

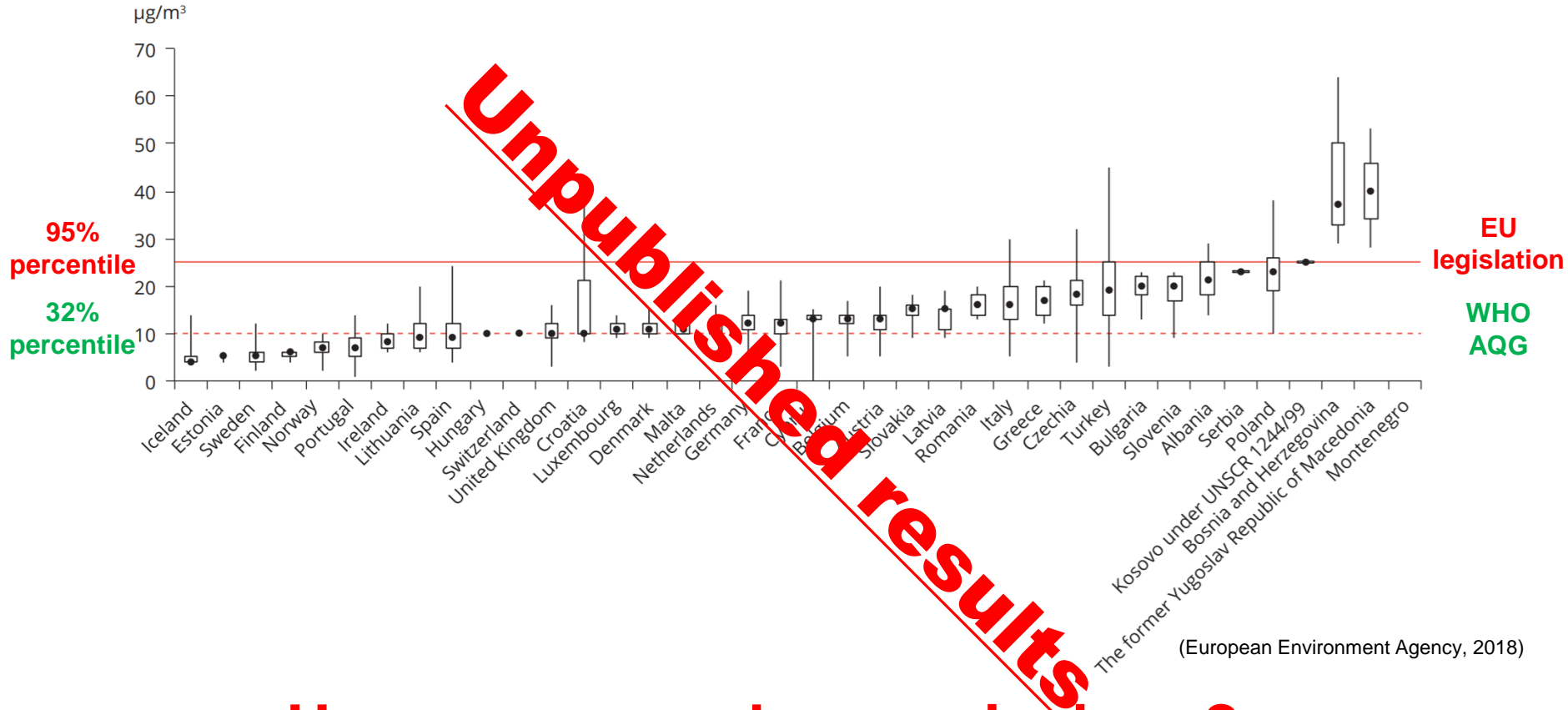
# European Overview for Source Apportionment of Organic Aerosol

Chen et al.

**Unpublished results**

# Motivations

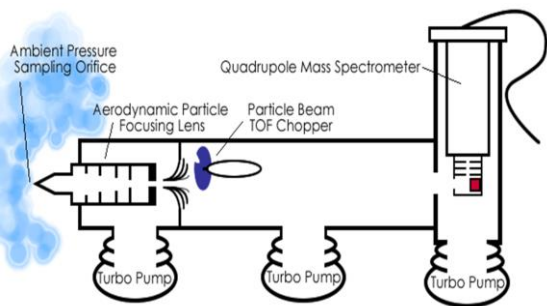
PM<sub>2.5</sub> concentrations in relation to the annual limit value in 2016



## How can we reduce emissions?

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

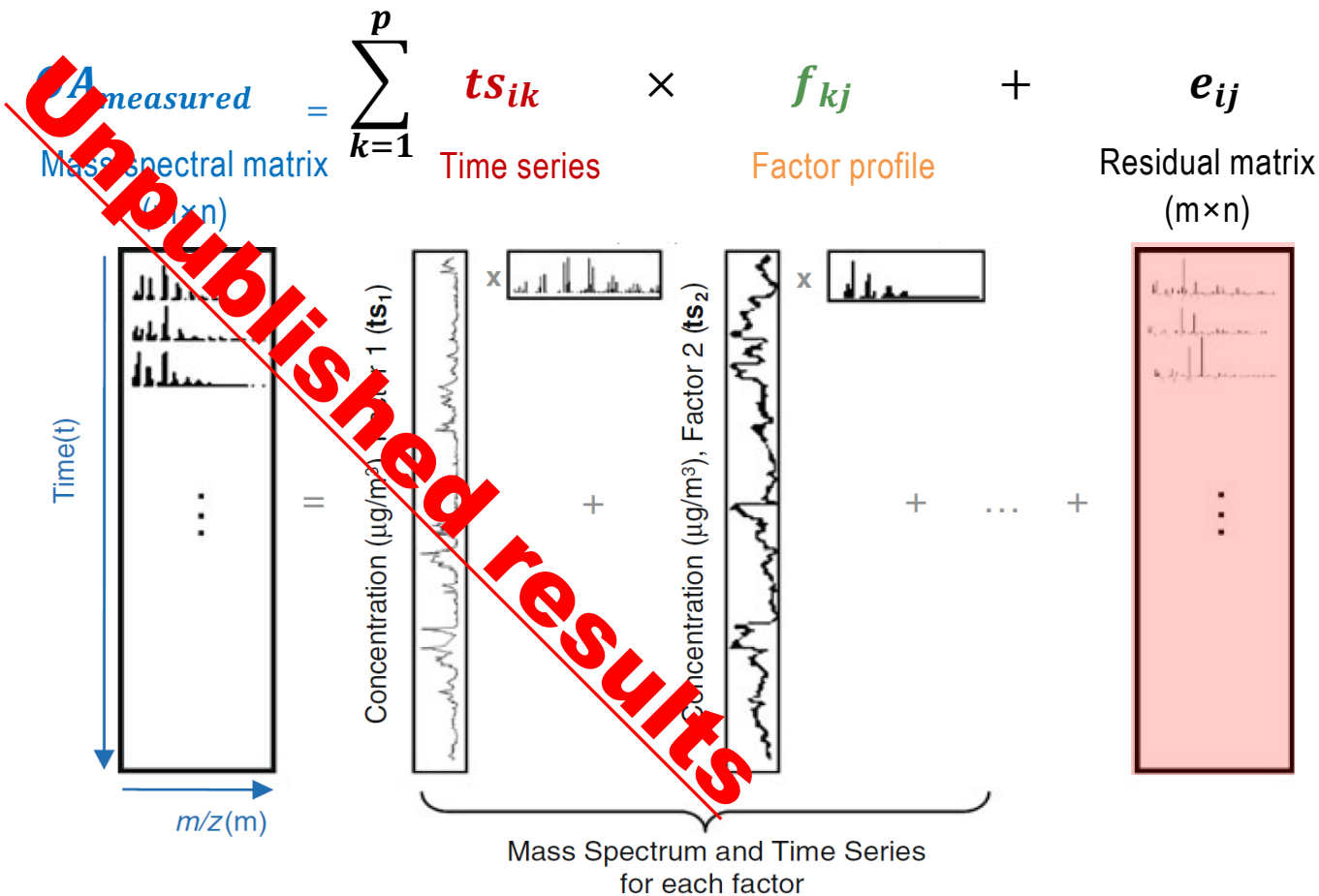
# Positive Matrix Factorization (PMF)



On-line determination of chemical composition:  
The Aerodyne Aerosol Mass Spectrometer (AMS)

$$Q = \sum_{i=1}^m \sum_{j=1}^n \left( \frac{e_{ij}}{\sigma_{ij}} \right)^2$$

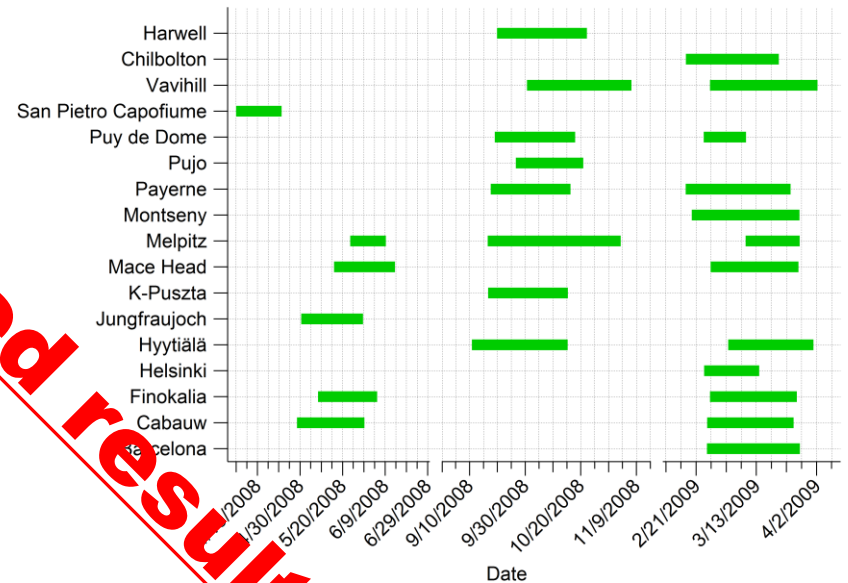
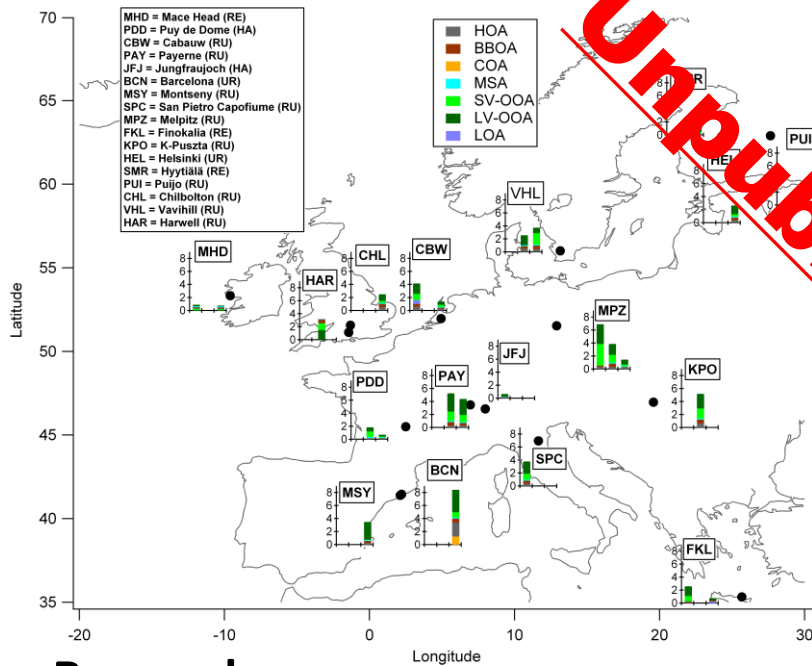
measurement uncertainties



Adapted from Zhang et al., (2011)

# 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)



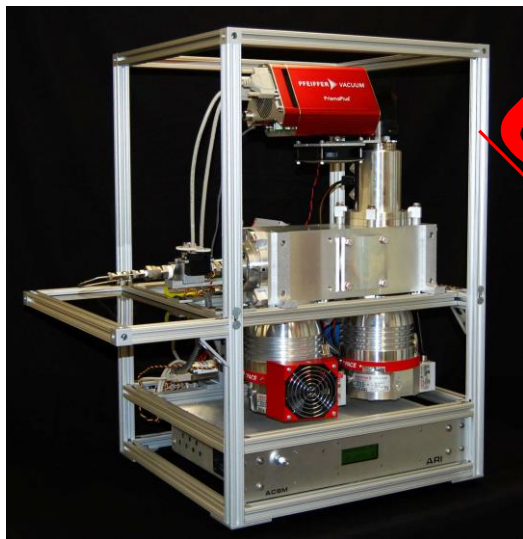
Unpublished results

## Research gaps:

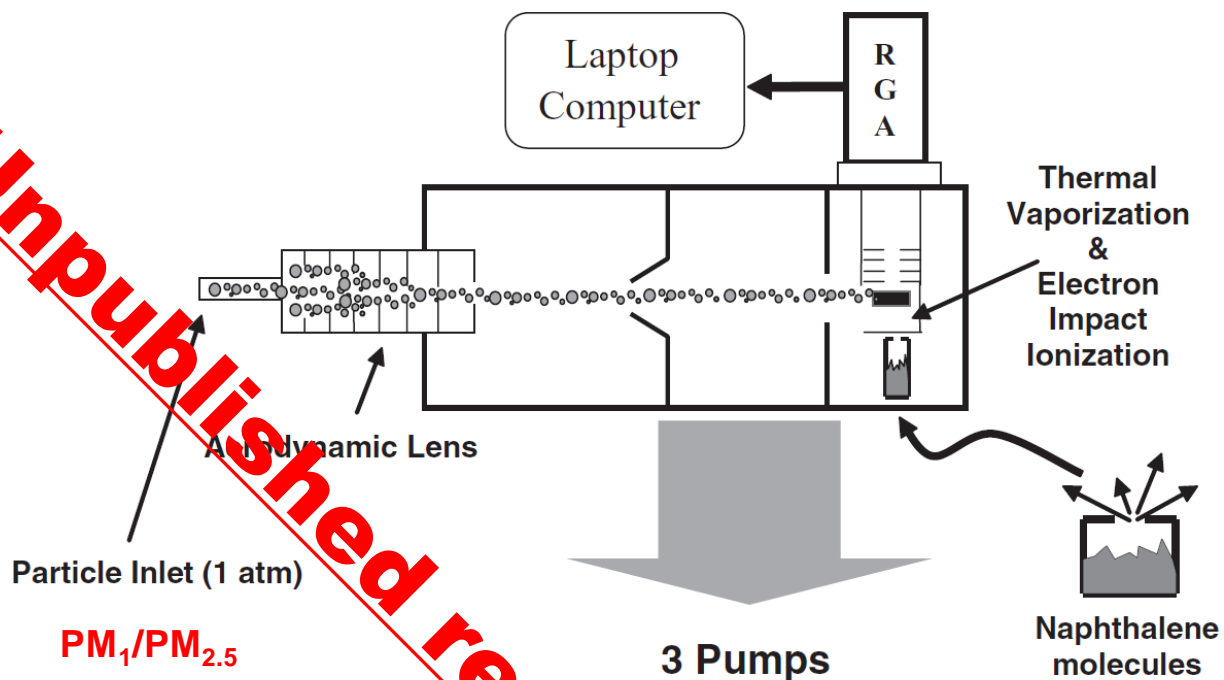
1. PMF suffers from **rotational ambiguity** and requires **subjective judgements**;
2. The conventional PMF does not consider the **evolutions of OA source profiles**;
3. AMS is a labor-intensive and expensive instrument, not desirable for long-term monitoring;
4. **Seasonal variations** of OA sources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

(Crippa, et. al., 2014)

# Aerosol Chemical Speciation Monitor (ACSM)



(Aerodyne, 2014)



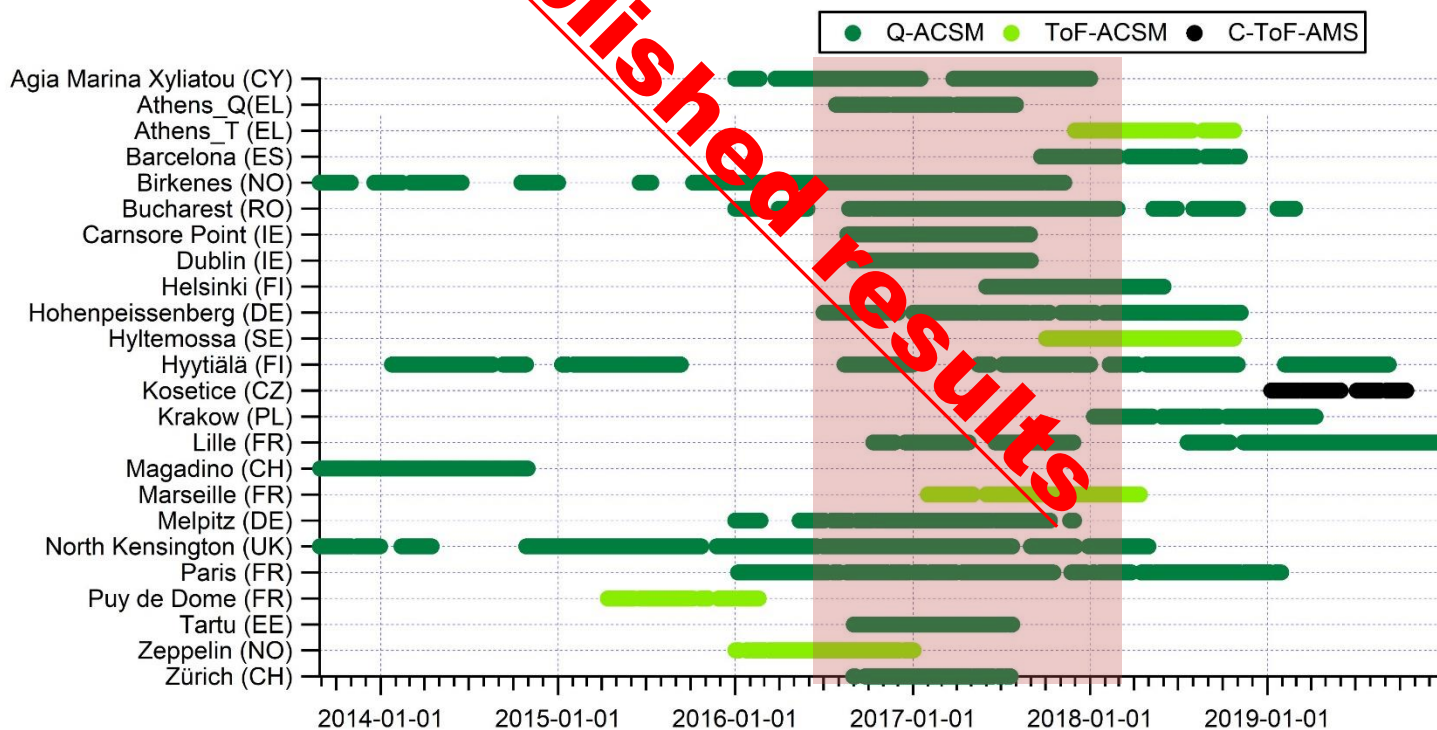
(Ng et al., 2011)

- Report mass concentration of OA, ammonium, sulphate, chloride, and nitrate with a 10-min resolution
- More **stable** for long-term measurement with less maintenance
- Low-cost and suitable for monitoring purpose

But...

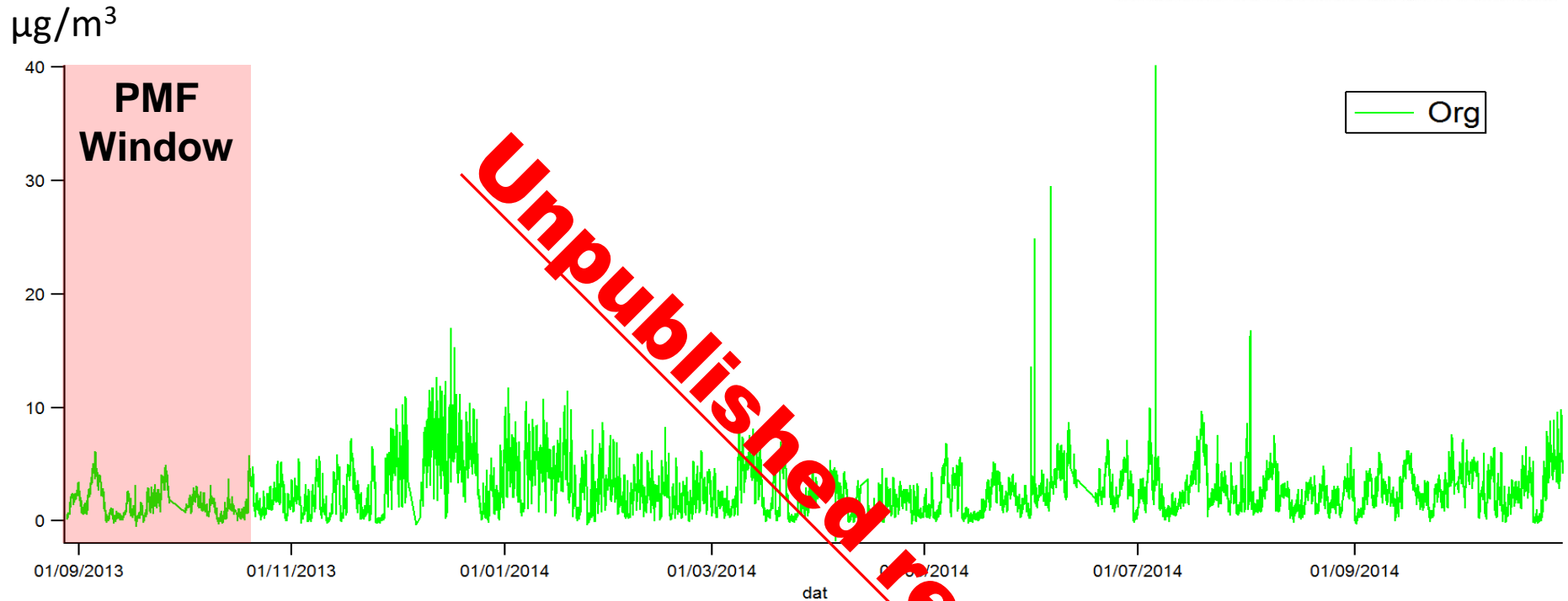
- Only provide a mass spectrum with unit mass resolution (UMR)

# Data Coverage



24 long-term datasets with some overlaps

# What is rolling PMF?



- PMF window can **shift** over the whole PMF input (>1 year) with a step of one day (or user defined) for many (e.g., 50) repeats
- Take the **temporal variations** of OA sources into account
- PMF runs can be assessed by using criteria-based selection, such as correlation with external tracers

# Standard procedures for rolling PMF

Seasonal PMF analysis

- Determine the number of factors

Bootstrap Seasonal solutions

- Test the stability of the seasonal solution;
- Find the thresholds for some criteria

Conducting Rolling PMF

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

Criteria-based Selection

- Define your own criteria to select and reposition "the good" PMF runs
- Select either 4/5 factor solution based on the differentiate criterion



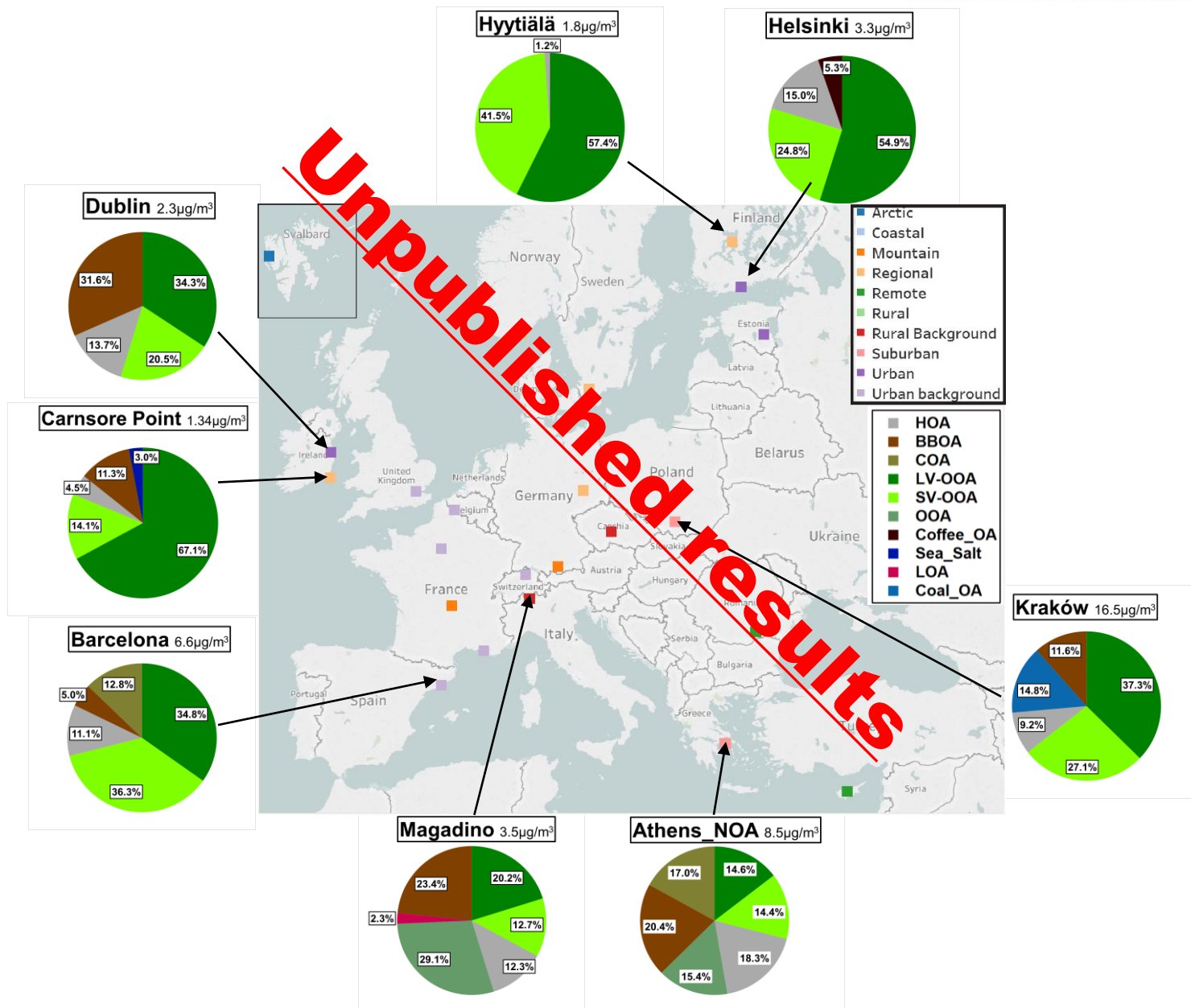
Number of runs in a year:

$$(365-14) \text{ days} \times 50 \text{ Repeats} \times 2 \text{ factors} = 36500$$

Unpublished results



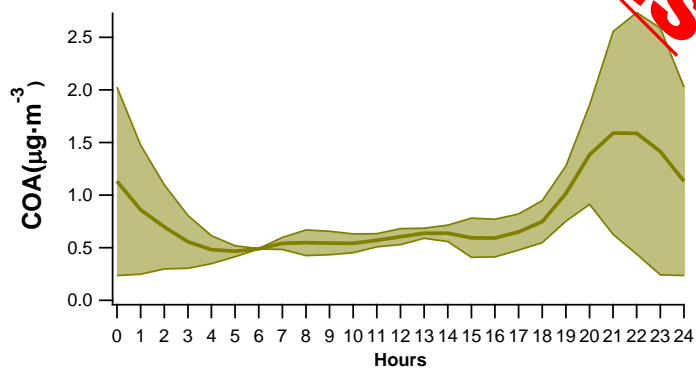
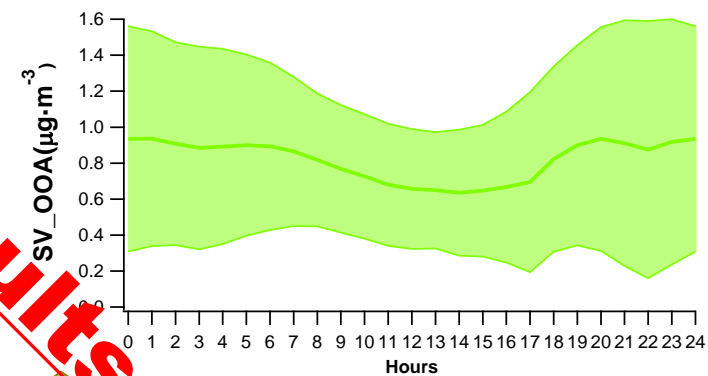
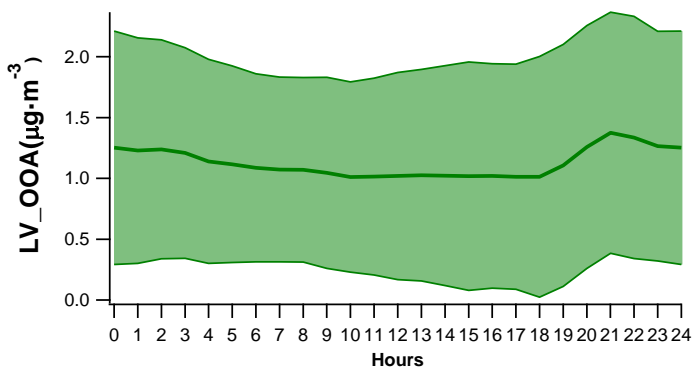
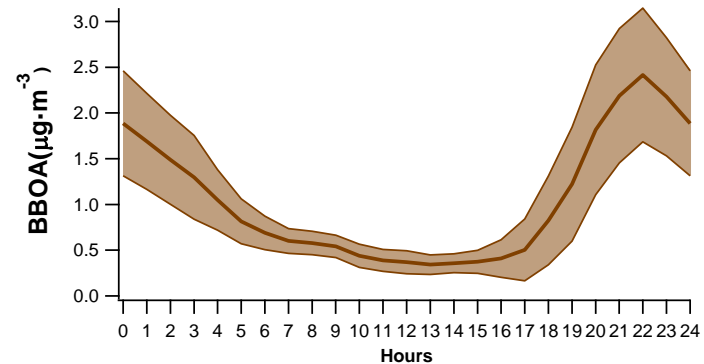
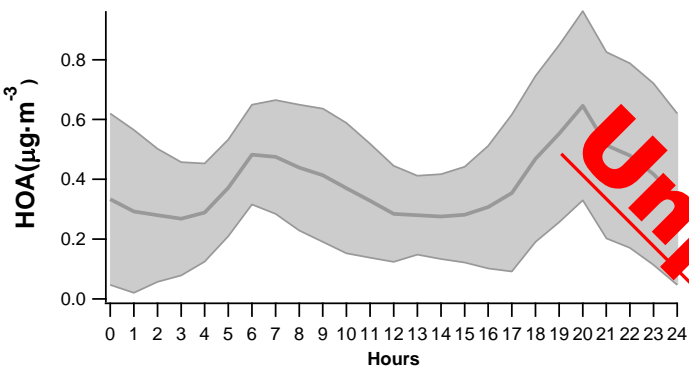
# Overview



# Relative Contribution

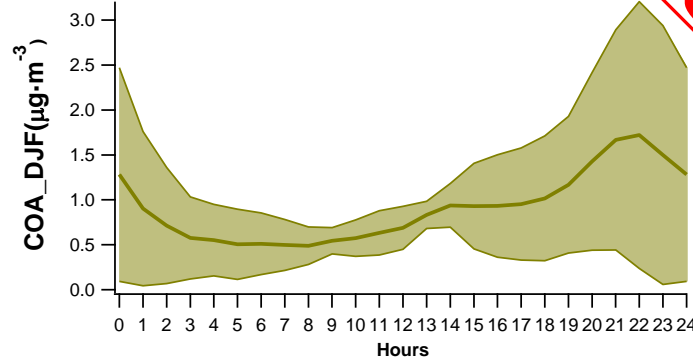
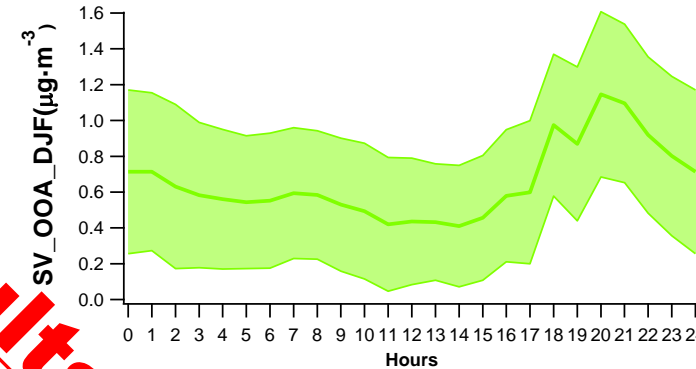
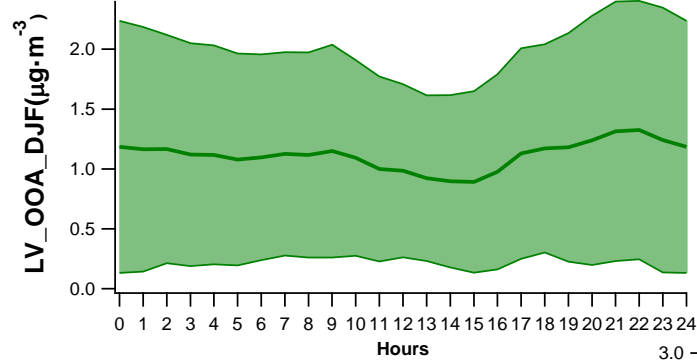
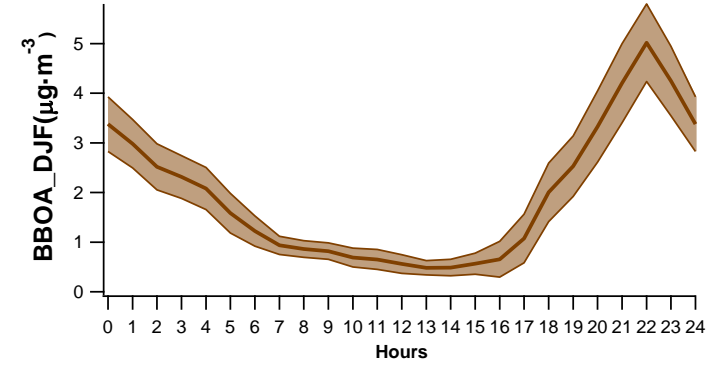
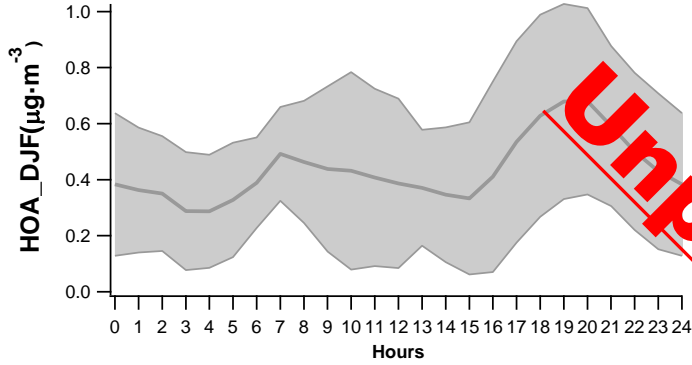


# Yearly averaged diurnals



Barcelona and  
Athens Only

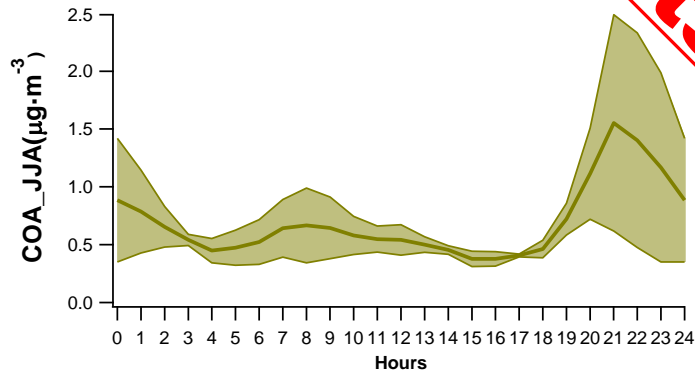
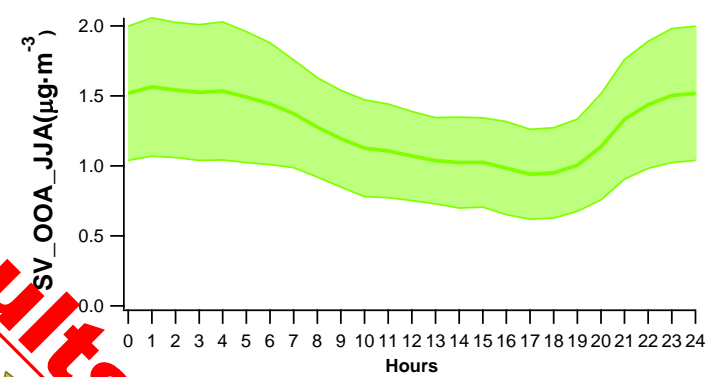
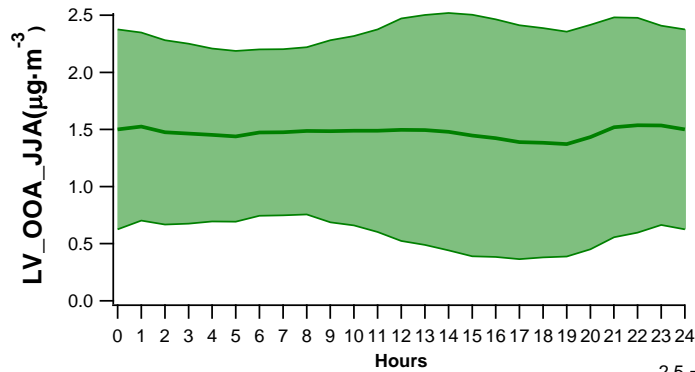
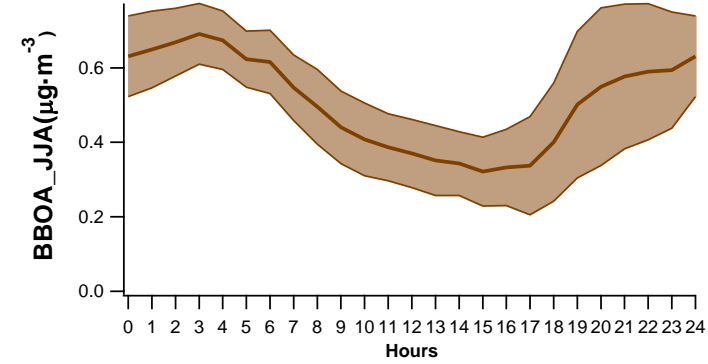
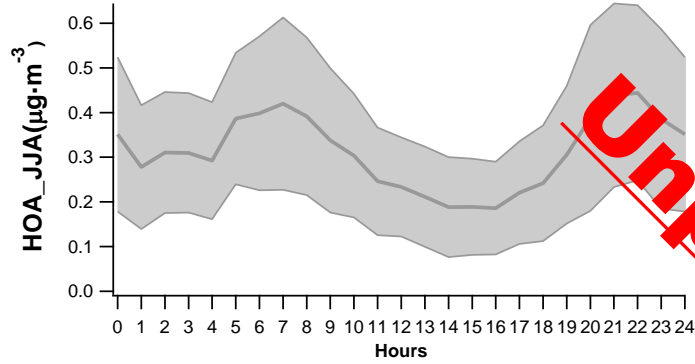
# DJF averaged diurnals



**Unpublished results**

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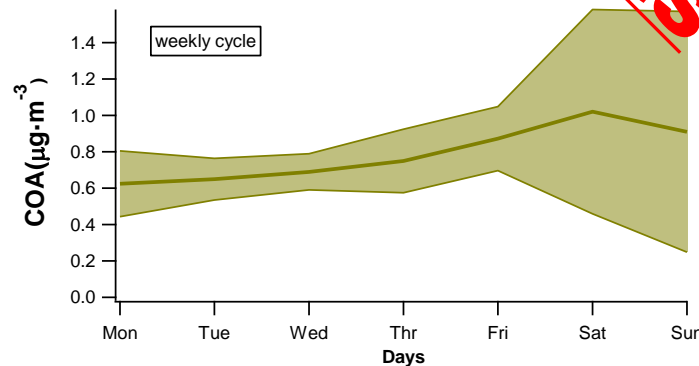
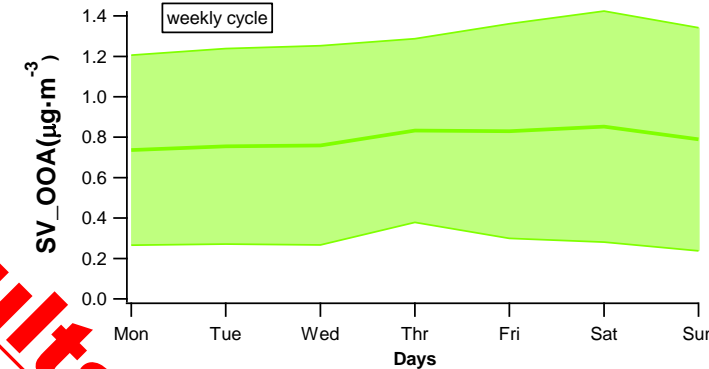
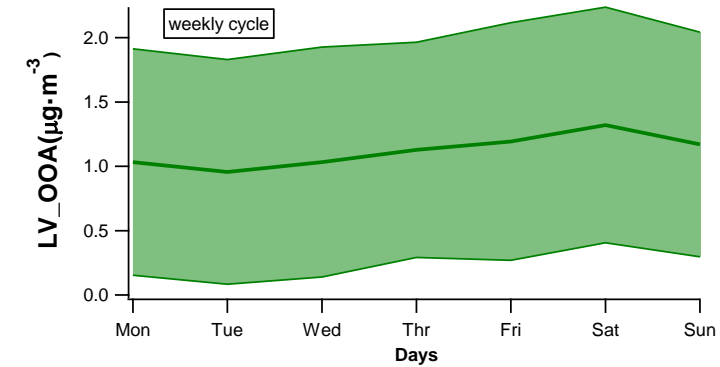
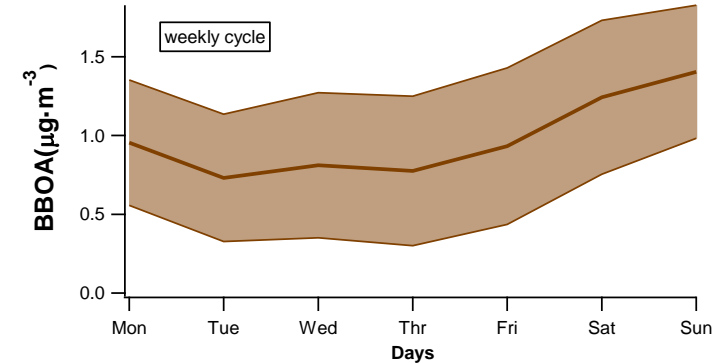
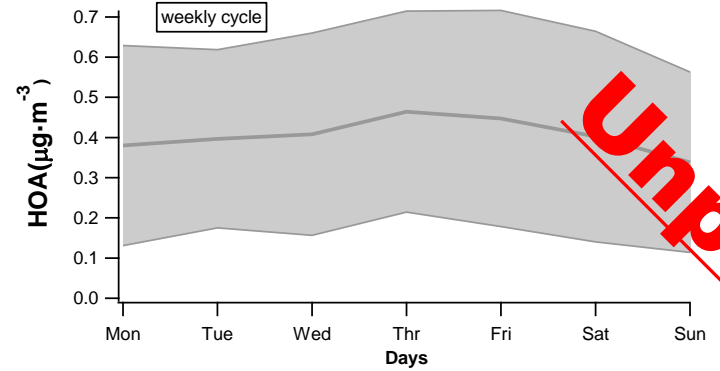
# JJA averaged diurnals



**Unpublished results**

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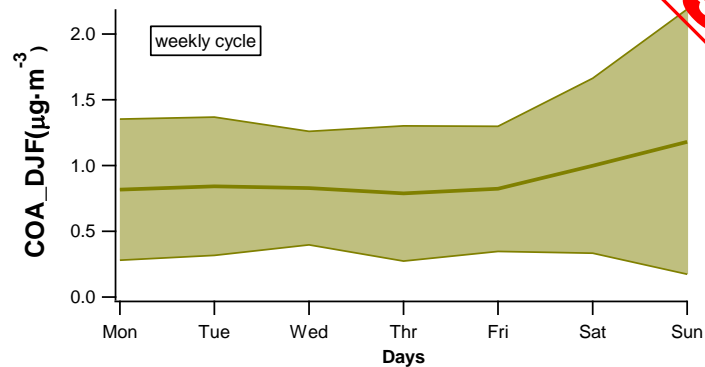
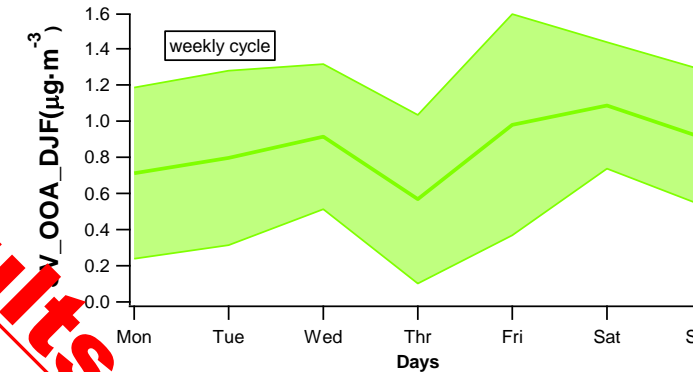
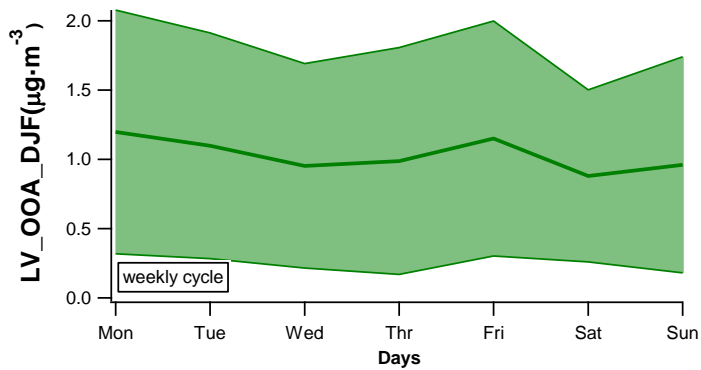
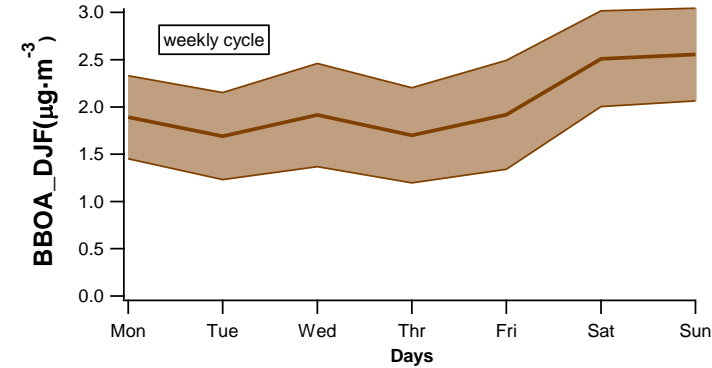
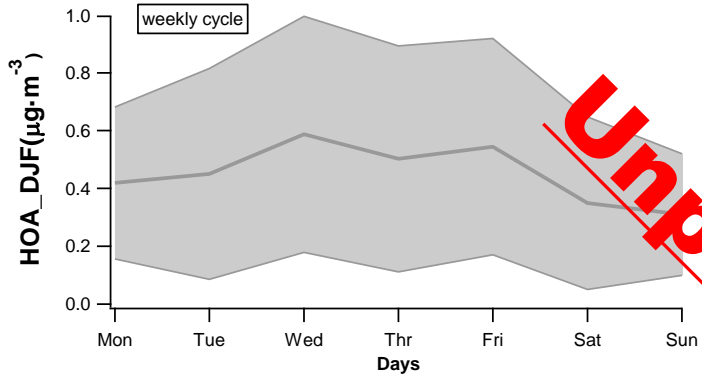
# Yearly averaged weekly cycle



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**Unpublished results**

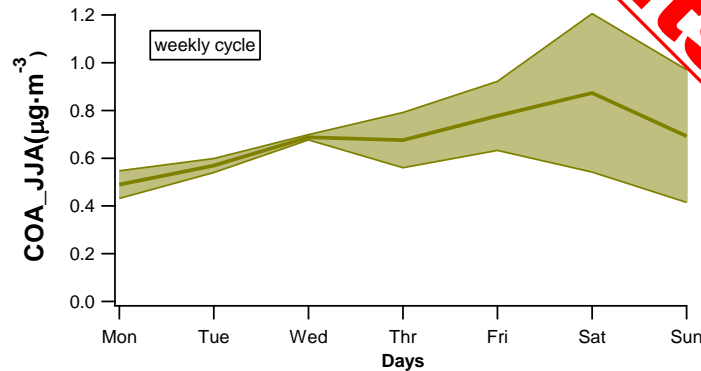
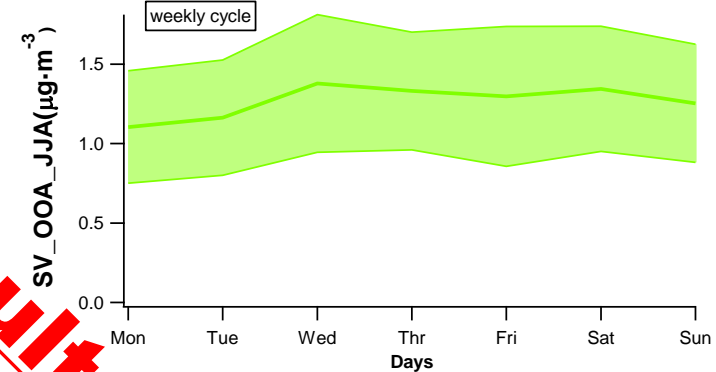
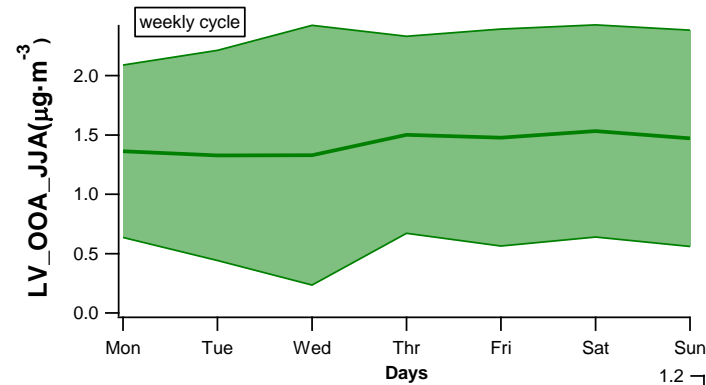
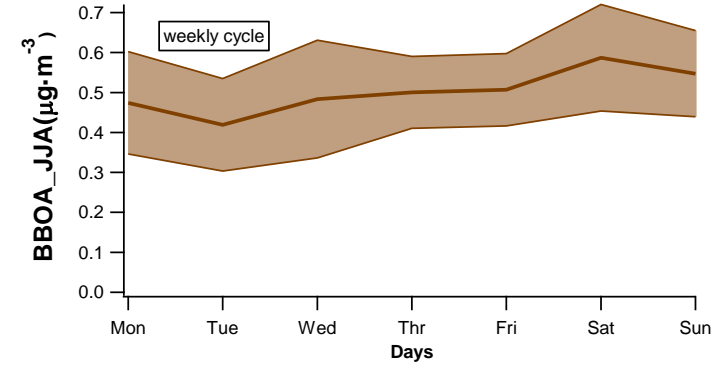
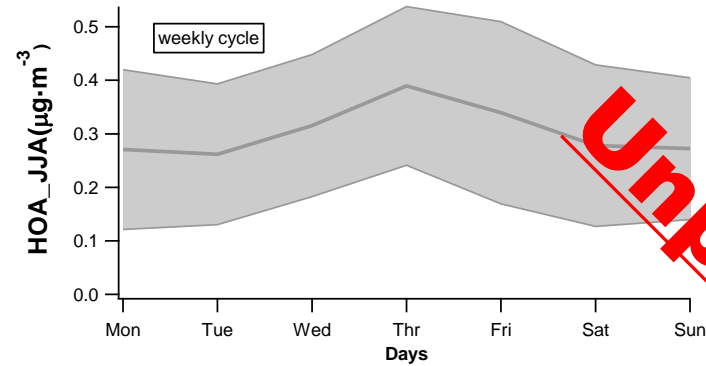
# DJF averaged weekly cycle



**Unpublished results**

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# JJA averaged weekly cycle



**Unpublished results**

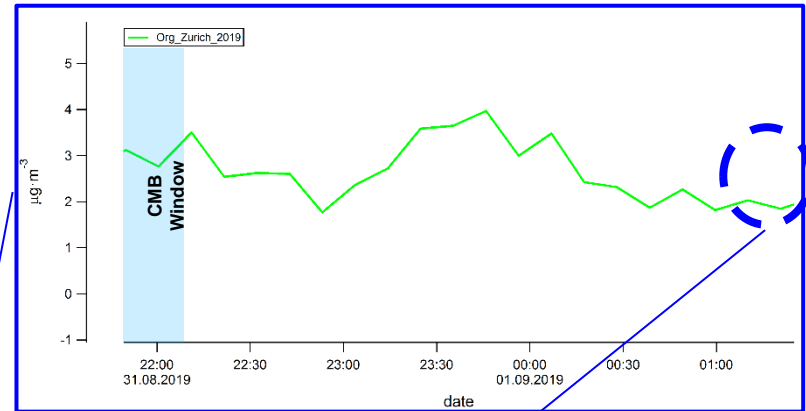
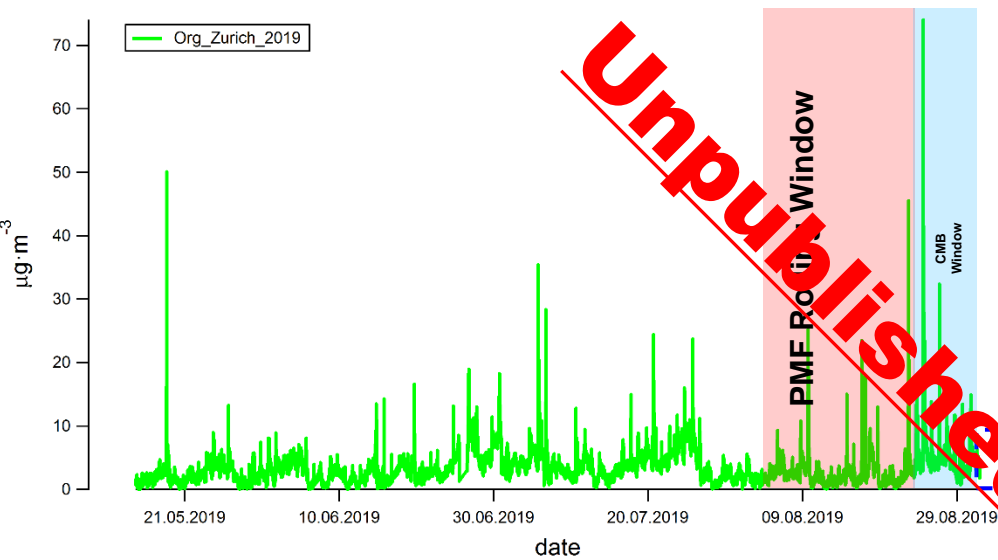
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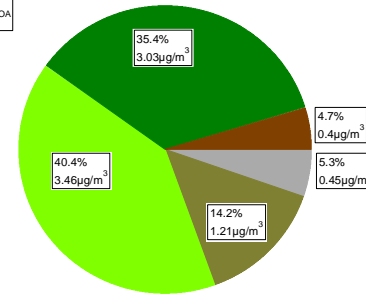
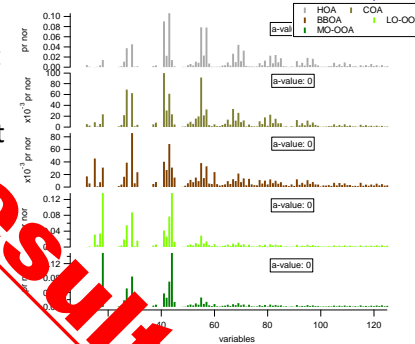
# Summary and Outlook

- 8/24 datasets have preliminary results so far;
- OOA is still the largest contributor over the Europe;
- Biomass burning is a considerable sources in most of the stations, especially during the cold period
  
- This study provides a standardized protocol to analyzing long-term ACSM data using SoFi Fra;
- 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 models;
- On top of the success of this project could potentially leads to the possibilities of **Real-time source apportionment**

# Real-time Source Apportionment



Next  
Data  
point



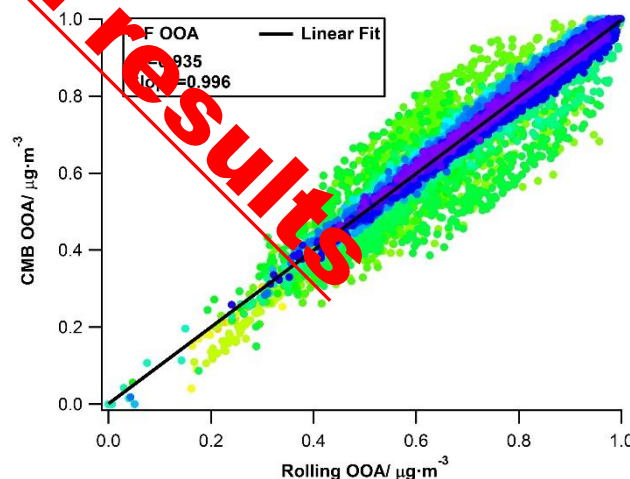
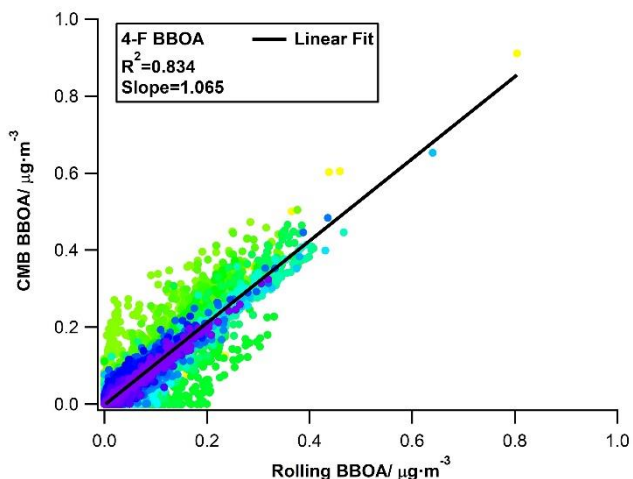
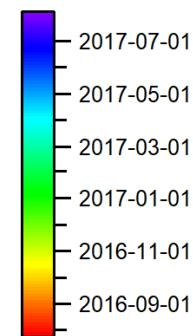
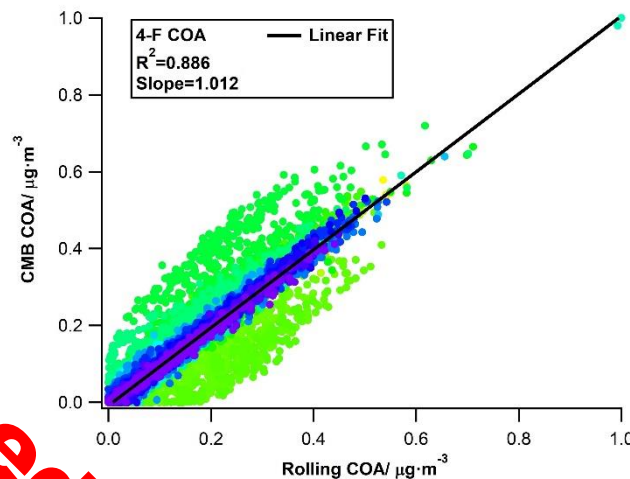
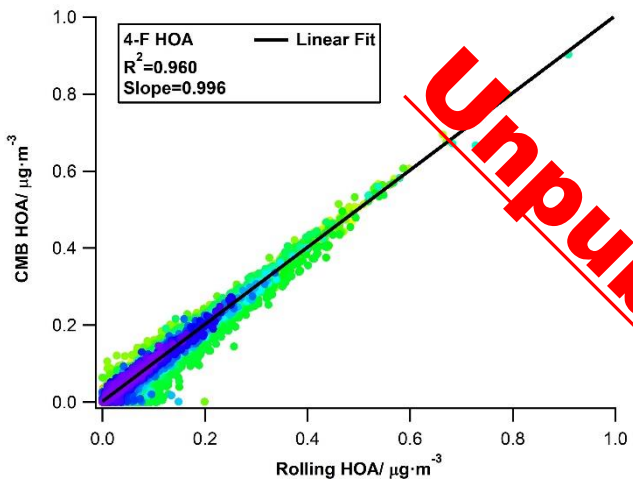
## Assumptions:

Source profiles are consistent within PMF window and the next data point

## Concept:

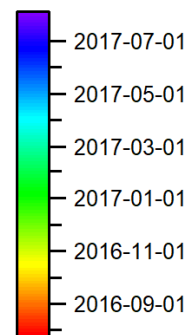
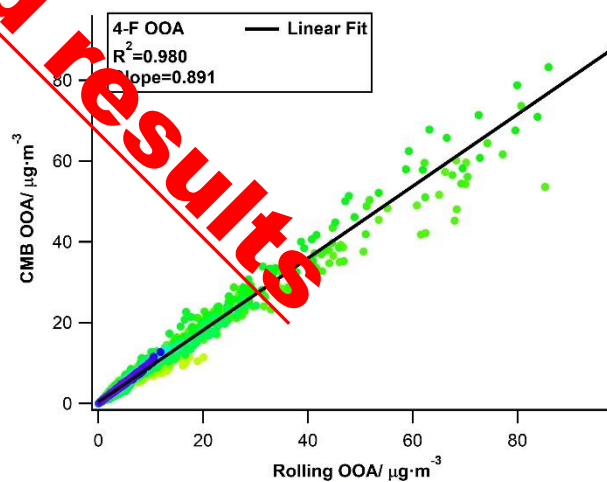
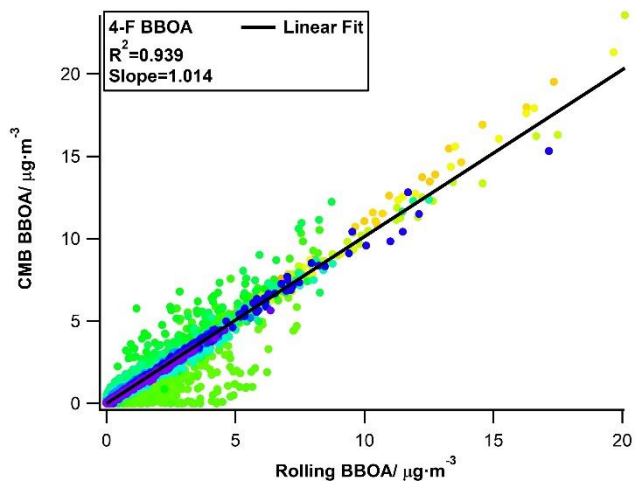
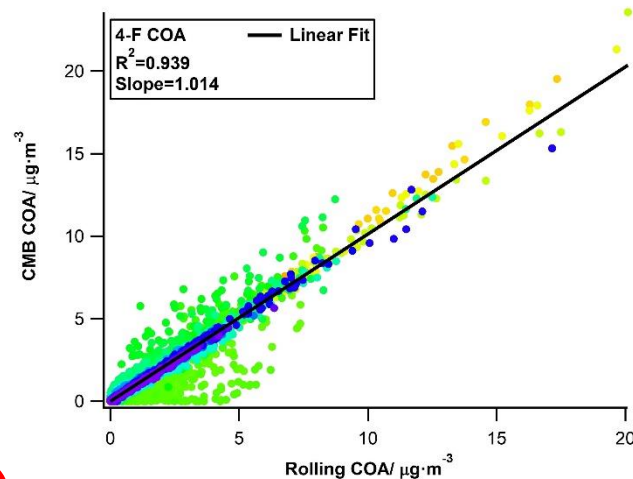
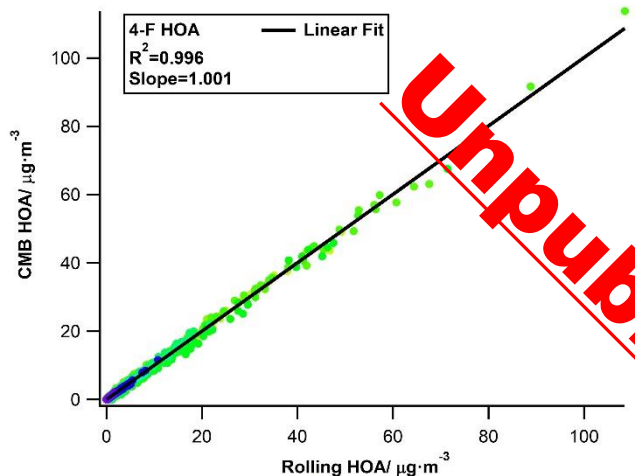
Run the rolling PMF with a small step (e.g., 1 day), using the source profiles from the most-recent solution to run chemical mass balance (CMB) for the next data point to retrieve the real-time OA sources.

# Comparison in relative contributions



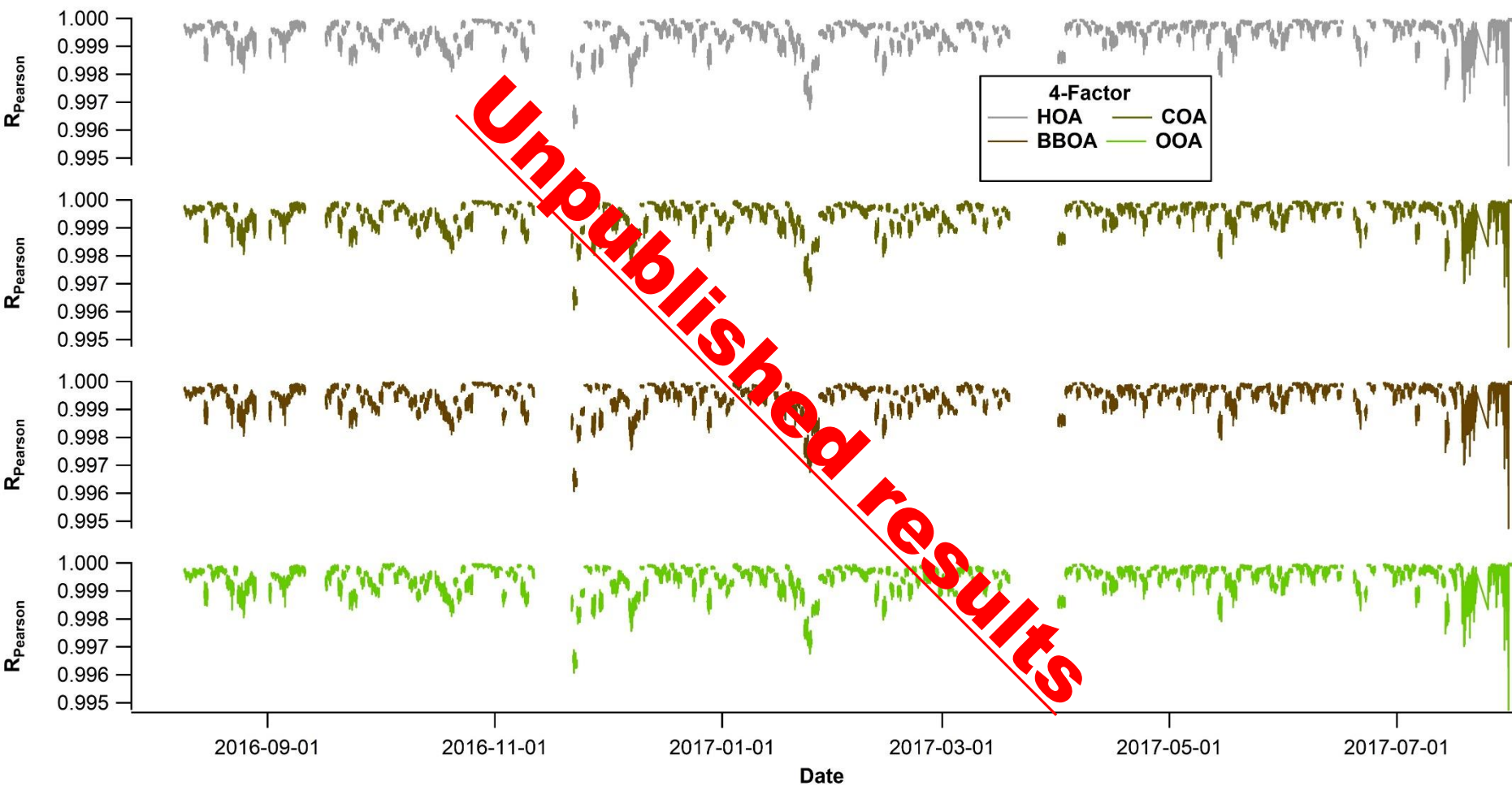
4-factor, 7-day window, 10 repeats/window

# Comparison in mass concentration



**Unpublished results**

# Correlation b/w factor profiles



profiles from the rolling results.

The factor profiles are rather static within 14-day rolling PMF window