PARIS

Process Attribution of Regional Emissions

GA 101081430, RIA

Completion of year-long source apportionment in 5 European locations

Milestone 28			
Delivery due date Annex I	Month 24		
Actual date of submission	Month 24		
Lead beneficiary: NUIG	Work package: WP7	Nature: Report	Dissemination level: PU
Responsible scientist	Jurgita Ovadnevaite		
Contributors	Vignesh Prabhu, University of Galway, former NUIG, Jurgita Ovadnevaite, University of Galway, former NUIG		
Internal reviewers	-		
Version: 1			

Horizon Europe Cluster 5: Climate, energy and mobility
"This project has received funding from the European Union's Horizon Europe Research and Innovation programme under HORIZON-CL5-2022-D1-02 Grant Agreement No 101081430 - PARIS".

Table of content

1. CHANGES WITH RESPECT TO THE DOA (DESCRIPTION OF THE ACTION) 4

2. DISSEMINATION AND UPTAKE 4

3. SHORT SUMMARY OF RESULTS 4

4. EVIDENCE OF ACCOMPLISHMENT 4

4.1 BACKGROUND OF THE MILESTONE 4
4.2 SCOPE OF THE MILESTONE 5
4.3 CONTENT OF THE MILESTONE 5
4.3.1 PM DATA COLLECTION: 6
4.3.2 PMF RESULTS 7
4.4 CONCLUSION AND POSSIBLE IMPACT 9
4.5 REFERENCES 9

5. HISTORY OF THE DOCUMENT 10

1. Changes with respect to the DoA (Description of the Action)

N/A

2. Dissemination and uptake

The dissemination of findings from M28 involves sharing data via email/SharePoint to internal project partners and external parties interested in the data. A possibility of data archiving in the EBAS database is explored to ensure it is publicly accessible. The public availability of the dataset through EBAS promotes transparency, collaboration, and informed decision-making in environmental protection. While an upload of the source apportionment data to the EBAS database was not originally planned, it was deemed important to do so in order to ensure open access and promote transparency, collaboration, and informed decision-making. Since currently there are no standardized submission tools or procedures available for the source apportionment data upload, the PARIS WP7 team has engaged EBAS on this. The working group with members from EBAS, PARIS and another EU project RI-URBANS was created to develop these procedures and data will be submitted after the tools are finalized.

To promote the data, findings will be shared with relevant stakeholders through scientific publications and conferences to enhance understanding and foster further research. This dissemination strategy aims to ensure the results are effectively utilized to improve the robustness and reliability of bottom-up emission inventories for Europe.

3. Short summary of results

The ground-based PM_1 from 15 different sites located in Europe was compared with the corresponding PM_1 derived from Copernicus Atmosphere Monitoring Service (CAMS) model data. The results revealed a high bias at sites located in urban and suburban areas. Hence, four sites with urban/sub-urban land use characteristics and one site with rural background were selected for the source apportionment (SA) study. With this approach, different pollution sources were identified and quantified.

For source apportionment, the OA concentration and error matrix from each of these sites were analysed using the Positive Matrix Factorization (PMF) receptor model. The results revealed seven distinct OA factors such as primary-OA from peat, wood, and coal burning; hydrocarbon-like OA, cooking-OA, and two types of oxygenated organic aerosols.

This detailed SA provided crucial insights into the relative contributions of different OA sources. These findings will help to highlight key discrepancies with emission inventory data and offer a pathway to address data gaps.

4. Evidence of accomplishment

4.1 Background of the milestone

Particulate matter, including organic matter (OM) aerosols and black carbon (BC), plays a critical role in influencing climate and air quality (Chen et al., 2022; Lin et al., 2020). OM and BC are significant components of PM_1 (particulate matter mass of particles with aerodynamic diameter ≤1 µm) which act as short-lived climate forcers with complex and sometimes opposing effects on the climate (AMAP, 2015; CCAC, 2019; UNEP, 2017). The diversity of aerosol sources and limited atmospheric data pose challenges in aligning bottom-up emission inventories with atmospheric observations. Addressing these gaps requires advancements in data analysis, source attribution methods, and atmospheric modelling.

Under work package seven (WP-7) of the PARIS (Process Attribution of Regional Emissions) project, non-GHG climate forcers such as OM and BC are the point of focus. Thereby, five European sites exhibiting significant discrepancies between ground-based PM_1 measurements and model-derived data were identified. Detailed source apportionment study at these locations revealed both major and minor sources of PM_1, thereby helping in enhancing the integration of bottom-up inventories with atmospheric data.

The project further aims to verify emission inventory consistency with the observed OM concentrations across major source sectors and refine methods for sector-specific BC emission estimation. This activity, to be described in upcoming deliverables, involves developing a European atmospheric modelling and inversion framework to quantify BC fluxes at national and sectoral levels, integrating observations from networks like ACTRIS (Aerosol, Clouds and Trace gases Research Infra Structure) and AC3 (Atmospheric Composition and Climate Change).

4.2 Scope of the milestone

Data collection and quality assurance: The OM data in PM_1 measured using a Quadrupole Aerosol Chemical Speciation Monitor (Q-ACSM) was collected from the respective laboratory supervisor through email. The quality assurance and quality control procedures were followed and documented. For more detail see Chapter 4.3.1.

Data preparation for PMF: The exported OA mass concentration and error (uncertainty) matrix was used as inputs for the PMF model.

Application of rolling PMF: An advanced rolling PMF method combined with Source Finder Pro (SoFi Pro) tool was employed on OA datasets obtained from the sites. The rolling PMF is a useful tool especially when the pollution sources have similar temporal distribution.

Development of novel approach: To identify the minor sources of OA, the hidden peaks in the OA data were explored. The bootstrap reference profiles were used for constraining the primary-OA factors.

Data documentation: The identified OM factors were documented with both their mass profiles and temporal variations. While the analysed data will be made available after research article publication, the raw data can be requested through email.

4.3 Content of the milestone

Data description: The dataset comprises the source apportionment results of OM in PM_1 collected from five cities for the year 2022-23. The OM dataset was measured by sophisticated Q-ACSM instrument (Ng et al., 2011). Further, the source apportionment of OM was performed using rolling-PMF method and several OM sources were successfully identified.

Metadata: The dataset is accompanied by comprehensive metadata, detailing information such as the measurement units, station names, land use characteristics, instrument settings, calibration parameters, contact information of the submitters, the name and descriptions of identified OM factors, and more.

Findability: The analysis dataset will be made available through report and research article publication.

Accessibility: The raw data will be openly accessible via. email request.

Interoperability: The analysis was performed through various methodologies besides following the guidelines mentioned by Chen et al. (2022).

Reusability: The analysis results include detailed mass profiles and temporal variations of the identified OM factors. This comprehensive documentation ensures that the data can be reusable for various research purposes and analyses.

4.3.1 PM data collection:

For ground-based PM, the data from reference grade monitors located aerially close to the ACSM station were collected (https://openaq.org/). Information of the ACSM stations across Europe was obtained from PSI (https://www.psi.ch/en/acsm-stations/acsm-and-emep-stations, accessed on 5th JULY 2024) and Chen et al. (2022).

For model-based PM, the CAMS data (spatial resolution: 0.75° × 0.75°) was obtained from the ECMWF (https://www.ecmwf.int/, accessed on 5th JULY 2024). Panoply software was employed for processing the CAMS data, and python program was used to extract the CAMS-PM_1 data corresponding to the grid cell of ground-based PM monitoring site.

The CAMS-PM_1 data was compared with the ground-based PM_1 data and the result for a few sites is presented in Table 1. In this analysis, only statistical models were utilized; scatter plots and time series plots were also generated. Key statistical metrics, including the coefficient of determination (R^2), root mean square error (RMSE), normalized root mean square error (NRMSE), and Pearson correlation coefficient (r), were calculated for each site to evaluate the model's performance. No outliers were excluded to ensure a comprehensive evaluation.

Table 1: Example of the model result comparison with the measurement data for the selected European sites.

ACSM station name (city)	Country	Land use characteristics	Correlation with Model data (Pearson's r)
Kosetice	Czech Republic	Background	0.41
Lyon	France	Urban - Traffic	0.15
Puy de Dôme	France	Suburban	0.28
Revin	France	Background	0.50
Lille	France	Background	0.59
Melpitz	Germany	Background	0.47
Bologna	Italy	Urban- Traffic	0.07
Monte Cimone	Italy	Urban	0.30
Padova	Italy	Background	0.42
Barcelona	Spain	Urban - Traffic	0.24
Montseny	Spain	Traffic	0.15
Carnsore Point	Ireland	Rural - background	0.67

This analysis revealed that the land use characteristics play an important role in determining the strength of correlation between ground and model-based PM_1. Specifically, the average r value (0.5) for sites under background category was considerably higher than the average r value (0.2) for sites in other categories. This poor association underscores the importance of investigating various types of PM_1 sources over different land use characteristics. Hence, four sites (Athens, Bologna, Bucharest and Sirta) with urban/sub-urban land use characteristics and one site (Carnsore point) with rural background were selected for SA study. Detailed explanation for the selection of these sites for the SA study is provided below:

- Athens in Greece is one of the sites included in the SA study. The city has complex air pollution mix from natural as well as anthropogenic sources such as vehicular emissions, thereby contributing significantly to the elevation of PM mass concentration. Hence, including this site can provide insights of lesser-known sources.
- The Po valley in Italy has a typical geographical and meteorological conditions that play a major role in influencing the pollution levels over the region. Bologna is one of the important cities in the region and it is ideal for a detailed SA study due to its diverse pollution sources such as traffic emissions, industrial activities, and residential solid fuel burning (Daellenbach et al., 2023).

- Bucharest in Romania has high levels of PM and NO_2 that often exceeds the recommended WHO limits. Additionally, the city has spatial and temporal variability in pollutant concentrations making it suitable for a detailed SA study (Ilie et al., 2023). It is one of the most polluted regions in Europe with very specific source characteristics.
- Carnsore Point in Ireland serves as an ideal regional background site for air pollution research due to its relatively clean air and distance from major urban centres with exposure to frequent long range transport episodes and providing insights into processed PM (Ovadnevaite et al., 2021). Its settings as a regional background site make it valuable for comparison with more polluted urban environments.
- Sirta in France provides a representative suburban environment, essential for understanding the impact of various pollution sources (Freney et al., 2019; Via et al., 2022).

4.3.2 PMF results

The result of source apportionment for the five sites revealed seven OA factors/components (Fig. 1). Sources such as peat, coal, wood, biomass burning OS (BBOA), hydrocarbon-like OA (HOA), cooking-OA (COA), low-oxygenated OA (LO-OOA) and more-oxygenated OA (MO-OOA) were identified.

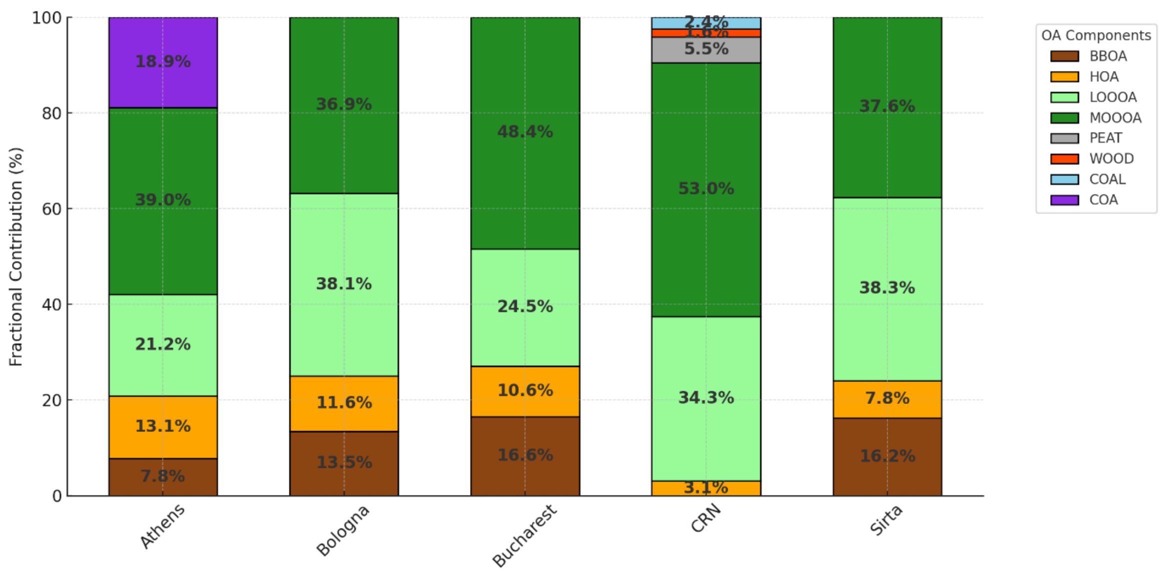

Fig. 1: Fractional contribution of OA sources in the study sites

Fig. 1 presents the fractional contribution of OA components across the five study locations—Athens, Bologna, Bucharest, Carnsore point (CRN), and Sirta. Hydrocarbon-like OA, a primary pollutant associated with vehicular emission can be observed in all sites while prominently in urban sites, contributing 10 to 13% of OA.

The secondary OA such as LO-OOA and MO-OOA dominate the OA composition in all the sites, indicating the prevalence of regional secondary aerosol formation. This process is likely driven by photochemical aging and long-range transport (Stavroulas et al., 2019).

The presence of substantial BBOA component at Sirta and Bucharest underscores the influence of residential wood burning, a major source during colder months. Similarly, the presence of peat, coal and wood burning at CRN suggests seasonal residential heating patterns and possibly episodic biomass burning events in rural areas. Overall, this analysis highlights the presence of both primary and secondary aerosols.

Fig.2 illustrates the timeseries of OA sources across five sites. The data highlights the seasonal fluctuations in concentrations of different OA components with MOOOA consistently contributing a significant source across all locations.

At CRN, the peat, HOA, and BBOA exhibit more variability, where HOA and BBOA contribute to the OA load during specific periods, such as summer when agriculture fires events occur. While the peat and coal emissions occur more during winter period. At BOLOGNA, BBOA was observed during summer season which can be attributed to agricultural burning events. The fire spot data for the region collected from (https://firms.modaps.eosdis.nasa.gov/download/list.php, accessed on 12th DEC 2024) revealed ~73% of fire events during summer season (APR-SEP), which substantiates the BBOA contribution.

The distinct seasonal signatures observed in the plot suggest that each location experiences a unique combination of anthropogenic and natural aerosol sources. These findings emphasize the importance of local sources and seasonal patterns in influencing OA concentrations and would be highly valuable for evaluating emission inventories with the regional model.

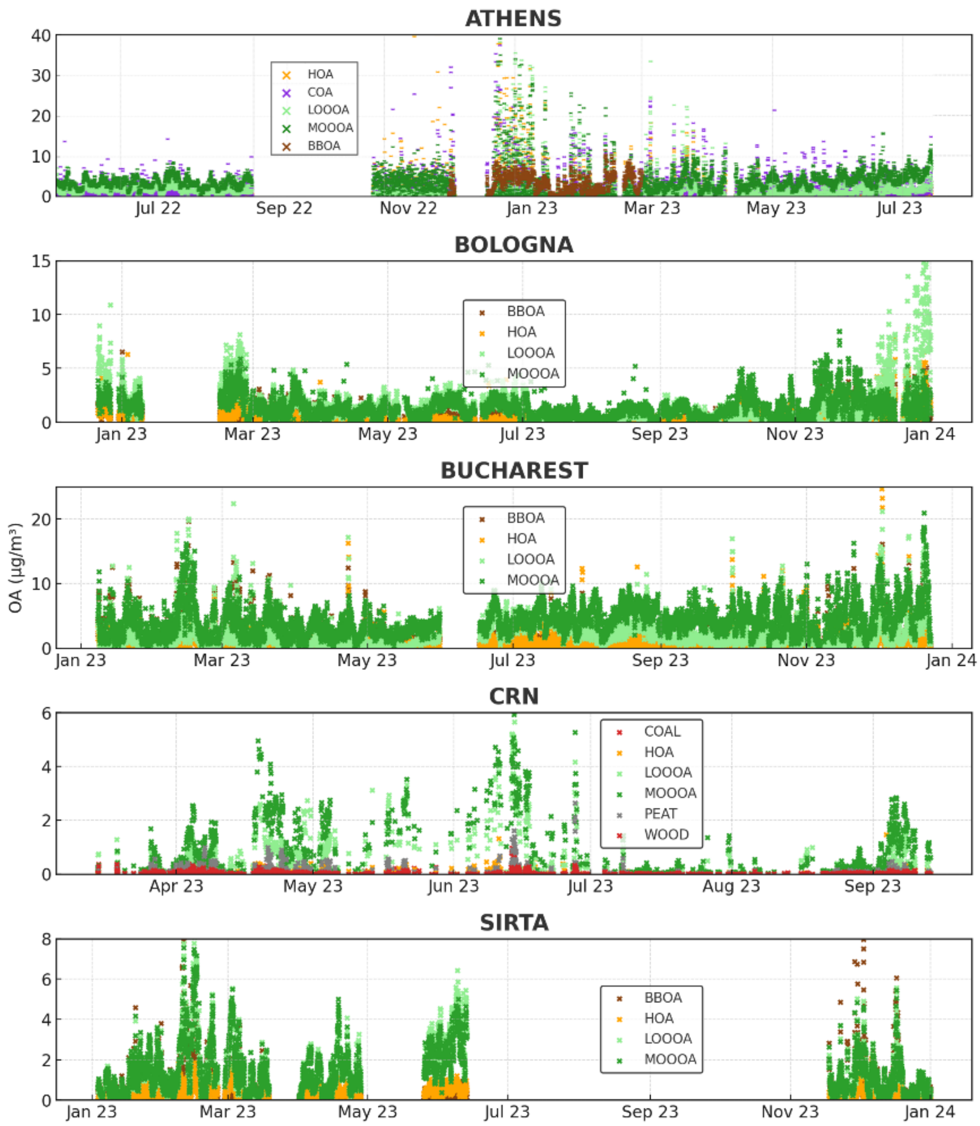

Fig. 2: Timeseries of OA sources in the study sites

4.4 Conclusion and possible impact

From the above analysis, it can be concluded that the sites located in urban and sub-urban areas showed higher bias with the CAMS-model derived PM. The source apportionment of OM in PM_1 conducted in five sites for the 1-year period within the time window of 2022-23 has yielded significant insights into the sources of OM. Overall, the results revealed seven distinct OM factors. These factors include primary organic aerosols (peat, wood, and coal burning; hydrocarbon-like organic aerosols), cooking OA, and two types of oxygenated organic aerosols (less oxidized OOA and more oxidized OOA).

The detailed source apportionment provides valuable insights of different OM factors, thereby enhancing our understanding of air pollution sources. This information is crucial for evaluating and improving model predictions, which in turn can enhance the accuracy of regional aerosol models.

In conclusion, the successful completion of M28 advances our scientific understanding of OM sources in five distinct sites across Europe.

4.5 References

AMAP: AMAP Assessment 2015: Black carbon and ozone as Arctic climate forcers. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, vii + 116 pp, 2015.

CCAC: Air pollution in Asia and Pacific: Science-based solution, United Nations Environment Programme (UNEP), ISBN 978-9-2807-3725-7, 2019.

Chen, G., et al.: European aerosol phenomenology − 8: Harmonised source apportionment of organic aerosol using 22 Year-long ACSM/AMS datasets. Environ. Int. 166. https://doi.org/10.1016/j.envint.2022.107325,2022

Daellenbach, K.R., et al.: Organic aerosol sources in the Milan metropolitan area – Receptor modelling based on field observations and air quality modelling. Atmos. Environ. 307. https://doi.org/10.1016/j.atmosenv.2023.119799, 2023

Freney, E., et al.: The second ACTRIS inter-comparison (2016) for Aerosol Chemical Speciation Monitors (ACSM): Calibration protocols and instrument performance evaluations. Aerosol Sci. Technol. 53, 830–842. https://doi.org/10.1080/02786826.2019.1608901, 2019

Ilie, A., et al.: Spatiotemporal Variability of Urban Air Pollution in Bucharest City. Atmosphere (Basel). 14, 1–16. https://doi.org/10.3390/atmos14121759, 2023

Lin, C., et al.: The impact of traffic on air quality in Ireland: Insights from the simultaneous kerbside and suburban monitoring of submicron aerosols. Atmos. Chem. Phys. 20, 10513–10529. https://doi.org/10.5194/acp-20-10513-2020, 2020

Ng, N.L., et al.: An Aerosol Chemical Speciation Monitor (ACSM) for routine monitoring of the composition and mass concentrations of ambient aerosol. Aerosol Sci. Technol. 45, 780–794. https://doi.org/10.1080/02786826.2011.560211, 2011

Ovadnevaite, J., et al.: Air Pollution Sources in Ireland, EPA Research. 2021.

Stavroulas, I., et al.: Sources and processes that control the submicron organic aerosol composition in an urban Mediterranean environment (Athens): a high temporal-resolution chemical composition measurement study. Atmos. Chem. Phys., 19, 901–919. https://doi.org/10.5194/acp-19-901-2019, 2019

UNEP: The Emissions Gap Report 2017, United Nations Environment Programme (UNEP), Nairobi, 2017.

Via, M., et al.: Rolling vs. seasonal PMF: Real-world multi-site and synthetic dataset comparison. Atmos. Meas. Tech. 15, 5479–5495. https://doi.org/10.5194/amt-15-5479-2022, 2022

5. History of the document

Version	Author(s)	Date	Changes
Version 1	Vignesh Prabhu, Jurgita Ovadnevaite	06/JAN/2025	New document
		30 Jan 2025	Sent for feedback
	S. Walter	10 Feb 2025	Finalised and uploaded