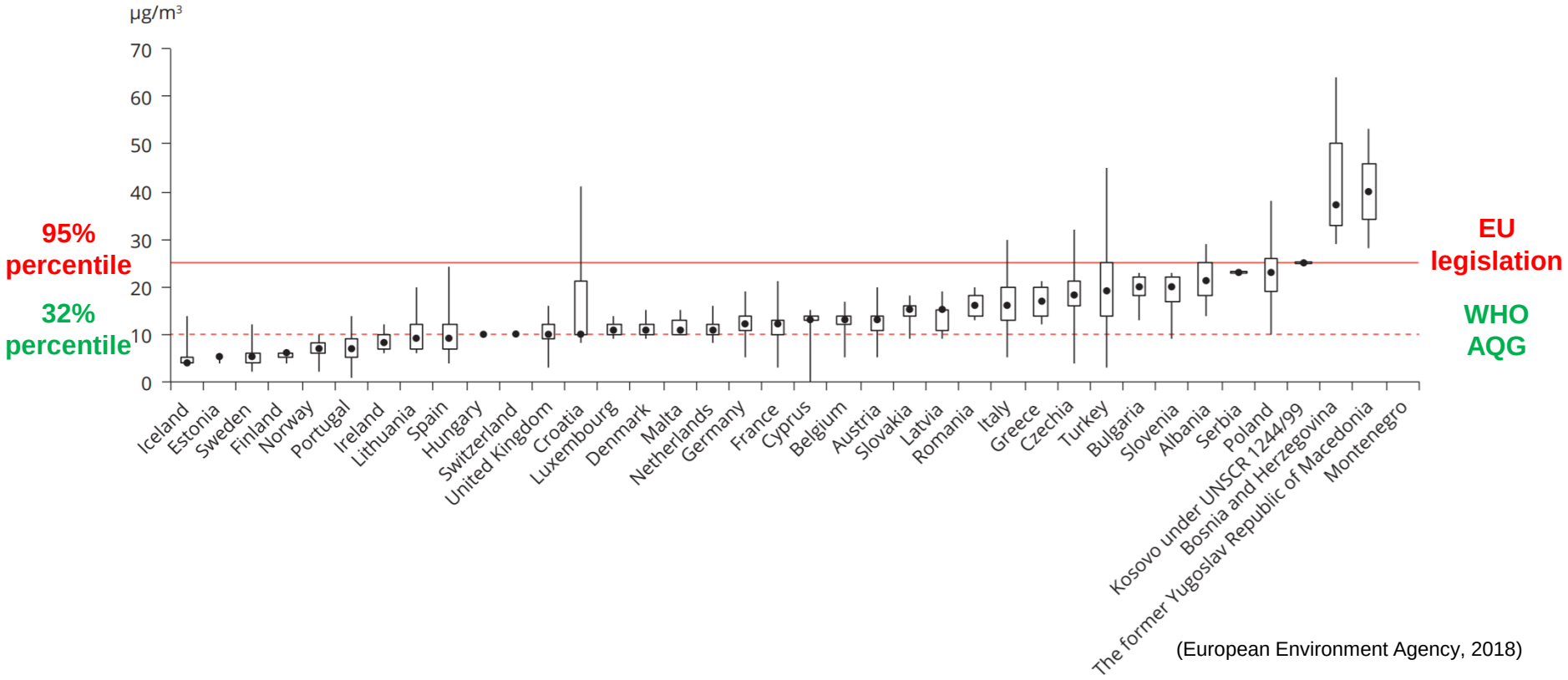
Motivations

$PM_{2.5}$ concentrations in relation to the annual limit value in 2016



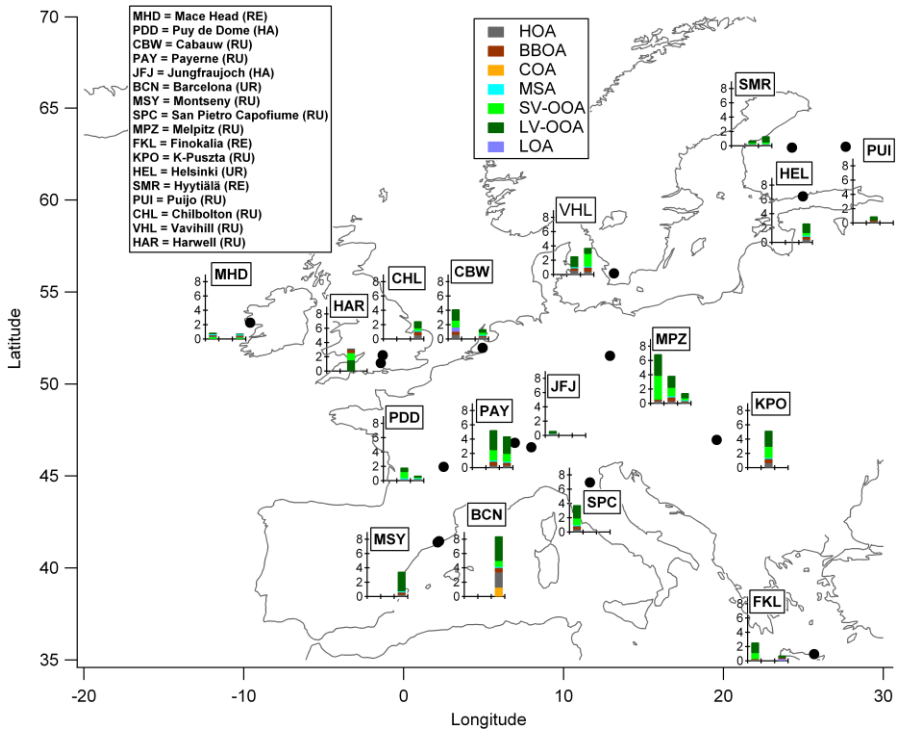
(European Environment Agency, 2018)

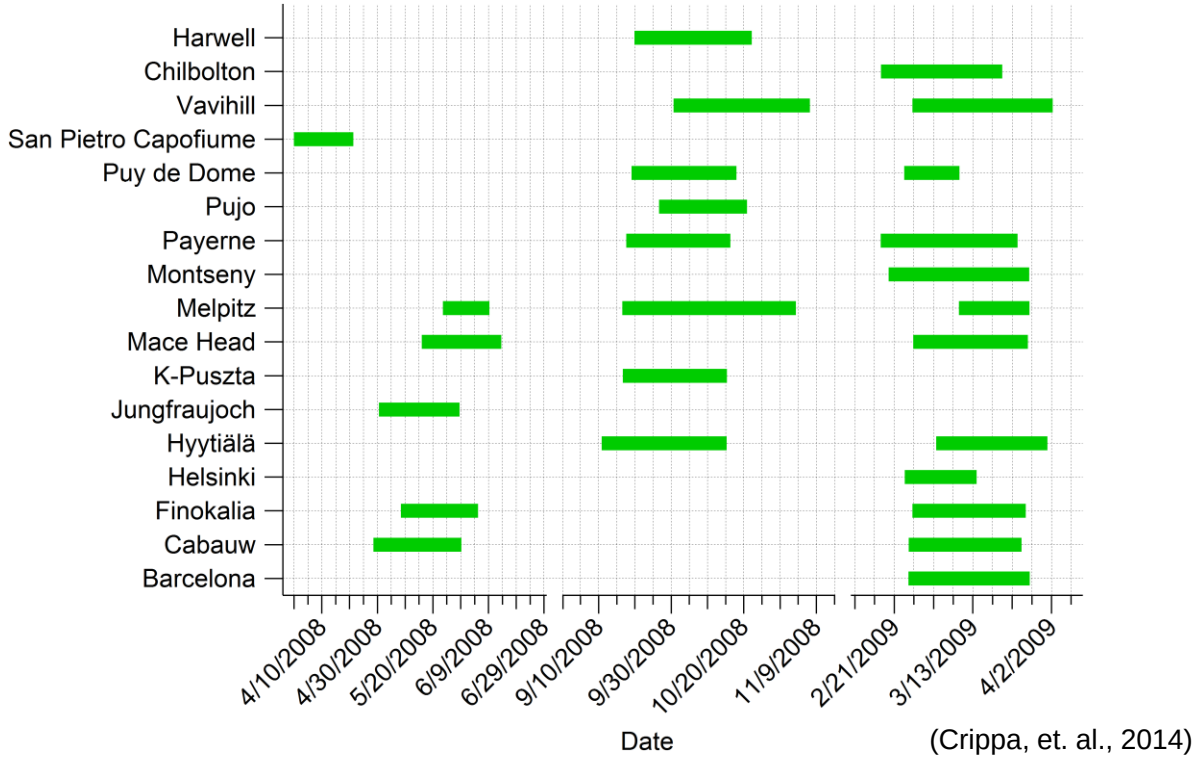
How can we reduce emissions?

The sources of organic aerosol (**20-90%** of total submicron aerosol) need to be characterized

Most-recent European Overview work

Crippa et. al., (2014) presents a spatial variabilities of organic aerosol (OA) sources with a consistent source apportionment (SA) guideline for 25 datasets collected using the Aerodyne aerosol mass spectrometer (AMS)



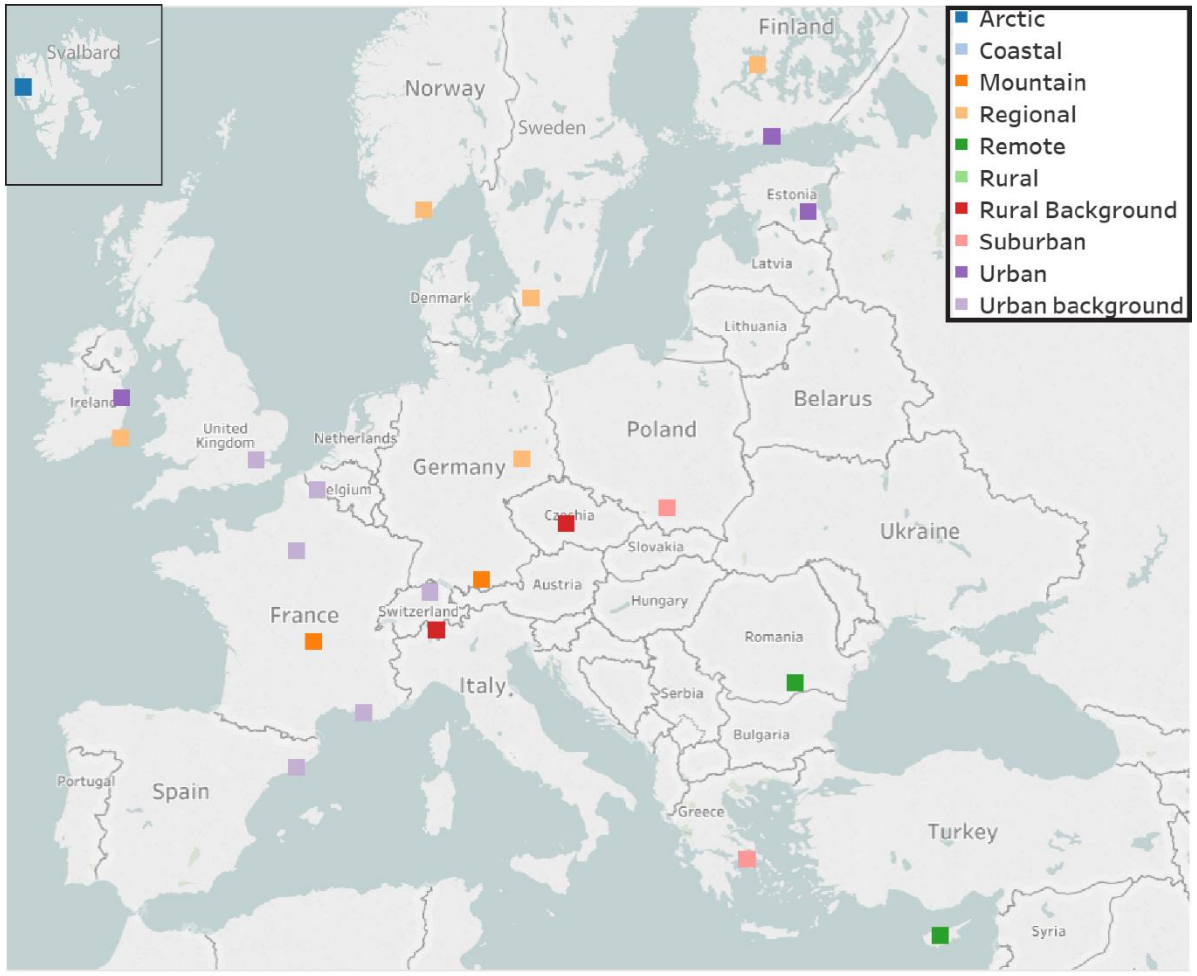


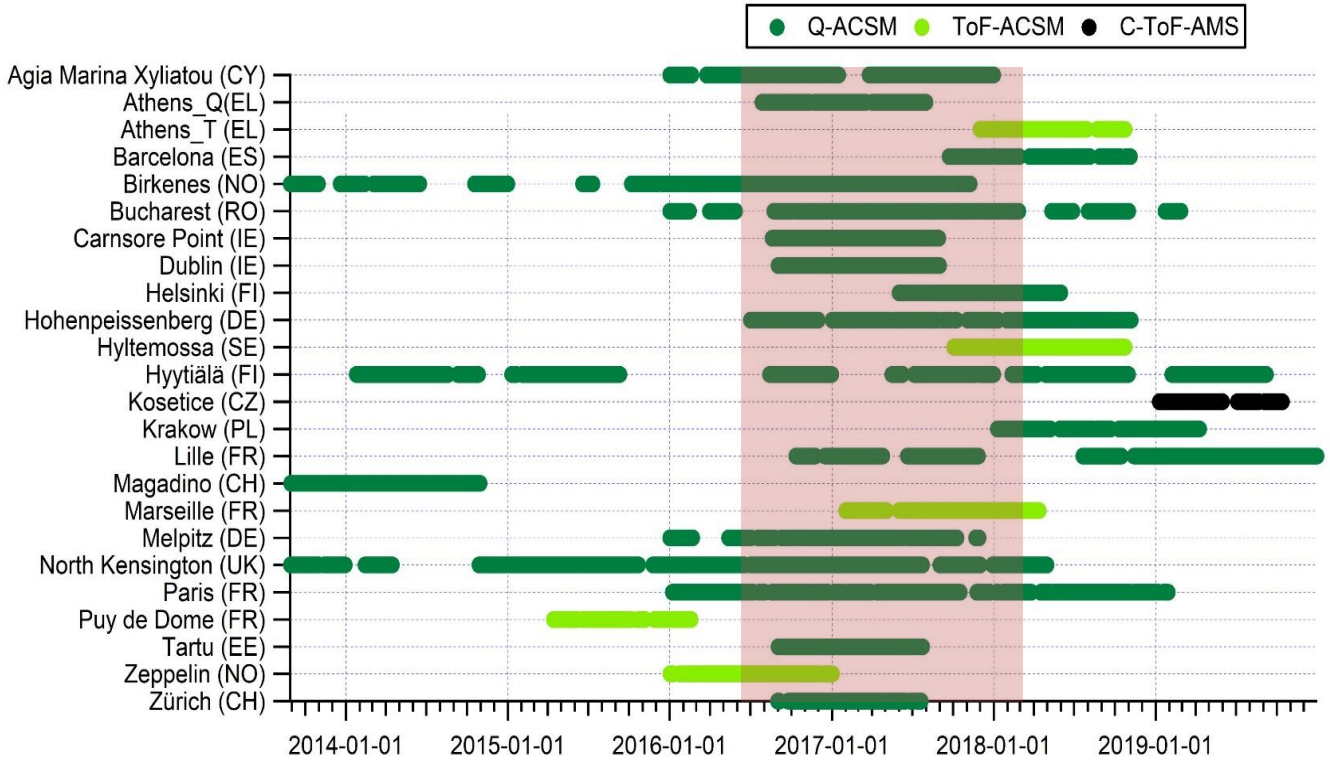
(Crippa, et. al., 2014)

Research gaps:

1. Positive Matrix Factorization (PMF) suffers from **rotational ambiguity** and requires **subjective** judgements;
2. The conventional PMF does not consider the **evolutions of OA source** profiles;
3. AMS is a labor-intense and expensive instrument, not desirable for long-term monitoring;
4. **Seasonal variations** of OA sources are still poorly understood without long-term datasets.

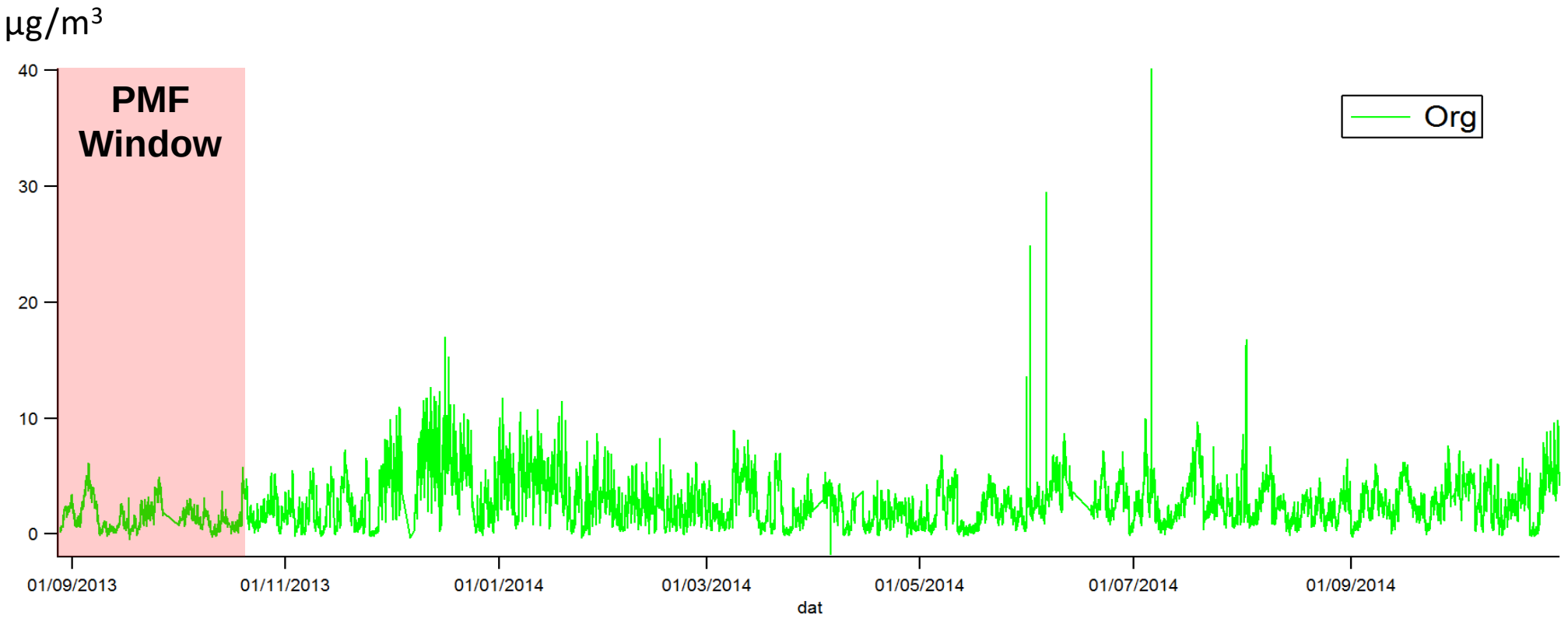
Data Coverage





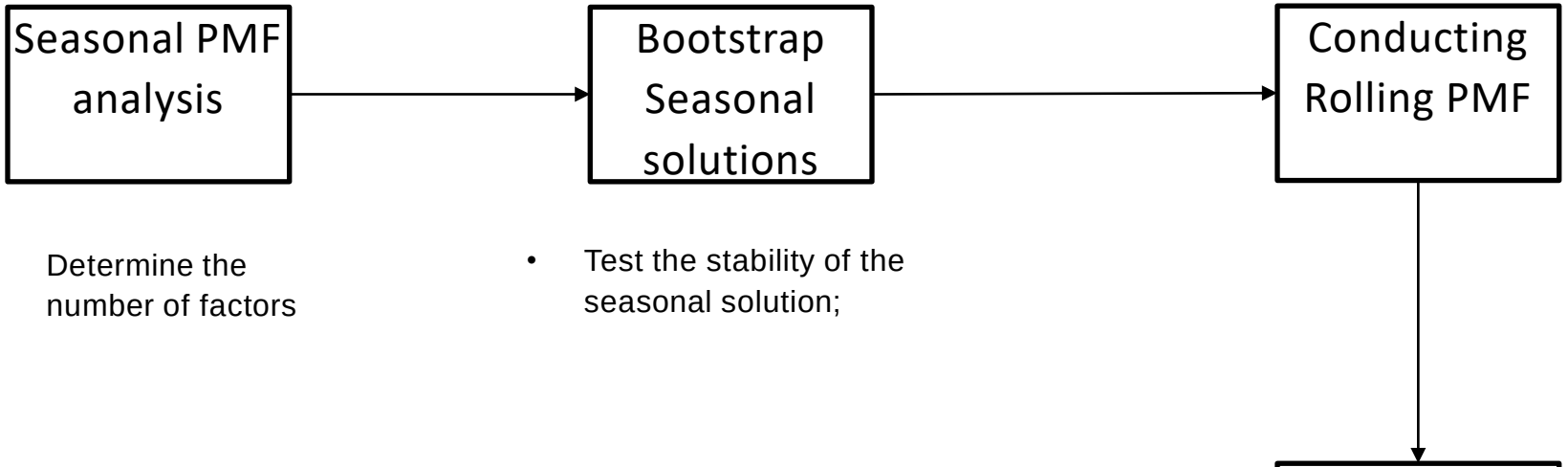
24 long-term datasets across Europe have Aerosol Chemical Speciation Monitor (ACSM)

What is rolling PMF?



- Smaller time window can **shift** over the whole PMF input (>1 year) with a step of one day for many (e.g., 50) repeats
- Take the **temporal variations** of OA sources into account
- Estimate rotational uncertainties of PMF while doing random a-values with bootstrap re-sampling
- Select “good’ runs using t-test of criteria

Standard procedures for rolling PMF



- Determine the number of factors

- Test the stability of the seasonal solution;

- Constrain the POA with reference profiles/seasonal solutions
- PMF window = 14 days
- Repeats = 50
- Step = 1 day

- Define your own criteria to select and reposition "good" PMF runs
- T-test of criteria to avoid subjective judgments

Number of runs in a year:

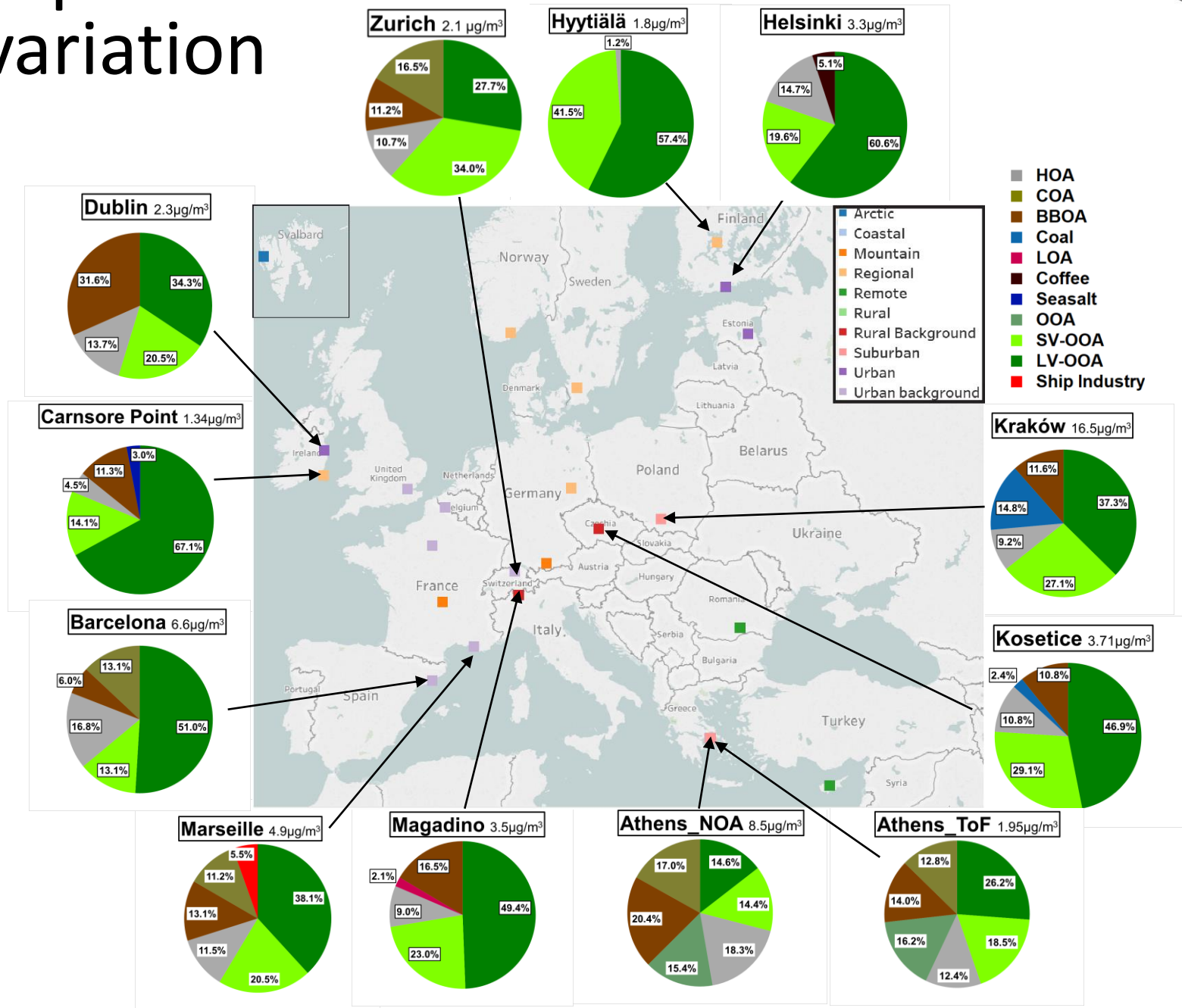
(365-14) × 50 × 2 = 36500

days Repeats 4 to 5 factors

?

A standardized protocol to analyze long-term ACSM data using SoFi Pro (Datalystica Ltd. Villigen AG Switzerland)

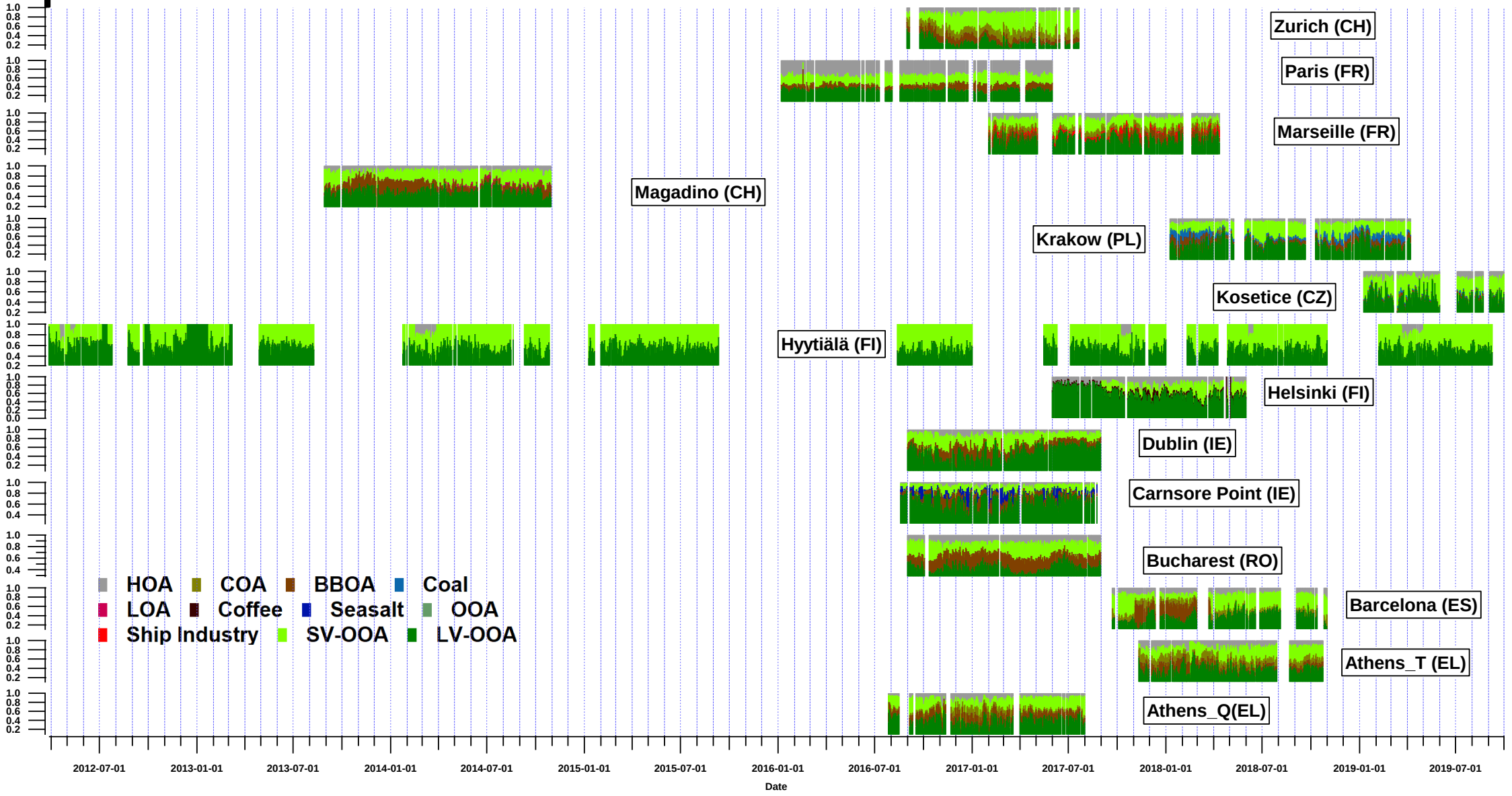
Spatial variation



- OOA is dominant
- BBOA is a considerable source in most of datasets;
- HOA contributions are alike except Hyytiala and Kosetice;

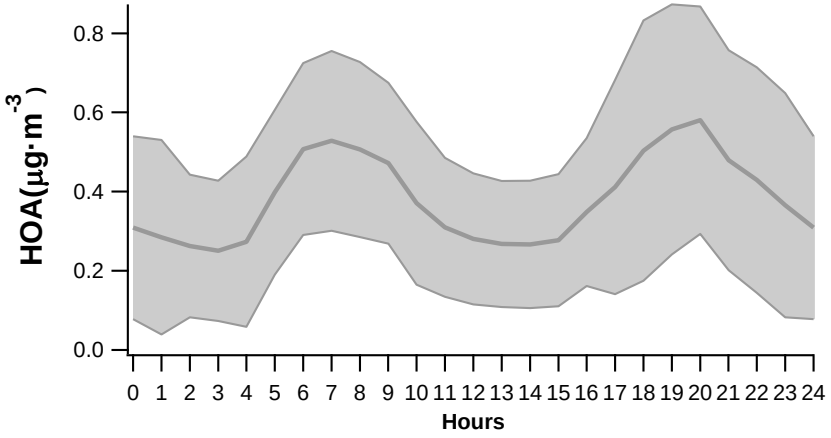
- Coal combustion source is present in eastern Europe;
- COA factor has been resolved in urban sites

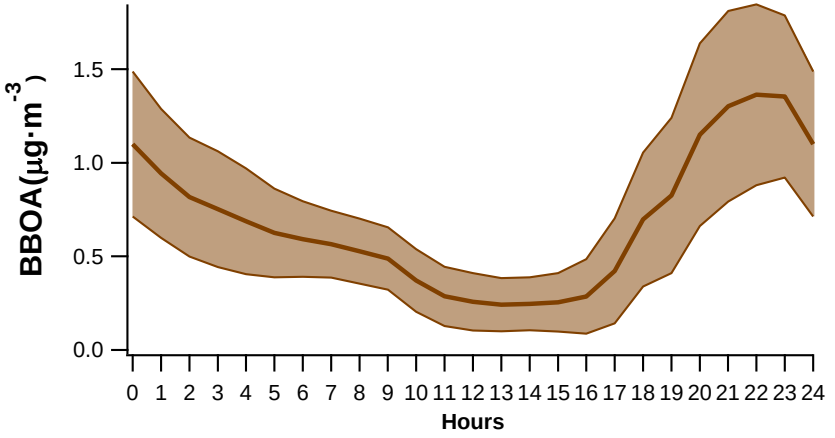
Temporal variation

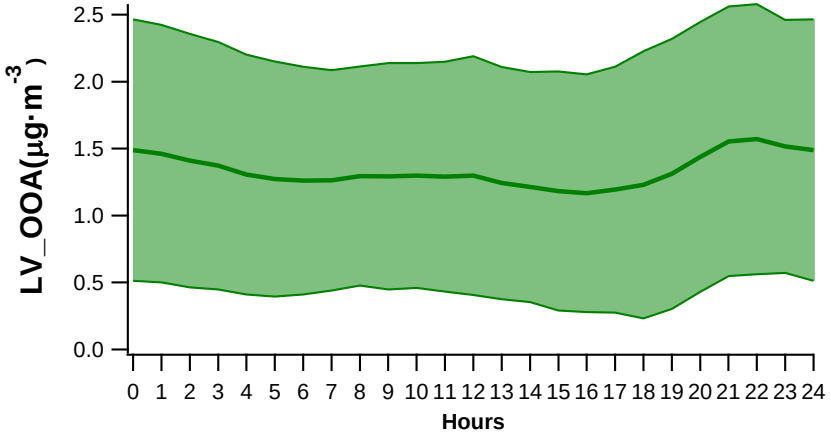


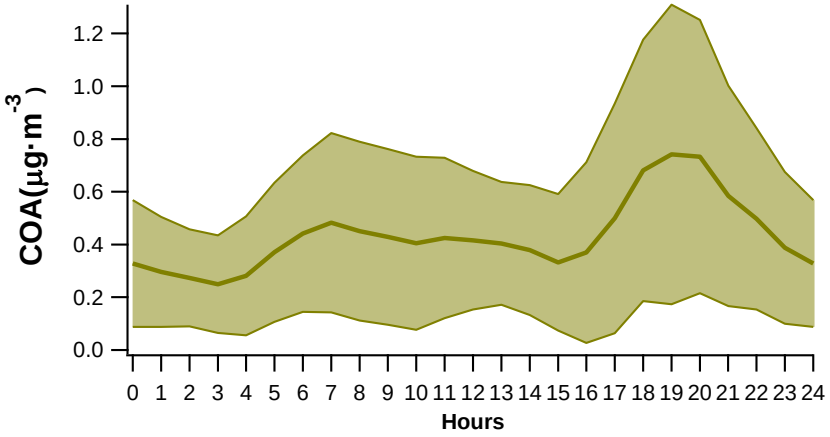
BBOA shows a strong temporal variation, HOA is rather consistent over time, OOA is dominant in all datasets

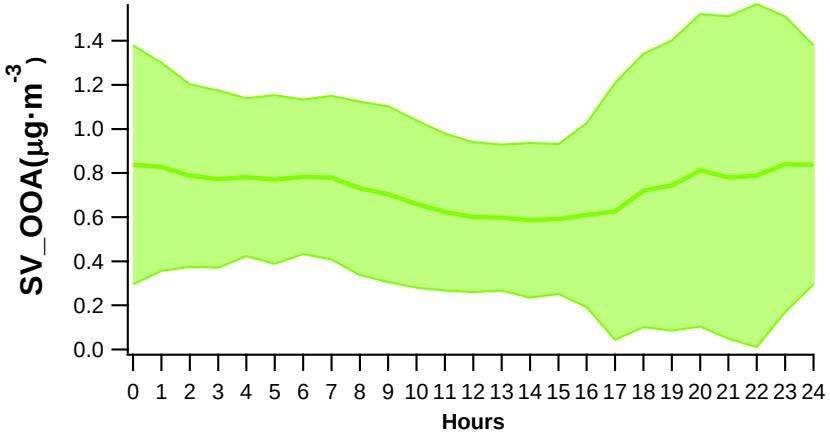
Yearly averaged diurnals





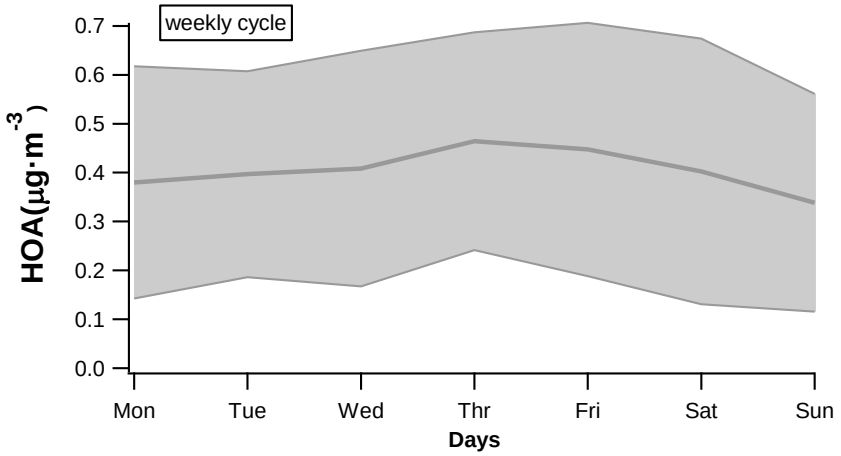


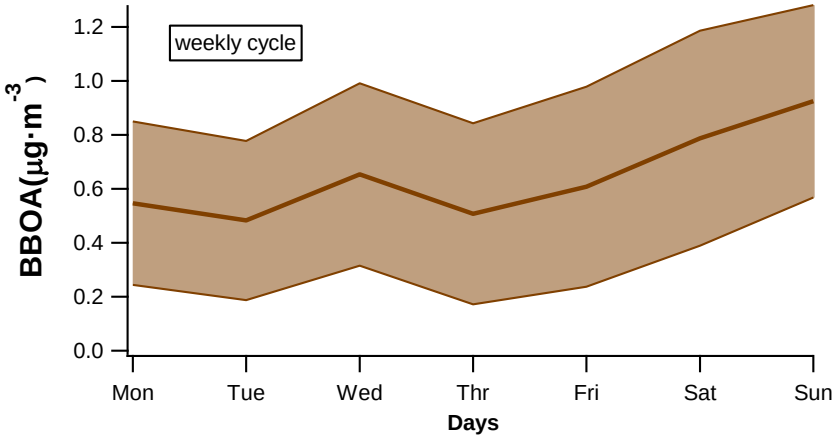


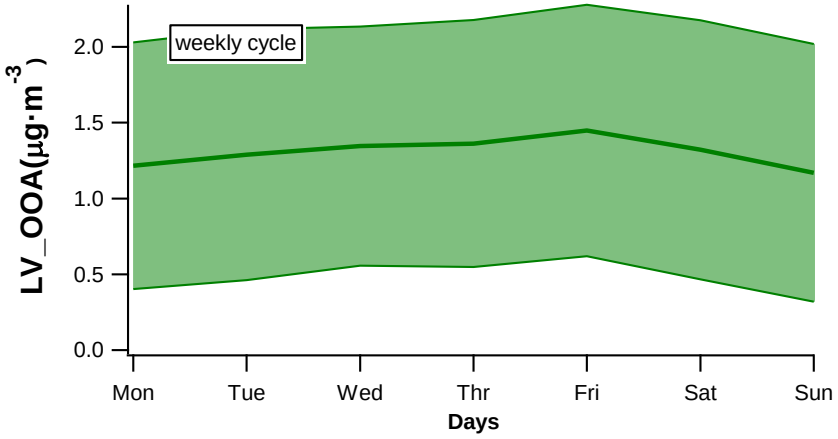


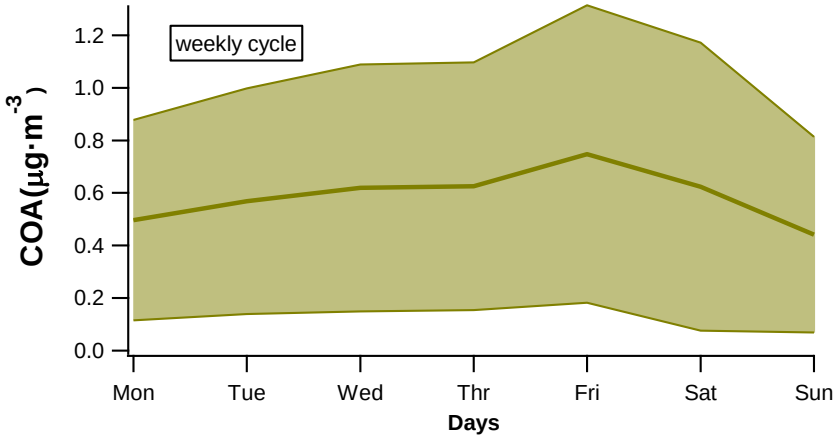
Both POAs show distinct diurnal patterns, OOA factors are rather stable

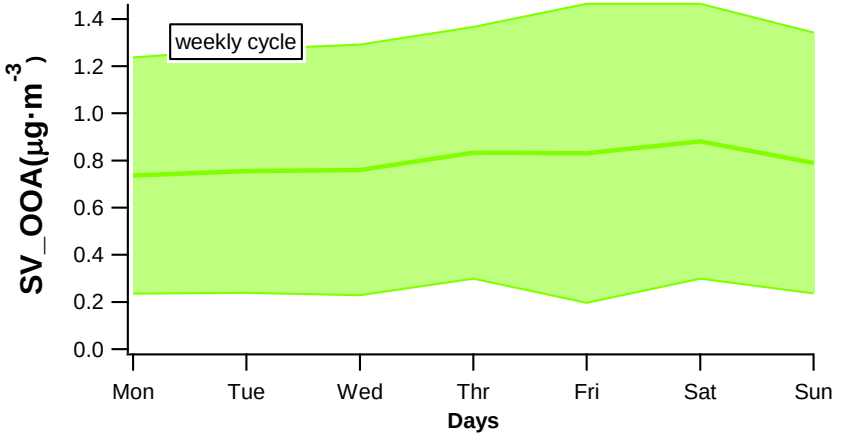
Yearly averaged weekly cycle











HOA seems to be decreased over the weekend, but the opposite for BBOA

Summary and Outlook

- This study provides a standardized protocol to analyze long-term ACSM data using SoFi Pro ;
- 12/24 datasets have preliminary results so far;
- OOA is still the largest contributor in Europe;
- Biomass burning is a considerable source in most of the stations, especially during the cold period

- It could provide a comprehensive overview of the temporal/spatial variabilities of the OA sources in Europe;
- With the overlap from 2016 to 2017, the origin of long-range transport aerosols could be determined;
- With highly time resolved OA sources, it could provide additional constrains for air quality/climate models