Deliverable D5 (D1.5)

NRT Source Apportionment Service Tools for submicron carbonaceous matter (final)

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

By
ACMCC (CNRS & INERIS)

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Comments	This document summarizes the work performed regarding the establishment and implementation of near real time (NRT) source apportionment (SA) service tools (STs) for fine carbonaceous aerosols. This Deliverable is produced in WP1, T1.2 on developing and implementing advanced SA-STs. This task aimed at providing STs, based on best procedures and methodologies, to apportion novel health-related AQ metrics.

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1. ABOUT THIS DOCUMENT

This Deliverable D5 (D1.5) presents the near real time (NRT) source apportionment (SA) service tools (STs) for fine carbonaceous aerosols, and their associated software packages.

This Deliverable is produced in WP1, T1.2 on developing and implementing advanced SA-STs. This task aimed at providing STs, based on best procedures and methodologies, to apportion novel health-related AQ metrics. It allowed to evaluate and apply the most suited SA receptor models for operational applications, considering previous work in FAIRMODE, EMEP and COLOSSAL (COST Action: Chemical On-Line cOmpoSition and Source Apportionment of fine aerosol) and to provide pilot NRT-SA functionalities (harmonised with CAMS21a development and outcome) (D1.4-D1.5), with operational requirements of the SA software and data transfer/formatting STs for the novel NRT-SA of non-refractory aerosols (ACSM) and BC measurements data products, for modelling STs (WP3), pilot applications (WP4) and upscaling activities (WP5).

Carbonaceous particles, including black carbon (BC) and organic aerosols (OA), are representing a substantial part (typically, in the range 40-80%) of fine particulate matter (PM) in urban environment. At ACTRIS national facilities, their in-situ high-time resolution monitoring is usually conducted using aerosol chemical speciation monitors (ACSM) and multi-wavelength aethalometer (AE33), for OA and BC respectively. In the last decades, research activities allowed to develop novel methodologies to identify and quantify the main sources of carbonaceous aerosols measured using these two types of instruments. The aim of the present task within RI- URBANS was to implement such methodologies at a centralized server to demonstrate their ability to be operated - and thus to gain knowledge on these sources - in NRT. This demonstration was then showcased over the year 2023, in various European pilot cities, as part of WP4 Task4.1 activity.

The list and weblinks of open-source procedures and software developed in this task to operate the NRT SA STs is notably made available by the present document.

Please note that ACSM are commonly measuring submicron (PM1) aerosols while AE33 are generally installed with a PM2.5 sampling head within air quality monitoring networks (AQMN). However, as BC particles are known to be overwhelmingly present in the smaller aerosol fraction (< 1µm), it is assumed that the STs established for this RI-URBANS activity are addressing PM1.

This is a public document, available in the RI-URBANS website (https://riurbans.eu/work-package-1/#deliverables-wp1). The document will be distributed to all RI-URBANS partners for their use and submitted to European Commission as the RI-URBANS deliverable D5 (D1.5).

2. DATA WORKFLOW AND SOFTWARE PACKAGES DEVELOPED FOR BOTH SERVICE TOOLS

Since AE33 and ACSM instruments have their own data format and data treatment procedure specificities, different software solutions have been set-up to establish BC and OA SA-STs. They are detailed in the subsections hereinbelow. Software packages used in some components are highlighted and numbered (eg **BCSA_Sx**). The full availability of these components is presented in section 2.3. Please also note that a detailed description of the codes are already available in RI-URBANS Deliverable D4 (D1.4) at https://riurbans.eu/wp-content/uploads/2023/01/RI-URBANS_D4_D1_4.pdf

2.1 Black Carbon Source Apportionment

Figure 1 illustrates the data workflow set-up for BC SA. Raw data are generated by an acquisition software installed at the station (**BCSA_S1**). **BCSA_S1** consists in several Python procedures for the acquisition of various aerosol in-situ instruments. It has been developed with ECAC-WCCAP (Topical centre unit coordinating in-situ measurements of aerosol microphysical properties within ACTRIS). Data are stored whether in an Influx or Postgres Database, where text/ascii-format file are automatically generated. Raw data are automatically formatted hourly (thanks to a task scheduler) into ACTRIS Level0 datafile (NASA-AMES format), containing meta information about the measurements (e.g., instrument information, measurement conditions, AE33 parameters) and results for the most recent measurements (previous last hour).

In order to optimize the development of the codes and their implementation for 13 sites during the WP T4.1 pilot phase, Level0 data were automatically transferred to a centralized server (**BCSA_S2**) located at AERIS-ICARE (CNRS) as part of ECAC-ACMCC (Topical centre unit coordinating in-situ online measurements of aerosol chemical speciation within ACTRIS). A cloud service (NextCloud) has been chosen for this task in order to simplify data transfer through an automatic back and forth synchronization. This allowed for instance to send back source apportionment results directly to the pilot station for evaluation. Because ACTRIS compliant data were generated, Level0 files from all pilot sites were also sent in NRT to EBAS, after proper configuration of **BCSA_S1**.

A data quality control is applied (**BCSA_S3**) in order to generate Level1 files. Automatic checks are applied at this stage, including the handling of measurements below the instrumental detection limit, the filtering of upper and lower limits for acceptable Aerosol Angstrom Exponent (AAE) values, r^2 score of the spectral dependence, ratios during spot change. The ACTRIS harmonization coefficient (H factor), and filter tape–dependant C-value are also applied, as defined by Savadkoohi et al. (2024) in the frame of the RI-Urbans pan-European overview.

BC Source Apportionment is subsequently conducted on Level1 data using **BCSA_S4**. Data are averaged over a 15-minute time resolution in order to increase the signal-to-noise ratio of BC RT outputs. A configuration file allows to fill out various items, such as general information on the station and the instrument, as well as explicit source apportionment parameters (α-values for liquid fuel and solid fuel fractions). As detailed in D1.4, the “aethalometer model” by Sandradewi et al. (2008) separates liquid and solid fuel combustion based on the fact that these two combustion sources exhibit a different light absorption wavelength dependence. Alpha values for liquid fuel (e.g., traffic exhaust emissions) is typically characterized by values between 0.9 and 1.1, whereas solid fuel is less well-characterized because it depends not only on the combustion material but also on the combustion efficiency and the burning conditions, e.g. values up to 2.2 (Sandradewi et al., 2008) can be found in literature. So for this reason, and given the variability of AAE distributions over several sites, using site-specific α-value has appeared to be essential in order for **BCSA_S4** to provide reliable and comparable results. **BCSA_S4** contains a dedicated script to be used “offline” to determine site-specific values based on statistical distributions and/or external tracers.

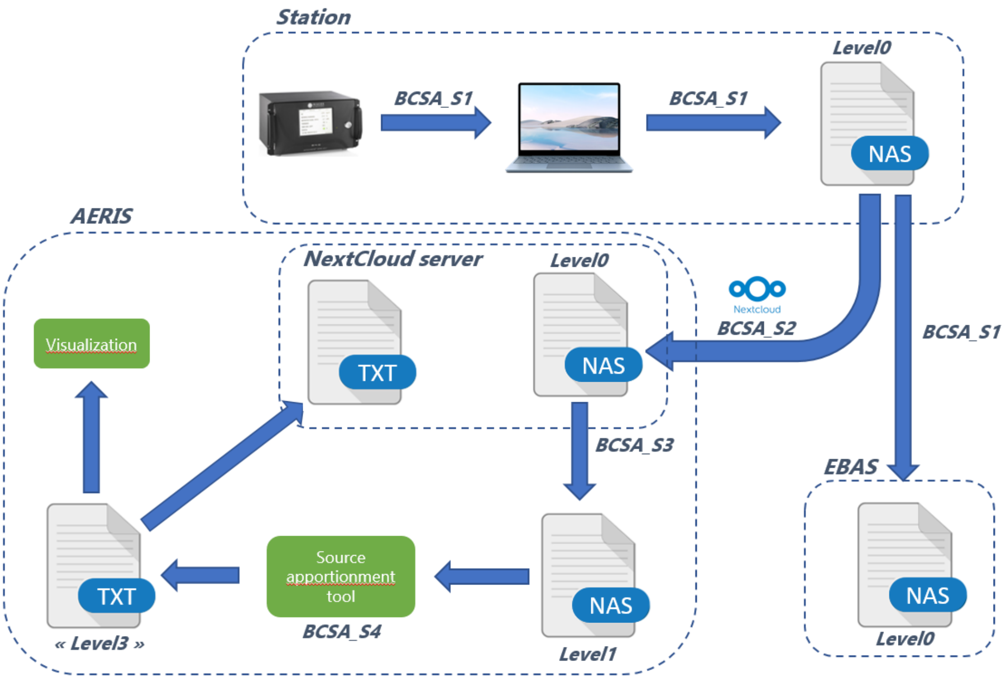

Figure 1. *Dataflow to be used in RI-URBANS for BC NRT source apportionment.*

Eventually, outputs (timeseries) of BC Source apportionment analysis have been stored in text files at the centralized server, where there are also made available automatically for data providers (i.e., pilot sites in WP4.1).

2.2. Organic Aerosols Source Apportionment

For OA, data workflow is illustrated in Figure 2. Data acquisitions are performed by **OASA_S1**, which have been initially elaborated by the instrument manufacturer (Aerodyne Res. Inc., USA). These procedures are based on Igor Pro (Wavemetrics Inc.). Since Quadrupole and Time-of-Flight systems deliver different kind of signals, and data workflows (as well as format) are different, specific tools shall be applied according to the type of mass spectrometer. Because the acquisition is based on Igor Pro, additional procedures needed to be developed in order to export data and generate interpretable files in ASCII format (**OASA_S2**). **OASA_S2** has been developed by ACMCC and runs independently of the acquisition. It is important to note here that input data for OA Source Apportionment are different from regular ACSM data submitted on EBAS for ACTRIS. This means that no ACTRIS template exists, and therefore no Level0/1/2 files of OASA input data can be generated at this stage. As a consequence, **OASA_S2** (as well as **OASA_S1** for ToF systems) have been updated in order to meet the requirements of **OASA_S4**.

Similarly, to BC source apportionment, data transfer to the centralized server has been performed with a cloud service (**OASA_S3**).

OA source apportionment was then achieved using **OASA_S4**, developed by Datalystica. **OASA_S4** is composed of SoFiRT, which is an IgorPro-based module based on the methodology presented in Chen et al. (2022a et 2022b), and the ME-2 solver executable. Both are commercial software tools. The choice of these tools is mainly linked to the fact that they are currently the only tools being able to perform NRT SA of OA, akin therefore to *Best Available Techniques*.

Over the course of the Pilot Phase, close interactions between T1.2 and T4.1 allowed to troubleshoot **OASA_S4** in a very reactive way, leading to sixteen updates of **OASA_S4**. One of the most important update is surely the development of configuration files in order to speed up the configuration of **OASA_S4**, which could require significant human-machine interactions through user-friendly panels. This has been a very valuable achievement in order to start and maintain OA source apportionment over 13 sites during the Pilot Phase. SoFiRT is not natively able to handle multisite analyses at once. Instead, several instances of Igor Pro needed to be used in parallel (which shall be consistent with the number of SoFiRT licenses).

Although raw results of OASA_S4 are stored in h5 files, timeseries of OA fractions are stored within generated text files.

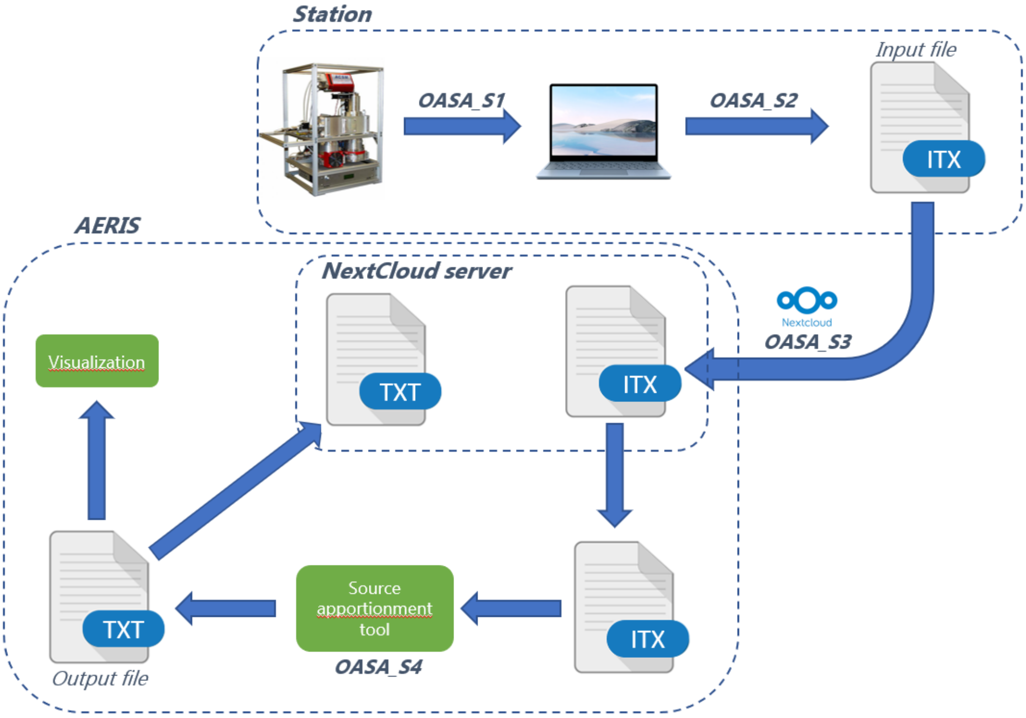

Figure 2: *Dataflow to be used in RI-URBANS for OA NRT source apportionment.*

2.3. Component availability

All the components of both STs - except OASA_S4, as explained above- are open-source et publicly available. The availability of these components is listed in Table 1. Proper guidance documents are available for each component, as well as licensing.

Table 1: *Availability of all components of Source Apportionment STs.*

Service Tool Component	Description	Website
BCSA_S1	Acquisition / Formatting / Transfer	https://gitlab.com/tropos_ecac/ebas_aerosol_nrt/-/tree/master/ *login/password required to access Repository*
BCSA_S2	Data Transfer	https://nextcloud.com/
BCSA_S3	Level0 to Level1	https://git.nilu.no/ebas-data-processing/cais-ecac-processing-software/-/tree/master/ae33

BCSA_S4	Source Apportionment	https://www.icare.univ-lille.fr/depot/ACTRIS/ACMCC/software/ae33_sa_code-v0.1.0.zip
OASA_S1	Data Acquisition	https://aerodyne.com/acsm/ *login/password required to access Aerodyne Knowledge Base*
OASA_S2	Data Export to ASCII files	https://www.icare.univ-lille.fr/depot/ACTRIS/ACMCC/software/actris_acsm_converter-latest.zip
OASA_S3	Data Transfer	https://nextcloud.com/
OASA_S4	Source Apportionment	https://datalystica.com/downloads/ *password required to access files*

3. CONCLUSION

All the developments performed within WP1 T1.2 and presented in this deliverable has allowed WP4 T4.1 to showcase the feasibility of NRT source apportionment of carbonaceous aerosols during the Pilot Phase (2023). Results of these piloting activities will be detailed in D4.3 (M40), demonstrating the potential of the developed STs to be implemented at a larger scale, but further work is intrinsically associated to upscaling, as described in the following sections.

4. FURTHER WORK

4.1 Black Carbon Source Apportionment

The BCSA ST currently generates timeseries in text files. In order to implement the ST on a database (e.g., EBAS) for upscaling, dedicated templates have to be adapted, along with a specific script for the generation of this advanced Level3 data products

Even if the relative simplicity of the “aethalometer model” is a very valuable strength, especially regarding NRT process, the algorithm can suffer from limitations which may increase the uncertainty of the results. In particular, the choice of source-specific alpha values is critical because it directly influences the concentrations of BC_{sf} and BC_{lf} (see Savadkoohi et al., 2023) Statistical approaches, as available in **BCSA_S4**, can restrain the solution space and help to determine site-specific values. But it should not prevent from developing other ways of optimization. Also, because the performance of the “aethalometer model” is limited especially in environments with various sources contributing to brown carbon (e.g., various solid fuel sources), alternative source apportionment may be considered, developed and evaluated. This is very valuable for the RI-URBANS stakeholders responding to the requirements of the new air quality directive.

4.2 Organic Aerosol Source Apportionment

The OASA ST currently generates timeseries in text files. To implement the ST on a database (e.g. EBAS) for upscaling, dedicated templates must be developed, along with a specific script for Level3 data files generation. More importantly, in order to follow FAIR principles, Level3 data must be linked to the input data. This means that the implementation of OASA ST is intrinsically linked to the development, structuration and consolidation of new data streams (Level0/1/2) that are currently not supported by existing tools and databases. These requirements and additional workflows to accommodate OASA ST are discussed and planned in collaboration between the ACMCC and ACTRIS Data Center.

Also, further interactions are needed with potential end-users in order to frame the usefulness of these new data and investigate if adaptations are necessary to best relate OA fractions to pollution sources and/or precursors. To this end, there are no limiting boundaries hindering dedicated efforts in developing open-source alternatives for source apportionment and evaluate their performances and applicability for NRT. Currently ACMCC is exploring options for the open-source alternatives e.g., with Atmospheric Science and Chemistry mEasurement NeTwork (ASCENT) network in the USA. This would allow full compatibility of the data analysis workflow between Europe and the USA.

5. REFERENCES

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