



# **ICARUS**

Integrated Climate forcing and Air pollution Reduction in Urban Systems

# Fine Particulate Matter Composition and Sources in 6 European: The ICARUS Project

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## Introduction

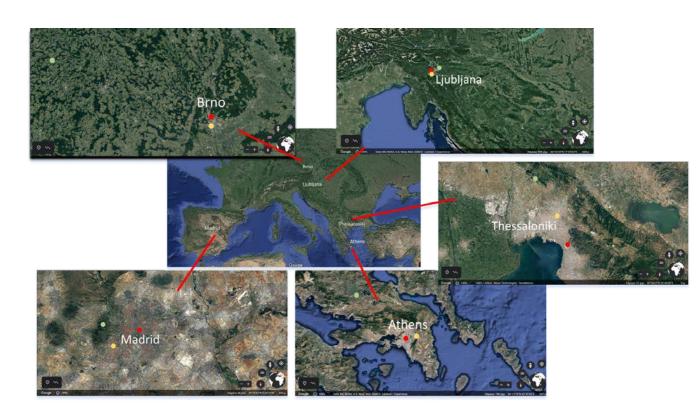
- ✓ PM2.5: air pollution metric widely used to assess air quality (EU targets for reduction in PM2.5 levels and population exposure)
- One of the major challenges for the scientific community is to identify, quantify and characterize, at the appropriate scale, the sources of atmospheric particles in the aspect of proposing effective control strategies to the public authorities.
- Although studies for source apportionment are rapidly spreading globally, revealing both PM local and regional origin, the comparability of results among the different sampling sites is often hampered, leading to *the need for harmonized source apportionment outcomes from multi-city studies*.





## Aim of the study

source apportionment application on PM2.5 data collected in six European cities (Athens, Brno, Ljubljana, Madrid, Stuttgart and Thessaloniki)



- PM2.5 samples collected from three different sites in each city (traffic, urban background and rural) during winter & summer 2017
- chemical analysis for 27 PAHs, 24 trace elements, anions (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>), cations (Mg<sup>2+</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>), elemental and organic carbon (EC, OC)
- **Meteorological data** (meteorological stations at the sampling sites/the nearest station from the national meteorological networks)
- The chemical composition data was introduced in PMFv.5 (Positive Matrix Factorization) model with the scope of identifying the main groups of sources and estimating their contribution to PM2.5 concentrations.





## Methodology

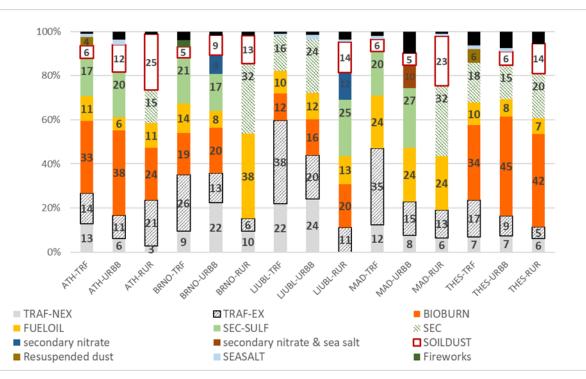
- **Receptor models** aim to re-construct the contribution of emissions from different sources of atmospheric pollutants (e.g. particulate matter PM), based on ambient measurement data (i.e. PM chemical composition) registered at monitoring sites.
- **Positive Matrix Factorization (PMF v.5.0)** introduces a weighting scheme taking into account errors of the data points, which are used as point-by-point weights. Adjustment of the corresponding error estimates also allows it to handle missing and below detection limit data. Moreover, non-negative constraints are implemented in order to obtain more physically meaningful factors (Paatero and Tappert, 1994)
- Data pre-treatment and evaluation of PMF solutions (Bootstrap, Displacement)





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- PMF Results (I)
  - 5-8 factors identified by PMF at each ICARUS site:
  - Main sources: traffic exhaust (TRAF-EX), traffic non-exhaust (TRAF-NEX), fuel oil combustion (FUELOIL) biomass burning (BIOBURN), soil dust source (SOILDUST), sea-salt (SEASALT), secondary aerosol (SEC)
  - un-apportioned fraction of PM2.5: from <1 to 9.88%



% contribution of each source to PM2.5 concentration for every site



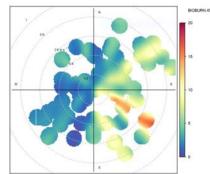
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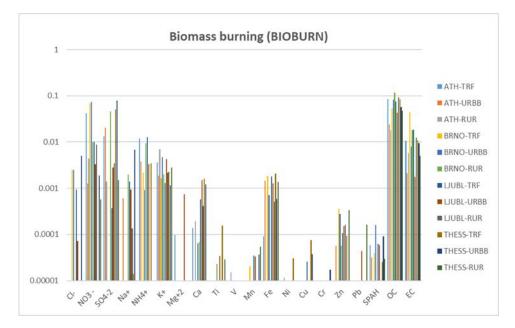
## PMF Results (II)

#### **Biomass burning (BIOBURN)**

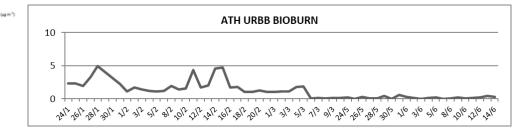
- Major tracers : (OC:30-78%; EC:3-15% of PM2.5 mass), K (7%) or K<sup>+</sup> (2-19%), SO<sub>4</sub><sup>2-</sup> (3-33%), NH<sub>4</sub><sup>+</sup> (<10%) and PAHs (53-74% of measured PAHs)
- prevalence of the source during winter (fireplaces or wood stoves burning), without excluding biomass combustion emissions from agricultural activities (in cases of Athens, Ljubljana and Thessaloniki rural sites).
- BIOBURN contribution to PM2.5: 22% at traffic sites, 30% at urban background sites and 28% at rural sites
- The highest percentage of BB contribution was found in THESS-URBB (45%), while the lowest in LJUBL-TRF (12%)
- Source orientation (R-plots of Factor contribution vs wind speed and velocity) at ATH TRF (city center).



#### Mass profiles (µg/µg) of BIOBURN sources)



#### Time variation of (normalized) BIOBURN source at ATH URBB



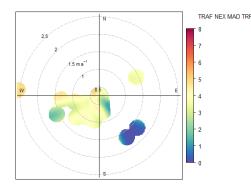


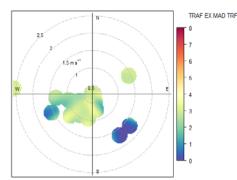


## PMF Results (III)

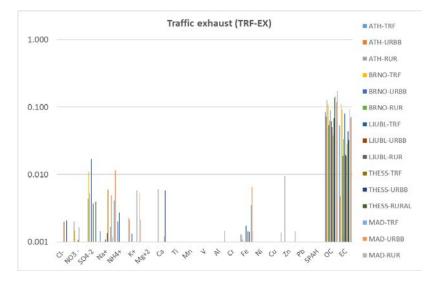
#### Traffic (TRAF-EX & TRAF-NEX)

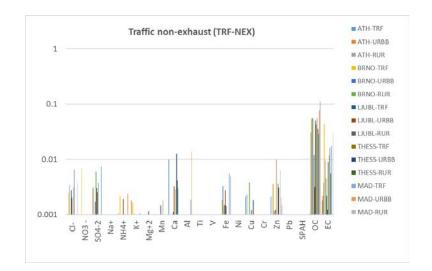
- Two traffic-related sources were distinguished in all sites: the trafficexhaust source (TRAF-EX) and the traffic non-exhaust source (TRAF-NEX)
- TRAF-EX contribution to PM2.5 : 23.3% at traffic sites, 13.3% at urban background sites and 8.8% at rural sites (excluding ATH-RUR site, where it was 21.2% due to frequent heavy vehicles circulation)
- The highest contribution is observed at LJUBL-TRF site (37.8%) while the lowest (5.6%) at BRNO-RUR and THESS-RUR sites
- TRA-NEX contribution: similar levels at traffic (12.6%) and urban background (13.5%) sites while being lower (6.1%) at rural sites.
- TRAF-EX and TRAF-NEX: Similar source orientation (R-plots of Factor contribution vs wind speed and velocity)











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## PMF Results (IV)

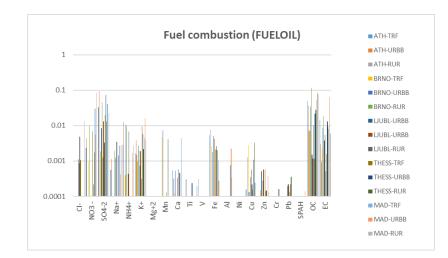
### Fuel oil combustion and industry (FUELOIL)

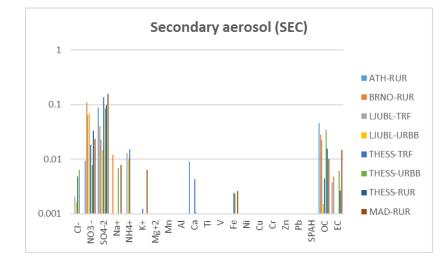
- high shares of OC, EC,  $SO_4^{2-}$ , K or K<sup>+</sup>, as well as Ni and V
- identified in all sites either as residential fuel oil combustion (for heating) or as refinery activities or shipping exhaust emissions or a combination of them
- Not clear predominance of % contribution in a specific type of areas (13.8% at traffic, 11.6% at urban background and 18.7% at rural sites)

#### Secondary aerosol

- identified either as secondary sulfate and organics (SEC-SULF) or as sulfate and nitrate-rich (SEC)
- SEC accounted for the 20-27% of PM2.5 while SEC contributed for the 16-34% of PM2.5
- secondary-particle factor may represent not only the formation of secondary aerosol over relatively long distances, but also a part of traffic-related pollution

#### Mass profiles (µg/µg) of FUELOIL/SEC sources)







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#### Mass profiles (µg/µg) of SOILDUST/SEASALT sources)

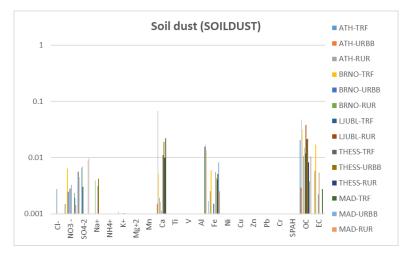
### Soil dust (SOILDUST)

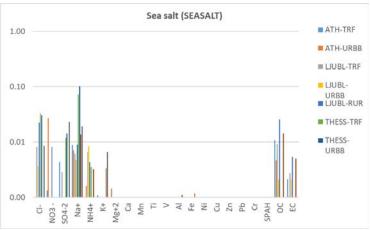
PMF Results (V)

- SOILDUST contribution: 5% 25%
- remarkable difference between traffic/urban background and rural sites is noticed, as SOILDUST average contribution was twofold in the latter (5% and 8% at TRF and URBB sites; 16% at rural sites)

#### Sea salt

- identified in Athens, Ljubljana, Madrid and Thessaloniki
- contribution 1-4%, not presenting a seasonal variation









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### Similarity in sources chemical profiles among the sites

✓ similarity indicators : the PD (Pearson distance) and SID (standardized identity distance):

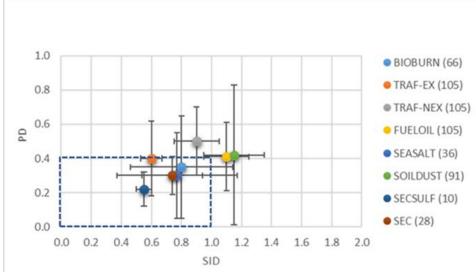
 $PD = 1 - r^2 ,$ 

where  $r^2$  is the Pearson coefficient and

 $SID = \frac{\sqrt{2}}{m} \sum_{j=1}^{m} \frac{|x_j - y_j|}{x_j + y_j},$ 

where x and y is the relative mass ( $\mu g/\mu g$ ) to the PM of two different sources and m the number of common specie in x and y (Weber et al., 2019). Practically, SID results from the comparison of n(n-1)/2 possible and unique pairs of profiles for each source.

- ✓ Based on Pernigotti and Belis, 2018, the acceptable PD and SID values for profile similarity are: P<0.4 and SID<1.</p>
- ✓ fuel oil combustion, traffic non-exhausts and soil dust source profiles are considered as dissimilar
- biomass burning, sea salt and traffic exhaust can be characterized as relatively homogenous
- secondary aerosol sources have been found to be characterized by similar profiles.



Similarity plot (PD-SID space) for all pairs of profiles belonging to the same factor/source category and the acceptable area for profile similarity. The number of pairs of profiles compared for each source category is given in the parenthesis.





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## **Conclusions – Positive Matrix Factorization**

- Main sources: traffic exhaust (TRAF-EX), traffic non-exhaust (TRAF-NEX), fuel oil combustion (FUELOIL) biomass burning (BIOBURN), soil dust source (SOILDUST), sea-salt (SEASALT), secondary aerosol (SEC)
- *Biomass combustion* is a well-distinguished source at all sampling sites. The factor's contribution to PM2.5 indicates the **prevalence** of the source during winter/fireplaces-burning periods, without excluding biomass combustion emissions from agricultural activities (e.g. in cases of Athens, Ljubljana and Thessaloniki rural sites).
- Fuel oil combustion source presents different temporal variation. This is because it is associated with either residential heating or industrial emissions or shipping emissions (Thessaloniki port city) or combination of them
- In the majority of the cases, *traffic* is represented by two different factors: *Traffic-exhausts* and *Traffic non-exhausts*. Contrary to traffic-exhausts, there is no clear trend that traffic non-exhausts has higher share at traffic sites
- A secondary aerosol source was identified either as secondary sulfate only, either as secondary sulfates and nitrate. The
  secondary-particle factor may represent not only the formation of secondary aerosol over relatively long distances, but also a part of
  traffic-related pollution
- Two natural-origin sources were identified: soil dust and sea salt.



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Thank you for your attention