Deliverable D24 (D4.3)

Summary: source apportionment pilots, sustainability and associated benefits

RI-URBANS

Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS (GA n. 101036245)

By

CNRS, FMI & ICL

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Deliverable D24 (D4.3): Summary: source apportionment pilots, sustainability and associated benefits

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Comments	This document describes the pilot phase, summary of main findings, as well as recommendations, next steps and future upscaling potential of NRT-SA tools. This is a result of WP4 from the task of the pilot cities focusing on this issue. The pilot on NRT-SA of carbonaceous aerosols employs the source apportionment tool for the Aerosol chemical speciation monitor (ACSM; organics, sulfate, nitrate, ammonium and chloride) and multi-wavelength aethalometer (AE, Black carbon) data. Expected outputs include contributions of primary sources such as traffic, wood burning, and cooking (depending on measurement site), quantification of the secondary organic aerosols (SOA) fraction, automatic transfer of data (organic aerosols matrices and aethalometer BC concentrations) to ACTRIS DC.

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1. About this document

WP4 aims to test and to demonstrate solutions for advanced urban Air Quality (AQ) monitoring systems and evaluation of exposures (WP 1-3) at representative areas and hot spots in Europe. Specifically, implementing 5 testing and demonstration pilots in (originally) 9 cities (Athens, Barcelona, Birmingham, Bucharest, Helsinki, Milano, Paris, Rotterdam-Amsterdam and Zurich, with at least 3 cities in each pilot) and create synergies with WP5 to devise the roadmap for upscaling service tools (STs). The pilot on NRT-SA of carbonaceous aerosols employs the source apportionment tool for the Aerosol chemical speciation monitor (ACSM; organics, sulfate, nitrate, ammonium and chloride) and multi-wavelength aethalometer (AE, Black carbon) data. Expected outputs include contributions of primary sources such as traffic, wood burning, and cooking (depending on measurement site), quantification of the secondary organic aerosols (SOA) fraction, automatic transfer of data (organic aerosols matrices and aethalometer BC concentrations) to ACTRIS DC.

D24 (D4.3) describes the pilot phase, summary of main findings, as well as recommendations, next steps and future upscaling potential of NRT-SA tools.

This is a public document that will be distributed to all RI-URBANS partners for their use and submitted to the European Commission as a RI-URBANS deliverable D24 (D4.3). This document can be downloaded at https://riurbans.eu/work-package-4/#deliverables-wp4

2. Summary pilot phase

Before the pilot phase, all stations started the measurements and established the dataflows to servers. During the pilot year (2023) the stations ensured that measurements are running and dataflows work. The responsibility of each station included maximizing the data coverage of the instruments, as well as running monthly manual source apportionment in order to establish information about the changes in the sources of organic aerosols. The results of monthly PMFs were compared to NRT-SA results. Monthly meetings were conducted to discuss the results and resolve issues as well as possible discrepancies between offline PMF and NRT-SA. The NRT-SA and visualization of results were done in a centralized way in the common data server. After the pilot year the stations conducted offline source apportionment for ACSM data using state-of-the-art methods (seasonal and rolling Positive Matrix Factorization (PMF) analysis). The results of NRT-SA were compared to offline PMF results. Figure 1. shows a summary of the actions taken before, during and after the pilot year in each site.

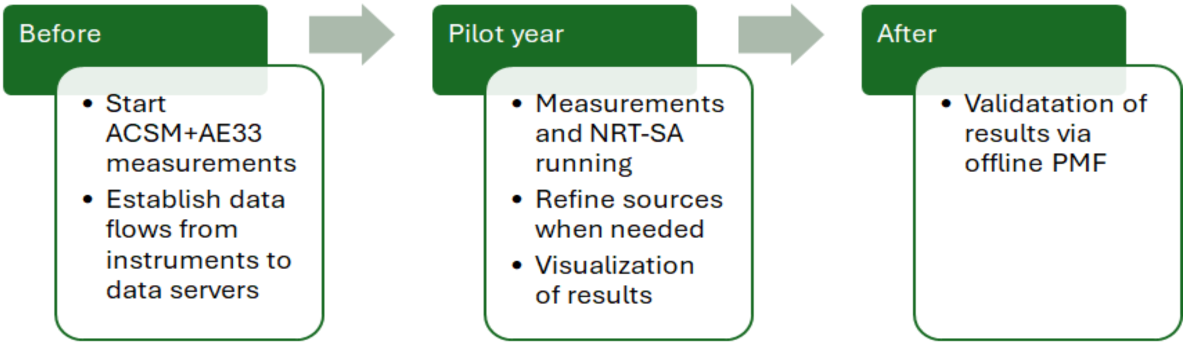

Figure 1*. Actions taken before, during and after the pilot year (2023).*

2.1 Pilot cities

During the pilot phase (year 2023) measurements were conducted at 12 stations (11 pilot sites + one follower site) situated in different environments (2 traffic, 9 urban and 1 suburban) across 8 cities. Figure 1 shows the locations of these pilot sites. Figure 2 includes locations of the sites and Table 1 includes information about the pilot sites.

***Figure 2**. Geographical locations of the near-real-time source apportionment (NRT-SA) pilot sites.*

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Table 1. *information about the pilot sites.*

Pilot city	Station Name	EBAS Station ID	Lat (°) N	Lon (°) E	EBAS Platform ID	EBAS Lab ID	framework	EBAS Station Name
Paris	SIRTA	FR0020R	48.719	2.149	FR0020S	FR01L	ACTRIS / RI-URBANS	SIRTA Atmospheric Research Observatory
Paris	Chatelet	FR0041U	48.86214	2.34462	FR0041S	FR21L	RI-URBANS	Paris Chatelet
Paris	BPEst	FR0042U	48.83857	2.412713	FR0042S	FR21L	RI-URBANS	Paris BPEst
Athens	AthensNOA	GR0004U	37.97312	23.71805	GR0004S	GR07L	ACTRIS / RI-URBANS	Athens NOA
Athens	AthensDEM	GR0100B	37.99532	23.81672	GR0100S	GR05L	ACTRIS / RI-URBANS	DEM_Athens
Bucharest	RADO-Bucharest	RO0010R	44.344°N	26.012°E	RO0010S	RO03L	ACTRIS / RI-URBANS	RADO-Bucharest II
Helsinki	Supersite	FI0039U	60.19635	24.95222	FI0039S	FI03L	RI-URBANS	Mäkelänkatu
Po Valley	Milano	IT0025U	45.47833	9.23144	IT0025S	IT006L	ACTRIS / RI-URBANS	Milano Pascal
Po Valley	Bologna	IT0022C	44.52366	11.33833	IT0022S	IT006L	ACTRIS / RI-URBANS	ISAC Bologna II
Zürich	Kaserne	CH0010U	47.37759	8.530419	CH0010S	CH02L	RI-URBANS	Zürich-Kaserne
Marseille	Longchamp	FR0035U	43.30529	5.394716	FR0035S	FR17L	RI-URBANS	Marseille Longchamp
Barcelona	PalauReial	ES0019U	41.38744	2.115306	ES0019S	ES05L	ACTRIS / RI-URBANS	Barcelona

Table 2. *Description of the stations where NRT-SA was piloted.*

Site name, coord.	Description (type, previous source apportionment studies)	Operator
SIRTA	SIRTA-Paris, an ACTRIS observatory located in Palaiseau (20 km southwest of Paris), is a major site for monitoring urban background air quality in the Paris region, which has significant traffic and heating emissions. Equipped with ACSM, AE-33, SMPS, and trace gas monitors (NOx, CO, O3), the site supports detailed source apportionment and particulate matter studies. Research shows seasonal PM1 variations, with organic aerosols often exceeding 50% of PM1 in colder months due to wood combustion and traffic. PMF analyses using SIRTA's ACSM data identify key sources, including hydrocarbon-like (HOA) from traffic, biomass burning (BBOA) in winter, and secondary organic aerosols (LV-OOA and MV-OOA) formed through photochemical and heterogeneous processes. These insights, underscored by recent studies (Petit et al., 2021; Chen et al., 2022), highlight local and regional emissions' influence on air quality, supporting ongoing PMF analysis for improved source apportionment.	CEA, INERIS, CNRS
Chatelet	Paris Chatelet (48°51'44" N, 2°20'41" E, 35 m a.s.l), also named Paris-1er Les Halles, is an urban background supersite located in downtown Paris (1st district) within the Nelson Mandela garden nearly to the Forum des Halles mall, in a much-used pedestrian environment with many shops and restaurants (Abbou et al., 2024). Also considered as one of the major sites for monitoring urban background air quality within the Paris region with SIRTA-Paris, the Paris Chatelet station is representative of the average public exposure to pollution levels in the Parisian conurbation (~7 million inhabitants). This supersite in use since October 2019 is equipped with the state-of-the-art instrumentation for the monitoring of gaseous pollutants (NOx, O3, CO/CO2/CH4,…) and physico-chemical parameters of interest	Airparif, the Air Quality Monitoring Network for the Greater Paris area

	related to aerosol health impacts (including PMx mass concentrations, PN, size distribution and chemical composition, precursor gases, oxidative potential etc.). Available measurements of atmospheric aerosol components from ACSM and AE33 data showed that OA dominates PM1 (~50 %), pointing a clear seasonality with higher concentrations during cold periods (due to enhanced wood combustion for residential heating) and in summer (reflecting an increased photochemistry favoring SOA formation from biogenic and anthropogenic sources) (Chebaicheb et al., 2024). Source apportionment analyses allowed identifying three local primary OA sources related to road-traffic (HOA), biomass burning (BBOA) and cooking (COA) activities, as well as two oxygenated OA sources depending on their oxidation state (LO-OOA and MO-OOA) (Chebaicheb 2023).	
BPEst	Paris BPEst (48°50'19"N, 2°24'46"E, 48 m a.s.l) is an urban traffic site located on the east side of the Parisian ring road which consists of 2x4 lanes for motorized traffic separated by a median strip, for a total width of 32 meters. The Annual Average Daily Traffic is around 243 400 vehicles per day (statistics from the Regional Council, 2017). The road fleet composition is characterized by a large proportion of private vehicles (77 %) followed by utility vehicles (13 %), two-wheeled vehicles and heavy goods vehicles (< 10 %) (Airparif' emission inventory, year 2018). Continuous measurements of NOx, PM10/PM2.5 (since 2012), BC (since 2014), PM1 chemical composition (since 2021) and PNC (started in 2024) are performed at the Paris-BPEst station. High PM1 mass concentrations were recorded, standing out notably on noticeable OA (44 %), BC (17 %) and SO4 (13 %) proportions. Significant contributions of BC at lower PM1 levels were observed, indicating a strong role of local combustion sources on OA levels (Chebaicheb et al., 2024). Indeed, the Paris-BPEst site can be influenced by the presence of fresh road traffic-related aerosol particles, especially when the wind speed is weak and blowing from north-east (NE) - east (E) to south (S). A first OA source apportionment analysis can be found in Chebaicheb (2023).	Airparif, the Air Quality Monitoring Network for the Greater Paris area
AthensNOA	The urban background Athens-NOA station is located at the Thissio area, close to the historical center of Athens, Greece, on top of a hill (105m a.s.l.) surrounded by a pedestrian zone where the influence of direct local emissions is limited. The site lies far from major roads in a moderately-populated residential area and is considered representative of urban background conditions in central Athens (Grivas et al., 2019). Based on previous source apportionment studies, four factors were identified during summer, namely HOA, COA, SV-OOA (LO-OOA) and LV-OOA (MO-OOA), and five factors during winter, the same as in summer with the addition of the primary biomass burning emissions (BBOA) which is 7-folded during winter nighttime highlighting the role of the residential heating (Stavroulas et al., 2019). The semi-volatile component of the oxidized organic aerosol SV-OOA was found to be clearly linked to combustion sources and a well-mixed type of aerosol (fast photochemical processes and oxidation of primary traffic and biogenic emissions) during the cold and warm period, respectively. Furthermore, LV-OOA presents a more regional character in summer.	National Observatory of Athens, Institute for Environmental Research and Sustainable Development
AthensDEM	The station is situated on the foot of Mount Hymettus in Agia Paraskevi, about 7 km northeast from the center of Athens (Vratolis et al., 2019) and is characterized as an urban background – suburban station, due to its proximity to the city-center. Therefore, it is influenced by fresh and aged traffic-related aerosol particles, especially when the prevailing wind has western (W) directions, and the regional background aerosol. MPSS, Nephelometer, AE33 aethalometer, EC/OC measurements are reported to ACTRIS database for over a decade.	ENRACT laboratory, NCSR Demokritos
Bucharest	RADO-Bucharest is a ground-based fixed ACTRIS National Facility located in south-eastern Romania, in a peri-urban area, flat terrain, 6 km south-west from Bucharest city. The aerosol in-situ measurements at the site were established in 2010, using state-of-the-art instrumentation for chemical and physical properties of particulate	National Institute of Research and Development for

	matter (e.g. composition, BC, size distribution, PN, PMs). The air quality is generally good, but several episodes with high PM concentrations are observed mostly during cold periods or long-range transport of pollutants (wood combustion burning or dust). Previous studies highlighted a clear seasonal trend of PM concentrations, with highest concentrations during cold season, when organic aerosols are the dominant fraction of PM1 at the measurement site, accounting for more than 50%. The source apportionment studies have shown four aerosol sources, among them the primary include traffic (HOA) and wood combustion (BBOA) sources, while secondary aerosols are represented by two oxygenated organic aerosols with oxidation degree (LO-OOA, MO-OOA) (Marmureanu et al. 2020, Chen et al., 2022).	Optoelectronics (INOE)
Helsinki – Supersite	Supersite is situated approx. 3 km north from downtown Helsinki at the kerbside of one of the main streets leading to the city centre. The heavily trafficked street consists of six lanes for motorized traffic, two rows of trees, two tram lanes, and two sidewalks, for a total width of 42 m (Hietikko et al., 2018). The traffic density is approx. 28 000 vehicles per weekday and a heavy-duty vehicle share of 10 % (statistics from the City of Helsinki). The Supersite established in 2015 is equipped with state-of-the-art instrumentation to measure concentrations and properties of gaseous and particulate pollutants (e.g. NOx, O3, PM, PN, size distribution, BC, LDSA, composition). The air quality at Helsinki is generally good, however episodes with elevated NO2, BaP and PM10 concentrations are occasionally observed due to traffic, wood combustion and road dust. Previous source apportionment studies at the site have shown that the main primary sources in the area include traffic (HOA), wood combustion (BBOA), Coffee roastery (CofR-OA) as well as , secondary aerosol formation (LV-OOA, MV-OOA) (Saarikoski et al. 2023, Chen et al., 2022).	Helsinki Region Environmental Services HSY, in collaboration with Finnish Meteorological Institute
Milano	The city of Milan is located in northern Italy, with a population of 3.22 million citizens it is considered the second most populated city in Italy after the capital Rome. The population of the wider Milan metropolitan area is estimated to be between 4.9 million and 7.4 million, which makes Milan the largest metropolitan area in Italy and one of the largest in the European Union. Milano Pascal is an Urban Background site, taking advantage of three different measurement sites located within the same area (200 m). The Milano Pascal area is located in the university area, mainly residential and it is considered to be the urban background station of the city; the average population density is 7500 km^{-2} in Milan. The CNR-ISAC mobile station "Voyager3" was located in the CNR Area for the RI-URBANS pilot. The field campaign started on January 19th, 2023, and ended on March 21st, 2024. The Voyager was equipped with Tof-ACSM, AE-33, VOCUS-ToF-PTR-MS, GHGs Picarro (since July 2023) and NOx (since September 2023). The ARPA Lombardia AQ station was located only 50m far away, while at Milano University PNSD measurements were carried out by Helsinki University (SMPS, PSM and NAIS).	National Research Council of Italy, Institute of Atmospheric Sciences and Climate
Bologna	Bologna is located in the southern part of the Po Valley (Italy), at the foot of the Apennines, and is an important population basin for the region (400,000 inhabitants) impacted by significant industrial and agricultural activities and crossed by several major highways. The Bologna measurement site is located in the CNR Research Area (44°31'29'' N, 11° 20'27' E), about 4 km apart from the city center, on the roof of a five-story building. A previous organic aerosol source apportionment characterization, based on multiple short-term campaigns, can be found in Paglione et al. (2020).	National Research Council of Italy, Institute of Atmospheric Sciences and Climate
Kaserne	Kaserne site is an urban background station located in downtown Zurich, Switzerland's most populous city with around 400,000 inhabitants. A Q-ACSM was deployed at this site during a field campaign from August 2022 to July 2023.	Paul Scherrer Institute (PSI)

	Continuous measurements of regulated PMx and gases are also conducted by the Swiss Federal Laboratories for Materials Science and Technology (EMPA). Several previous source apportionment studies using Q-ACSM organic data at Kaserne have identified key sources (Canonaco et al., 2015, 2021; Chen et al., 2022), including HOA, COA, BBOA, secondary organic aerosol components separated based on their oxidation degree (LO-OOA, MO-OOA), and a cigarette-smoke-related organic aerosol factor (CSOA).	
Longchamp	Marseille-Longchamp supersite is an urban background station located in Marseille, the second most populated city in France (about 870 000 inhabitants). The site is located in the heart of the Parc Longchamp, in the fourth district of Marseille (43°18'20'' N; 5°23'41; m a.s.l.). The city encounters dense traffic and hosts one of the largest harbors of the Mediterranean Sea (2 km away). In addition, the large industrial complex of Fos-sur-mer with petroleum refining, shipbuilding, steel facilities, and coke production plants is located 40 km north-west of the city. The region is well known for active photochemistry, inducing high secondary pollutants formation during summer periods. Since 2017, the site has been equipped with state-of-the-art instrumentation for continuous aerosol measurements (ToF-ACSM, AE33, Xact, SMPS). OA is the dominant fraction of PM1 accounting for 45-50% in all seasons (Chazeau et al., 2021). Previous source apportionment studies have identified six OA sources (Chazeau et al., 2022; Chen et al., 2022). SOA factors, separated based on their oxidation degree (MO-OOA, LO-OOA), dominate the organic composition. The other sources included HOA, COA, BBOA and the new defined Sh-IndOA which was related to the mix between shipping and industrial plumes.	Laboratoire Chimie Environnement (LCE UMR7376 CNRS - AMU) and AtmoSud (the regional air quality monitoring network)
Palau Reial	Barcelona- Palau Reial (PR; 41° 23' 15" N; 02° 07' 05" E; 80 m a.s.l.) is a supersite representative of the urban background in Barcelona, Spain. It is located in a residential area at the NW of Barcelona at 200 m distance from one of the most concurred avenues of the city (>60.000 vehicles per working day in 2014-2018). The atmospheric dynamics of the area are dominated by breeze regimes, consisting in a nocturnal NW wind component, a diurnal breeze development turning from SE to SW direction, and highest wind speeds around noon. Source apportionment revealed OA was 46% to 70% of secondary origin (SOA), with different oxidation status: less-oxidized (LO-OOA) and more-oxidized (MO-OOA). The anthropogenic primary OA sources identified were cooking-like OA (COA), hydrocarbon-like OA (HOA), and biomass burning OA (BBOA). They have shown a decreasing trend from 2014 to 2018 (Via et al., 2021; Via et al., 2022). Advanced source apportionment (SA) techniques were applied: multi-time resolution PMF (MTR-PMF), using a) non-refractory submicronic species measured by Q-ACSM, b) black carbon measured by an aethalometer, and c) major and trace elements in PM1 measured by offline techniques using samples collected on quartz-fibre filters. The MTR-PMF resolved eight PM_1 sources: ammonium sulphate + heavy oil combustion (25%), ammonium nitrate + ammonium chloride (17%), aged SOA (16%), traffic (14%), biomass burning (9%), fresh SOA (8%), cooking-like organic aerosol (5%), and industry (4%) (Via et al., 2023).	Institute of Environmental Assessment and Water Research (IDAEA), Spanish National Research Council (CSIC)

2.2 Instruments

An aerosol chemical speciation monitor (ACSM), with either quadruple (Q) or Time-of-flight detector (ToF), and filter photometer. A summary of used instruments is collected to table 3. See Milestone **M17 (M4.1): Source apportionment started,** for the detailed description of the instrumentation.

Table 3. *The used instrumentation, cut-off sizes and references related to the previous studies conducted at the site.*

Name of Site	Station type	ACSM type	Reference for SA	Filter photometer type	Size Cut-off	Reference for SA
Paris, SIRTA	Periurban	Q-ACSM		AE33	PM10 & PM1	Petit et al., 2021
Paris, Chatelet	urban background	Q-ACSM	Chebaicheb H., 2023	AE33	PM2.5	Savadkoohi et al., 2023
Paris, BP Est	Urban traffic	Q-ACSM	Chebaicheb H., 2023	AE33	PM2.5	Savadkoohi et al., 2023
Athens, NOA	Urban	Q-ACSM	Stavroulas et al., 2019	AE33	PM2.5	Savadkoohi et al., 2023
Athens, Demokritos	Suburban	ToF-ACSM	Zografou et al., 2022	AE33	PM10	
Bucharest, INO	Periurban	Q-ACSM	Chen et al., 2022; Marmureanu et al., 2020	AE33	PM10	Savadkoohi et al., 2023
Helsinki, Supersite	Street canyon	Q-ACSM	Chen et al., 2022, Saarikoski et al., 2023	AE33	PM1	Helin et al., 2018, Saarikoski et al., 2021
Po Valley, Milano	Urban	ToF-ACSM	Chen et al., 2022 Paglione et al., 2020	AE33	PM10	Sandradewi et al., 2008 Zotter et al., 2017
Po Valley, Bologna	Periurban	Q-ACSM	Chen et al., 2022	AE33	PM10	Sandradewi et al., 2008 Zotter et al., 2017
Zürich, Kaserne	Urban	Q-ACSM	Chen et al., 2022	AE33	PM2.5	Savadkoohi et al., 2023
Marseille, Longchamp	Urban	ToF-ACSM	Chazeau et al., 2022 and Chen et al., 2022	AE33	PM2.5	Savadkoohi et al., 2023
Barcelona	Urban	Q-ACSM	Via et al., 2021 ACP	AE33	PM2.5, PM10	Savadkoohi et al., 2023

2.3 Software tools

Several software packages were needed to conduct NRT-SA as described in figure 3.

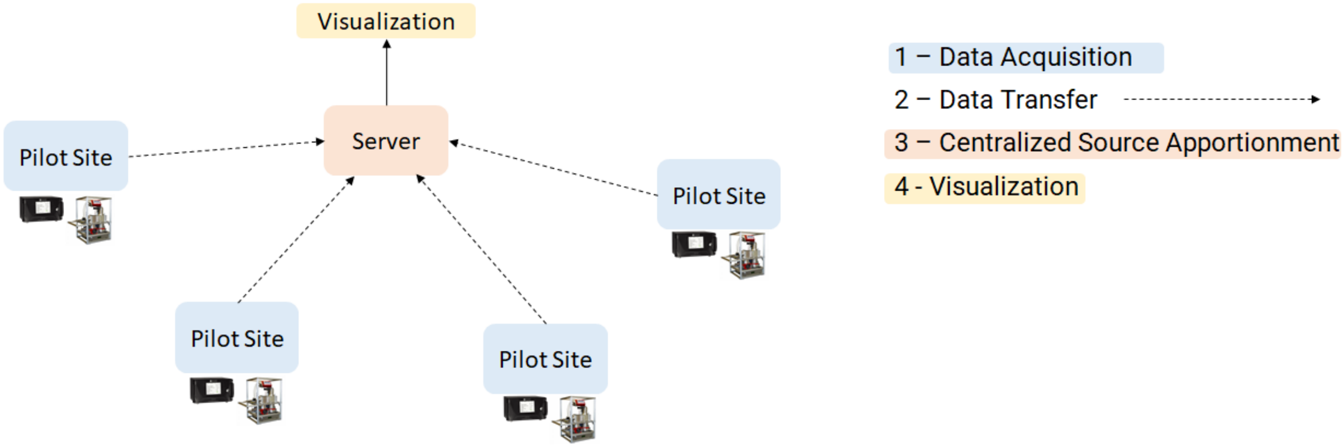

Figure 3. *Data flows from station to server and visualization*

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Following software packages were used in the measurements

- **IGOR Pro software, version 6-9,** (Wavemetrics) in ACSM to conduct measurements, data handling and to prepare PMF matrices
- **ECAC tools** to transfer data (ACSM + AE33) from pilot sites to NextCloud server
- **NextCloud** to store data from pilot sites and conduct centralized source apportionment
- **SOFI-PRO,** Datalystica to conduct source apportionment offline and online
- **SOFI-RT** for organic source apportionment.
- **Python scripts** for BC source apportionment

A more detailed description of softwares can be found from Milestone M17 (M4.1).

3. Summary and quality of the results

Most of the results from the pilot phase are already available in previous deliverables and milestones:

- D22 (D4.1): https://riurbans.eu/wp-content/uploads/2024/04/RI-URBANS_D22_D4_1.pdf
- D23 (D4.2): https://riurbans.eu/wp-content/uploads/2024/05/RI-URBANS_D23_D4_2.pdf
- M17 (M4.1): https://riurbans.eu/wp-content/uploads/2022/09/RI-URBANS_M17.pdf
- M18 (M4.2): https://riurbans.eu/wp-content/uploads/2023/02/RI-URBANS_M18.pdf

The quality of NRT OA SA outputs has been evaluated with manual PMF analyses (monthly analyses, and a year-long rolling PMF) with a harmonized protocol, following Chen et al. (2021). Figure 4 presents an overview of the comparison at each site between NRT and manual analyses for each OA fraction. For the sake of the comparison, sub-OOA fractions (LO-OOA & MO-OOA), if present, were summed as OOA. Global performance metrics (Pearson r, Mean Bias and Root Square Mean Error) are also presented in Table 3. Even if global metrics are encouraging, a striking feature of the comparison is that the reliability of NRT results is highly dependent on the site. Indeed, the variability of NRT primary OA factors (HOA, BBOA or COA) at Site#2 and Site#3 is inconsistent with rolling PMF. This is for instance a clear case of misconfiguration of the NRT SA software, causing unrealistic values (sometimes > 100 µg/m^3). This also demonstrates the need of QA/QC procedures for NRT SA outputs in order to avoid these kinds of values from being considered. Interestingly, higher correlation is obtained for OOA fraction, despite highest MB and RMSE. Similarly, rather good performance is observed for the sum of primary OA factors (HOA, COA, BBOA) with lower site-to-site variability, suggesting that at that stage, the apportionment between POA and OOA fractions is more consistent than individual OA factors.

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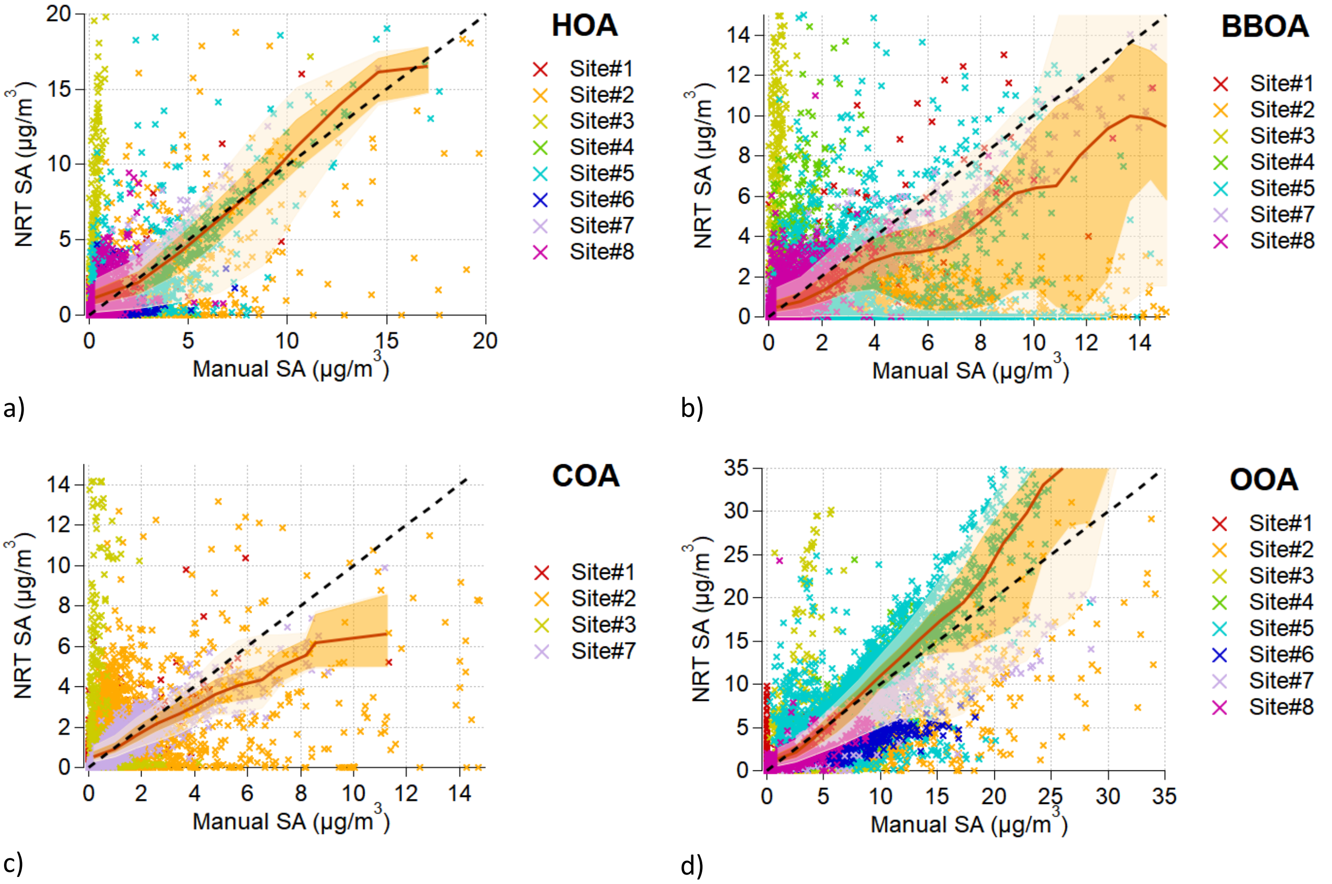

a) b) c) d)

Figure 4. *Scatter plots of hourly NRT SA results versus Manual SA for HOA (a), BBOA (b), COA (c) and OOA (d). Colored dots correspond to the data of each individual site; red line and shaded areas correspond respectively to the median and 10th, 25th, 75th, 90th percentiles concentrations (calculations were performed without Site#2 & Site#3).*

Table 3. *Global performance metrics (GPM) for the different OA fractions: Pearson r coefficient, Mean Bias (MB), and Root Square Mean Error (RMSE). Calculations were performed without Site#2 and Site#3; values in parenthesis correspond to calculations with those sites.*

	HOA	**BBOA**	**COA**	**∑POAs**	**∑OOAs**
r	0.50 *(0.11)*	0.61 *(0.24)*	0.89 *(0.12)*	0.80 *(0.31)*	0.78 *(0.46)*
Mean Bias (µg/m3)	-0.05 *(0.08)*	-0.16 *(-0.24)*	-0.01 *(0.15)*	-0.20 *(-0.14)*	-0.18 *(-0.5)*
Root Square Mean Error (µg/m3)	1.41 *(7.28)*	1.27 *(2.69)*	0.34 *(4.23)*	1.53 *(5.76)*	3.07 *(6.5)*

As presented and discussed in previous documents, the discrepancies observed throughout the pilot phase, and summarized here, may arise from several causes, some of which have been already identified:

- Wrong input data leads to wrong results. Acquisition configuration is critical. No prior QA/QC of input data was performed during the pilot phase.
- NRT SA parameters need to be optimized (e.g. number of PMF runs, of factors, constraining profiles).
- External tracers (such as BC fractions) may help to refine the solution space.
- Technical QA/QC of NRT SA outputs (e.g. residuals, or other metrics) was not implemented.

BC SA is technically less challenging than OA SA, because:

- Source apportionment calculations require less computational effort, and were based on Python, enabling efficient and collaborative maintenance, debugging and updates.
- Source apportionment calculations are based on data which are already supported by ACTRIS DMP.

To this matter, BC SA can be considered as a more mature process than OA SA.

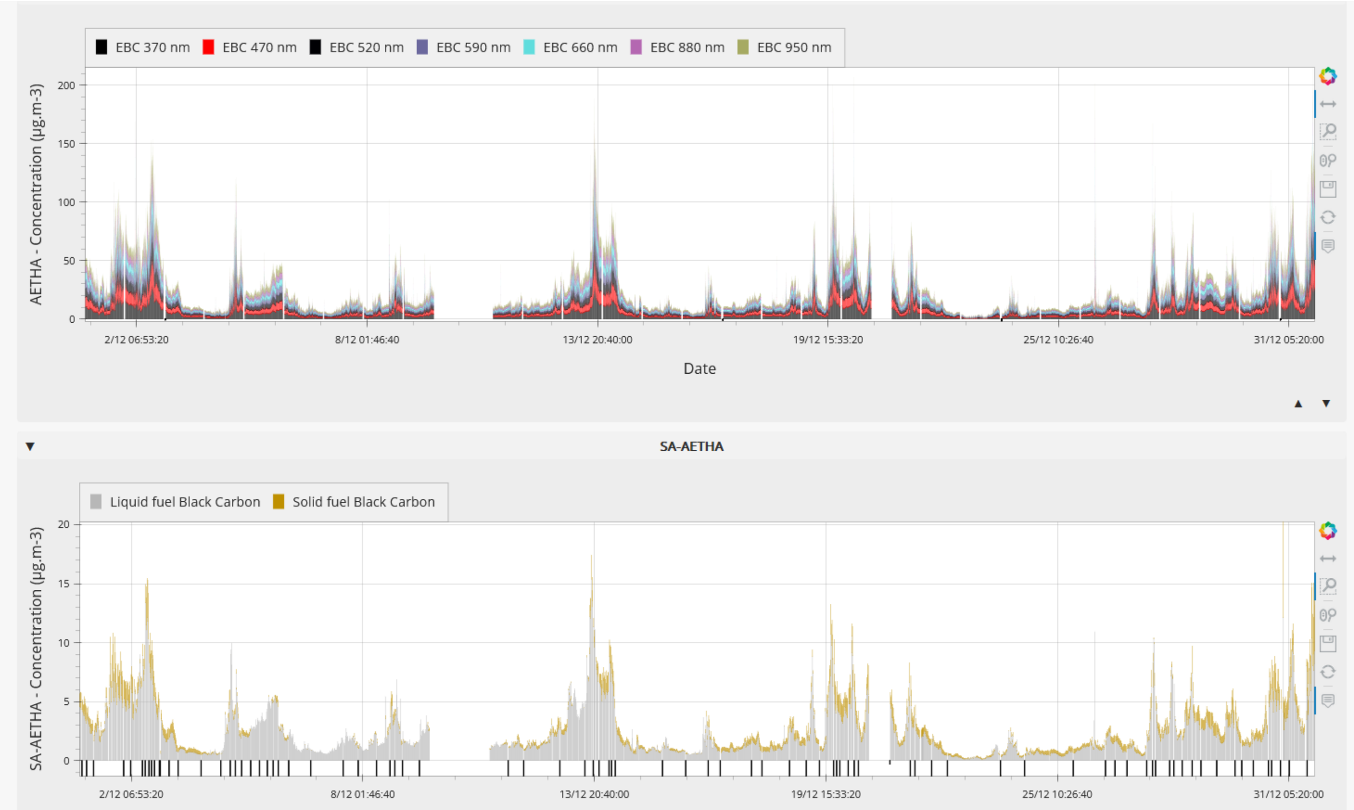

Figure 5. *Temporal variations of BC (upper panel) and BC fractions (Liquid fuel & Solid fuel, bottom panel) in December 2023 at Site#5*

These results highlight that **NRT source apportionment of OA and BC is feasible**. They also **validate the proof of concept of a centralized process** with simultaneous calculations, paving the way for future work from the perspective of having an **operational service**. The further necessary steps towards operationality are described and discussed in the following sections.

4. Strengths and limitations of NRT-SA

The NRT-SA was run for the pilot year using data from the ACSM and aethalometer separately. The software used to conduct the source apportionment were fairly different, thus the strengths and limitations are separately listed below for the ACSM and aethalometer SA.

Regarding, Near Rear Time Source apportionment of ACSM data, **multiple strengths for conducting organics NRT-SA based on ACSM data were found:**

- NRT-SA approach automatizes the process and at same time minimizes the influence of the user on the result. During the pilot year, NRT-SA was conducted in a centralized way, so that at each site a software was used to construct needed datafiles that were sent to a centralized server and analyzed automatically without prior expectations about the result. However, prior information of the number of the sources and/or representative factor profiles at each individual site need to be known for NRT-SA.
- The NRT-SA provides the results to users in real-time ensuring that authorities, public and scientific community have immediate information of the sources that affect the air quality. The conventional offline source apportionment is time consuming for long-term datasets. With a traditional approach, it takes months after the measurements to retrieve high-quality SA-results for one year of data. Also, the source apportionment results are typically available for certain campaigns, not continuously, which limits the usability of the results. NRT-SA data would be continuously available.
- NRT-SA results can be made available in easily understandable format in realtime to anybody and can be made available for further use e.g. by API developers or air quality modeling community. The offline source apportionment results are typically published as scientific articles, that are not understandable for the general public.

However, multiple limitations/challenges for conducting organics NRT-SA based on ACSM data were found:

- Organic Aerosols SA is not PM SA. Although OA can make up a significant fraction of PM1, the results provided by OA SA do not provide information about the sources of the other components of PM1.
- Experienced users with long-term experience on ACSM measurements and source apportionment are still preferred to run the measurements and conduct the NRT-SA, particularly the quality control of the analysis.
- Knowledge about the sources and source profiles are essential for NRT-SA. Users have to give a number of factors, their names and profiles of primary factors (i.e. mass spectra) into the software. Primary sources (e.g hydrocarbon-like, biomass burning organic aerosol) or especially their characteristic mass spectra need to be constrained. If prior information is not available, the number of sources can be guessed and factor profiles taken from the literature. However, this strongly affects the reliability of the results.
- It is important that the measured input data is correct, as errors in the input data cause wrong SA results. Also changes in the input data caused e.g. by technical reasons such as filament change can cause changes to instrument response which affect input data and thus the NRT-SA results. Fragmentation tables need to be consistent throughout the process.
- Changes in particle sources over time were found to be problematic for the NRT-SA. Current NRT-SA software version cannot identify when the sources appear/disappear, thus the user has to be aware and modify the prior information file in time. For example, biomass burning factor (BBOA) is typically only seen in winter, thus at some point in autumn the factor needs to be added to the list of factors and removed in spring. We note that this is problematic also for offline PMF.

- Optimizing the NRT-SA parameters was found to be essential. Chapter 5 gives an overview of optimization done in this project.
- Disruptions in measurements due to technical issues were observed during pilot year, causing breaks also to source apportionment. They can come e.g., from instrumental break down, internet outage preventing the data from being sent, and bugs in the centralized server.
- Validation of NRT-SA results is important. We chose to compare the NRT-SA results on state-of-the-art PMF analysis results done based on offline data by experienced source apportionment users. This is both time consuming and needs experienced personnel to conduct. NRT-validation processes are needed in order to evaluate the quality of the apportionment.
- NRT OA SA may not always be self-sufficient. External co-located air quality data (e.g. BC, NO_x, PM2.5 mass concentration, aerosol particle number size distribution) may be needed to ensure and improve the quality NRT-SA results.
- Software is not open access, which may limit the potential users and upscaling.

Regarding, Near Rear Time Source apportionment of BC data we identified several strengths and weaknesses as follows:

Strengths

- Mature measurement technique that is easy to run continuously. The data gaps due to technical issues or other malfunctions for aethalometer were very uncommon during the pilot year, which made the NRT-SA process fairly easy to implement.
- Soot photometers (e.g. Aethalometer) produces data as a compact .txt files. Thus, the data amounts that need to be sent to the server and for the visualization are very small making the process feasible.
- The SA using the “aethalometer model” is easy to automate and produces reliable results. The aethalometer model with two fixed sources (solid and liquid fuel burning emissions) used in source apportionment is straight forward and can be easily automated.
- NRT-software is made in the project and can be made openly accessible to all users.

Limitations

- A priori knowledge of BC sources is preferable for the application of the aethalometer model. Optical signatures for liquid fuel (AAE_{LF}, mostly from road traffic) and solid fuel (AAE_{SF}, mostly from biomass and in some cases coal combustion) are needed for the SA analysis and these values can change based on the prevailing sources and their properties (Savadkoohi et al., 2023, Helin et al., 2021).
    - Values of around 1 and 2 for the two wavelengths of 470 and 950 nm are commonly used for traffic and residential, commercial sources (e.g., biomass burning and coal combustion), respectively (Sandradewi et al., 2008a), if site specific values are not available.
    - External tracers (e.g. m/z 60 from ACSM measurements, formed in incomplete combustion of levoglucosan) can help optimizing AAE_{SF} and AAE_{LF} .
    - Seasonal percentiles can be used when the site only has AE33 measurements.
- Amount of information produced by the SA can be limited, as it only splits the sources to traffic (liquid fuel) and biomass (solid fuel) combustion, which may not exist in all environments and seasons (e.g. summer). Also, technique is not optimal in an environment where one source is dominating.

- Naming convention: “liquid” and “solid” nomenclature does not explicitly refer to pollution sources but to the type of fuel used. Bud liquid in urban areas mostly refer to road traffic and solid to biomass and/or coal combustion.

5. Recommendations

As the used NRT-SA software were very different for the ACSM and BC, the recommendations are separated based on the instrument providing the data.

Source apportionment of organic aerosols (ACSM)

- Ensure the quality of the input data used for the source apportionment. We recommend developing automatic quality control protocols to prevent publishing clearly wrong NRT-SA results. The automated QA/QC developed in ACTRIS from ACSM data can be a relevant starting pillar, but specific procedures shall be implemented for the PMF input as well.
- Ensure the data transfer system is robust to avoid data gaps. Ensure there is a plan for data gaps e.g. due to technical issues. In a case of a data transfer gap, the system should be able to check for the gaps and reprocess the NRT retrospectively to provide a continuous data set.
- Optimize the source apportionment parameters. Based on the experiences in this pilot study, some recommendations for the NRT-SA are provided particularly for the aerosol sources as follows:
    - Refine the POA factor profiles for rolling PMF in on-line SA analysis by using site-specific factor profiles from previous seasonal PMF analysis if data is available.
    - For sites without a priori information and no sufficient data available, following factor profiles are suggested: BBOA from Ng et al. (2011), HOA and COA from Crippa et al. (2014) which can be downloaded here: https://datalystica.com/spectral-database/. In addition, one OOA factor is recommended if there is no comprehensive information for the sampling site. Once sufficient data is available, following the first recommendation to get the site-specific factor profiles.
    - The rolling PMF settings in online SA analysis need to follow Chen et al. (2022b):
        - Using random a-values with upper a-value of 0.4 (or smaller) to constrain POA factors.
        - Enable bootstrap with >50 iterations for each window.
        - 14-day window with 1-day step.
    - Criteria-based selection is essential to retrieve solid factor profiles for Chemical Mass Balance (MCB)-like analysis, therefore, complementary data, like BC, NO_x shall be imported in a real-time manner for determining the “goodness” of the HOA factor. For BBOA factor (if exist), explained variation of m/z 60 shall be applied; f44 shall be sorted for unconstrained OOA factors (if two OOA are present) to situate the MO-OOA in the designated position. T-test of temporal criteria (i.e., correlation, explained variation) shall be applied with a p-value of 0.05 to determine the environmentally reasonable solutions from the on-line rolling PMF analysis.
    - Start a new on-line SA analysis with different rolling PMF settings if different number of factors are expected at different times, for instance, due to the absence of BBOA in summer.
- QA/QC of output data is important. We recommend evaluating the SA output against results given by state-of-the-art offline source apportionment methods such as PMF conducted by experienced users. Regular off-line QA/QC will improve the quality of NRT SA and the insights from the off-line analysis can be implemented

into the NRT process, for example linked to seasonal cycle of sources and identification of new site specific sources.

Source apportionment of black carbon

- Ensure the latest software and firmware versions are used in the measurements and instruments.
- Include all necessary information in metadata in order to harmonize the parameters between instruments if data from several measurement locations are used:
  - Time local-vs Coordinated Universal Time (UTC).
  - Standard Temperature and Pressure (STP) conditions or local atmospheric conditions.
  - Time resolution of measurements.
- Ensure the quality of input data:
  - Remove impact of filter changes from final data prior SA.
  - Automatic QA/QC of data prior to SA with status codes and other technical metrics.
- Document all changes and observations to the measurement diary to ensure traceability of the results in case of doubt. Also, it is important to document any changes e.g. in filter type, parameters or software. Make sure that this information are accessible automatically, in eg metadata.
- It is important to conduct quality assurance for SA e.g. against other co-existing measurements to ensure correct operation of both instrument and SA.

6. Next steps towards harmonized SA, upscaling and future work

Upscaling of SA to many sites will likely be seen in future, as the SA provides important data that would be beneficial to authorities, city planning as well as to the general public. In order to reach harmonized real time source apportionment, following aspects need to be considered and improved prior upscaling:

- **Reliability and trustability** of the results is utmost important as they are communicated to the public in real-time. Significant efforts are needed to ensure that instruments operate well, and that real time SA input and output data are valid. Automated QA/QC procedures shall be developed by defining variables, thresholds and metrics to assess the quality of the data. Specific additional work shall also focus on the determination of the overall uncertainties of the outputs.
- **Traceability** of the process shall be achieved by the determination of harmonized submission procedures. Because outputs are directly related to inputs, all the data used for source apportionment need to be accessible and well documented. **Input data and results need to be available**. Dedicated repository for SA matrices as well as SA results is needed.
- **Fairness and open-source:** optimally the SA tool would be openly accessible and available freely for other users and for further developments. The user community could help in the further development of an open access tool. This may also promote the use of the SA tool in air quality measurement networks. Fairness may also be achieved with a centralized process, which allows the intercomparability of the results and makes sure that all data go through the exact same procedures.
- **Standardization**: Harmonization of terms used for the sources needs to be done, so that the results between different stations are comparable. In this study, the sources were first split between constrained primary sources (e.g. HOA, COA, BBOA, coal burning) and un-constrained secondary sources (LO and MO-OOA). Naming conventions need to be settled, especially regarding how primary OA factors relate to sources, as well as secondary factors.

- **PM source apportionment**: SA end-users may have different needs. For air quality purposes, and quantitatively speaking, PM SA results should be more useful to OA or BC SA. To this perspective, dedicated efforts must be undertaken in order to go beyond OA source apportionment and be able to provide valuable information on PM sources.
- **Connection between different source apportionment analysis**. When comprehensive observations (e.g., aethalometer, ACSM, aerosol particle number size distribution, Volatile Organic Compounds) are performed in a co-located manner, connecting the source apportionment from the different instruments should be compared and interpreted together for a comprehensive view on the sources influencing air quality.

8. Conclusions

Running a centralized source apportionment in a way that all results are available in real-time is a big effort, that needs expertise from users as well as good skills with a variety of software, data transmission and servers. The results were observed to strongly depend on the correct input data and used SA tools. The impact of the results depends on capability to visualize and make the results available for end users.

For black carbon, the used source apportionment method, Aethalometer model, is a well-established and widely used, and non-complex approach to SA. The Aethalometer model-based SA was fairly easily automated and run in realtime. The dataflows were modest and results easy to visualize and understand. However, in some conditions, the approach, dividing the sources only to BC emissions from liquid and solid fuel combustion, was problematic. Especially, when there was only one very dominating source, e.g. in a traffic environment, the approach did not provide reliable results.

For ACSM state-of-the-art SA combining PMF and CMB was used. This needs experienced users to setup, previous knowledge on sources, as well as the SA software to run it. The mass spectrometer, ACSM, produces a very large and complex dataset, and a separate tool was developed to transfer the needed data to the server. Establishing the dataflows from all 13 sites was challenging. Furthermore, the SA tool needed significant amounts of information about sources, including source profiles, constraints etc. However, the SA provided significantly more information that could be further utilized in the emission mitigation, future emission limits and city planning. Also, real-time data could be utilized in the dissemination and further used in the air quality related applications and services. We note that real-time data validation is utmost important when the results are visualized and communicated in a real-time.

Both SA tools were found useful, however further development is needed in order to produce tools that can be easily adapted to any air quality network as a part of daily routine work.

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