Milestone M34 (M5.6)

Upgrading source apportionment tool

RI-URBANS

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

By

FORTH, INERIS, TNO & METNO

17th September 2024

RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

Milestone M34 (M5.6): Upgrading source apportionment tool

Authors: Maria Kanakidou (FORTH), Spyros Pandis (FORTH), Elena Poulikidi (FORTH), Evangelia Siouti (FORTH), Augustin Colette (INERIS), Florian Couvidat (INERIS), Alicia Gressent (INERIS), Renske Timmermans (TNO), Floris Pekel (TNO), Ruud Janssen (TNO), Janot Tokaya (TNO), Hilde Fagerli (MetNo), Peter Wind (MetNo) and Willem van Caspel (MetNo)

Work package (WP)	WP5 Strategic guidance for upscaling RI-URBANS Service Tools
Milestone	M34 (M5.6)
Lead beneficiary	FORTH
Means of verification	Report on preliminary design developed
Estimated delivery deadline	M36 (30/09/2024)
Actual delivery deadline	17/09/2024
Version	Final
Reviewed by	WP3 leaders and the project coordination team
Accepted by	Project coordination team
Comments	This document summarizes the proposed source modelling methodologies for the source apportionment tools of health relevant indicators (e.g. PM mass, OP, UFP, OA, BC) developed and applied in WP3 in order to be evaluated them before being proposed to CAMS in D5.4 by the end of the project. The source apportionment results will be provided in a user-friendly environment that will enhance citizens' implication, and thus, contribute to citizen's science.

RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

Table of Contents

1. ABOUT THIS DOCUMENT

This document was prepared as part of the "Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS" (RI-URBANS) EU-project that connects the atmospheric observation expertise from Aerosols, Clouds and Trace gases Research InfraStructure (ACTRIS) as well as the urban air quality observation capacities of the regulatory air quality monitoring networks.

The document summarizes the proposed methodologies for the source apportionment tools of health relevant indicators (e.g. PM mass, OP, UFP, OA, BC) developed and applied in WP3 in order to be evaluated them before being proposed to CAMS in D5.4 by the end of the project. The source apportionment results will be provided in a user-friendly environment that will enhance citizens' implication, and thus, contribute to citizen's science.

The document reports on the design of source apportionment methods developed and used within WP3. All of these applications were illustrated in WP3 at both the regional and local modelling scales, with the exception of OA which is primarily a focus for regional scale models.

This public document will be distributed to all RI-URBANS partners for their use and submitted to the European Commission as RI-URBANS milestone M34 (D5.6). It can be downloaded at https://riurbans.eu/work-package-5/#deliverables-wp5.

2. SOURCE APPORTIONMENT TOOLS OF SOURCE MODELLING

Different source apportionment methods used by the air pollution modelling community and within Ri-Urbans. The most common are the zeroing emissions, sensitivity method reducing emissions from sources with e.g. 15% (Pommier et al., 2020) or using a surrogate model (Colette et al., 2022), tagging method (e.g. Kranenburg (2013) which includes the Particulate Source Apportionment Technology (PSAT) developed by Wagstrom et al. (2008)) and the local fraction methodology (Wind et al., 2020). Within RI-URBANS, LOTOS-EUROS and PMCAMx models are using the PSAT methodology, while EMEP and CHIMERE are using sensitivity methods.

PMCAMx was coupled with the modified Particulate Matter Source Apportionment Technology (PSAT) algorithm that performs pollutant mass source apportionment calculations at multiple scales at the same time and can be now used for pollutant mass source apportionment. This methodology is further described in the document as applied for PM2.5 in Athens.

For UFP modelling, an innovative framework for including source apportionment in the PMCAMX-UF model was proposed in order to make use of the UFP source apportionment data. It will be possible for local or national AQMN to implement this approach to develop the uptake of UFP modelling and evaluation in the future. This methodology is described in the document as applied for UFP in Athens.

Several models are using the brute force methodology to evaluate the contribution of the zeroed source to observed pollutant levels. This is a simple methodology that however requires several (*n+1*) simulations in order to attribute pollutants to a number (*n)* of sources. EMEP is using the local fraction (LF) methodology that gives the fraction of pollutants that have a local origin, e.g. originating from emissions from the same grid as the relevant concentrations (Wind et al. 2020) and can be efficiently used to track pollutants from a large number of sources, over large distances. LF method can give the same results as a direct ("brute force") method for small emission changes. The advantage of the LF method is that it can do so at a lower computational cost when a large number of scenarios are to be computed and the scenarios only differ by small changes in emissions. The surrogate model Air Control Toolbox developed within CAMS with the CHIMERE model avoids this difficulty related to the number of scenarios required in the sensitivity approach by fitting a non-linear multivariable model surface capturing the sensitivity of the full chemistry-transport model in an automated learning environment.

For Black Carbon (BC), the source apportionment methodology is mature enough to propose a fair comparison between models and observation, considering the uncertainty of the emission fluxes and observations in deriving equivalent BC (eBC) and quantifying the source apportionment of liquid and solid fuels. A first demonstrator was developed to perform near real time QA/QC of the 11 operational CAMS models retrieved from the Atmosphere Data Store against 11 stations reporting NRT data from the RI-Urbans pilot cities. This demonstrator will be proposed to CAMS for future uptake in an operational Service Tool (ST).

For Organic Aerosol (OA), the methodology proposed in WP3 to compare models with ACSM source apportionment data (OOA, HOA and BBOA) from 31 sites could be tested with the advanced version of 4 different CTMs (CHIMERE, CAMx, EMEP and LOTOS EUROS). While the regular CAMS service provides total OA mass, detailed information of apportioned OA requires sophisticated aerosols modules which are not implemented by all of the eleven operational CAMS Regional CTMs. The OA ST for model evaluation will be focused on the methodological matching between models and measurement. While NRT ACSM data will soon become available there remains substantial uncertainties regarding the correspondence between ACSM source apportionment and emission activity sector. It is only when advanced OA model parameterisation is implemented in a regular service such as CAMS that routine evaluation will be possible.

For the Oxidative Potential (OP), the robustness of the methodology was strengthened by aligning the approach of 3 different modelling teams (CHIMERE, LOTOS-EUROS and EMEP) in implementing the OP data obtained from observation providers in WP2 (IGE and PSI, themselves relying also on data collected by CSIC, EMPA, and NABEL). Although the maturity of the source apportionment was significantly improved, the ST for potential downstream user still requires further research work.

3. POLLUTANT MASS SOURCE APPORTIONMENT - THE EXTENDED PSAT ALGORITHM

The PSAT algorithm has been designed to calculate the source contributions to primary and secondary air pollutants in the particulate phase (Wagstrom et al., 2008; 2011a; 2011b; Skyllakou et al., 2014; 2017; 2021). It runs in parallel with a Chemical Transport Model (CTM) and the source types are defined by the user and the corresponding emissions inputs of each source are provided separately. PSAT has been designed to determine source contributions within a single modeling domain with a fixed spatial resolution. In RI-URBANS the FORTH team has extended PSAT to provide source contributions using multiple grids and moving from the regional to the urban scale with increasing spatial resolution. As a results PSAT can now perform source apportionment calculations at multiple scales at the same time.

The fifteen emission source categories used in the RI-URBANS applications of PMCAMx are: "industry", "fugitives and solvents", "aviation", "agriculture", "combustion", "road-transport (exhaust)", "road-transport (non-exhaust)", "shipping", "non-road transport", "biomass burning", "sea-salt", "biogenic", "wildfires", "initial conditions" and "boundary conditions" which is the long-range transport from outside Europe.

The extended algorithm used the following steps:

Step 1: PSAT is used at the 36x36 km^2 modeling domain over Europe.

Step 2: For each 36x36 km^2 grid cell, the average concentration of each pollutant due to each source is calculated.

Step 3: PSAT is used at the 1x1 km^2 modeling domain over the city of interest. The city occupies a few 36x36 km^2 computational cells in the European domain.

Step 4: For each 1x1 km^2 grid cell, the average concentration of species coming from each local source is calculated.

Step 5: The total transported concentrations in the 1x1 km^2 domain for each source are calculated by subtracting the total local average concentrations from Step 4 from the total 36x36 km^2 average concentrations obtained in Step 2.

Step 6: The results of Steps 4 and 5 are synthesized to provide the contribution of all the sources separated into local and transported contributions for each pollutant of interest.

4. APPLICATION FOR PM2.5 SOURCES IN ATHENS

We applied the PMCAMx/PSAT model over Europe using a 36x36 km^2 spatial resolution during a typical summer (July 2019) and winter (January 2019) month. The modeling domain covers a region of 5400x5832 km^2 (Figure 1a). For each 36x36 km^2 grid cell, PSAT calculated the contribution of each source to $PM_{2.5}$. Additionally, we applied the PMCAMx model in another simulation over Europe, focusing on Athens, using three nested grids with increasing spatial resolutions during the same periods (Figure 1b and c). The inner domain, which is the domain of Athens, had a 1x1 km^2 horizontal spatial resolution and 72 columns and 72 rows. The modeling domain of Athens (72x72 grid cells) corresponds to four 36x36 km^2 grid cells of the European domain. The long-range transport contribution for each 1x1 km^2 grid cell was further separated to its sources, based on the source predictions from the corresponding 36x36 km^2 grid cell without considering the local concentrations from the 1x1 km^2 simulation.

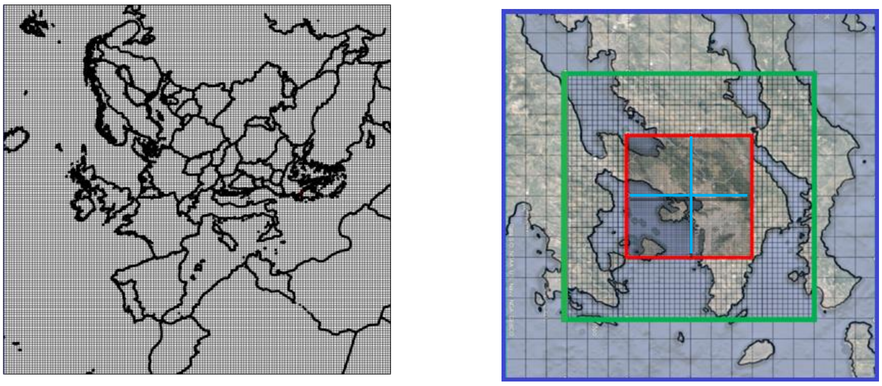

Figure 1. *(a) The European modeling domain used for the first simulation with 36x36 km^2 spatial resolution, (b) the European modeling domain for the second simulation and the three nested grids and (c) the nested domains focusing on the 1x1 km^2 modeling domain of Athens (red). Blue: 1st nested domain with 12x12 km^2 spatial resolution, green: 2nd nested domain with 3x3 km^2 spatial resolution and red the 3rd nested domain with 1x1 km^2 spatial resolution. Light blue inside the red box corresponds to the four 36x36 km^2 Athens' grid cells.*

The summer and winter results of application of the extended PSAT algorithm on $PM_{2.5}$ in Athens are shown in Figure 2. Long range transport dominates in the summer with wildfires and industrial sources away from Athens dominating. Local residential biomass burning is the major wintertime source.All this information is spatially resolved. The corresponding source contribution maps for local sources and total long range transport for July are given in Figure 3.

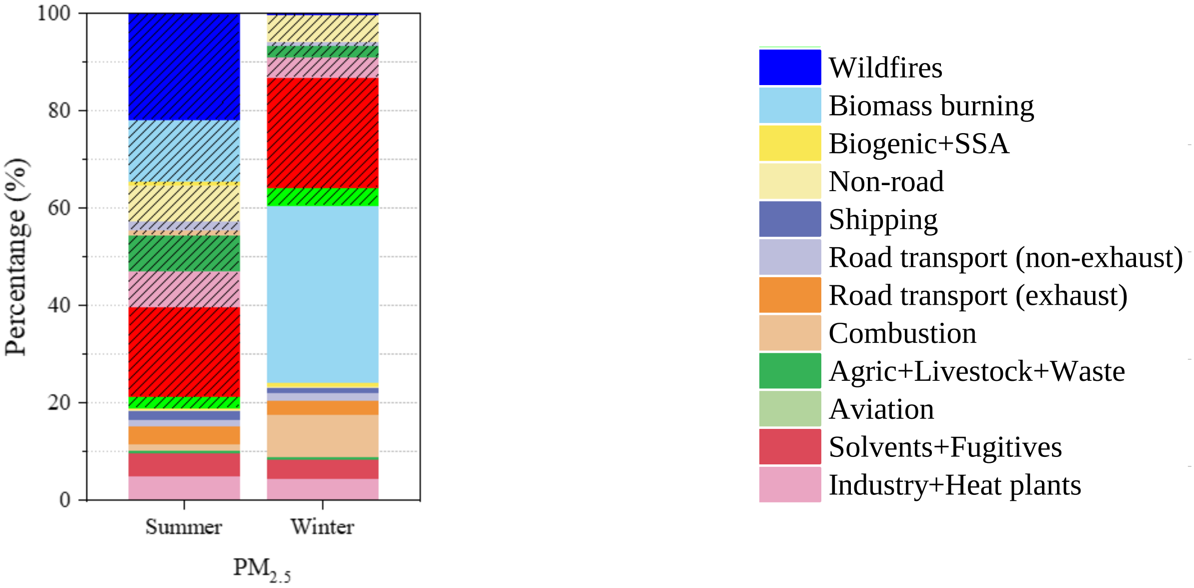

Figure 2. *Average predicted source contributions for $PM_{2.5}$ for the urban center during July and January 2019. Long-range transport is shown with shaded areas.*

RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

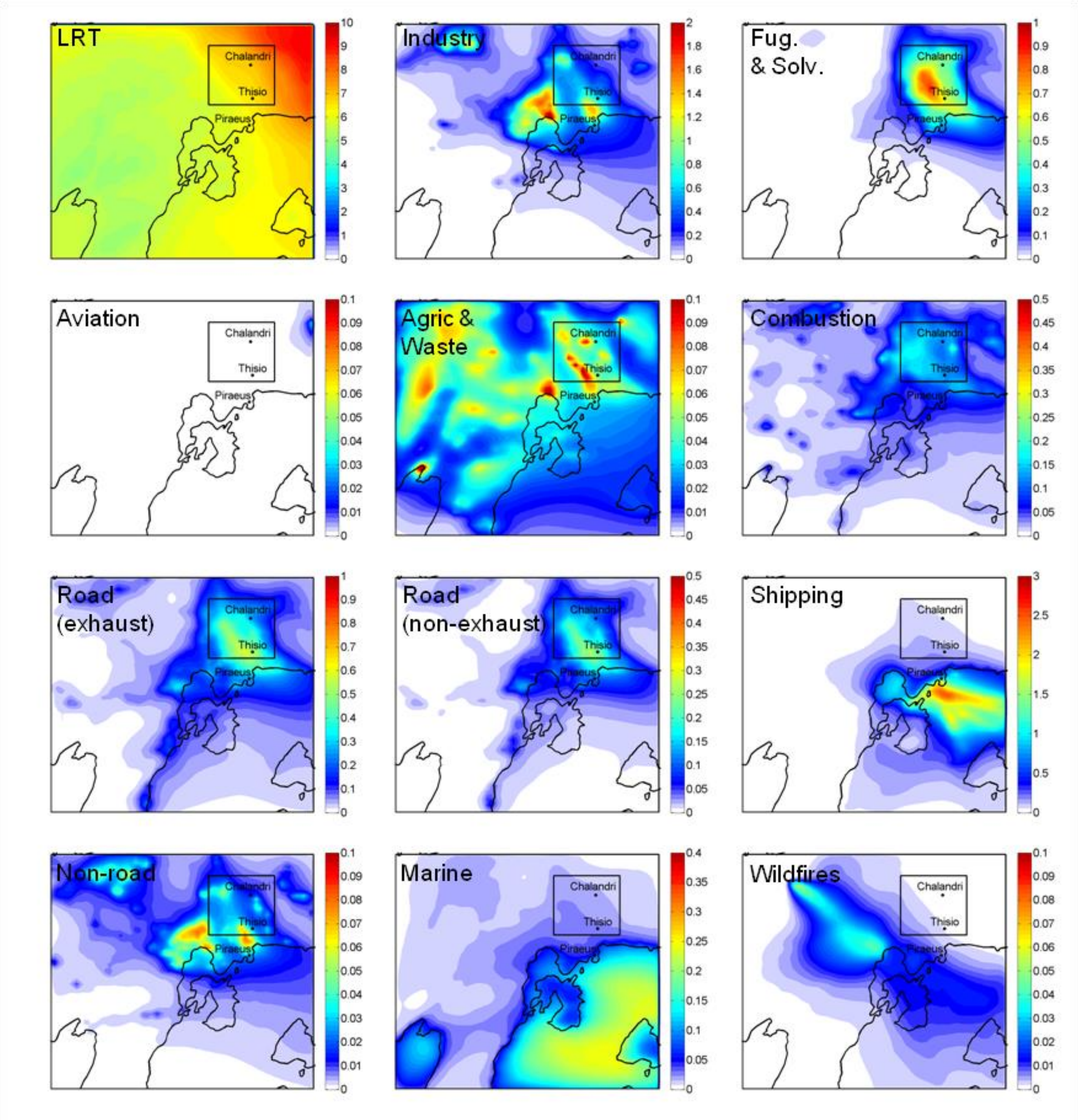

Figure 3. *Average predicted concentrations of PM$_{2.5}$ sources in μg m^{-3} during July 2019 in Athens. Different scales are used.*

RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

5. NUMBER CONCENTRATION SOURCE APPORTIONMENT WITH PMCAMX-UF

PMCAMx-UF is a three-dimensional transport model that simulates independently both the number size distribution and the mass/composition distribution of the multicomponent atmospheric aerosol. Number source apportionment in atmospheric models can be difficult. Eliminating all but one source's number emissions affects greatly the condensation and coagulation sinks, which can result in disproportionate and unpredictable effects on the overall predicted particle number concentrations. For that reason, we have extended the Posner and Pandis (2015) approach for application in Europe in the framework of RI-URBANS. This zero-out method eliminates emissions from specific sources below a certain diameter threshold. This approach allows us to retain most of the surface area and mass of the emitted particles while still capturing the source's contribution to the total number concentrations. The threshold diameter for each source is chosen to eliminate approximately 90% of its number emissions. The sources examined in this work are the following: nucleation, industry including power generation, small combustion, gasoline road transport, diesel road transport, shipping, off-road vehicles, other sources (fugitives, LPG road transport, non-exhaust road transport, waste, agricultural sources), and biomass burning. Table 1 displays the chosen diameters for the RI-URBANS project.

Table 1. *Zero-out diameters used for source apportionment simulations.*

Source Category	**Bin diameter range (nm)**	**% eliminated**
Public power and industry	81-102	88
Other stationary combustion/ Residential wood burning	129-162	89
On-road gasoline	51-65	89
On-road diesel	102-129	90
Shipping	41-51	90
Off-road	81-102	89
Aviation	65-81	90
Others (fugitives, solvents, waste, etc.)	162-205	88

The major extension of the approach is its application for the first time at multiple grid resolutions. The following approach is followed:

1. We apply PMCAMx-UF over Europe with a 36x36 km^2 grid resolution with three additional nested grids. The inner domain which is used to describe the desired city has a 1x1 km^2 grid resolution.
2. We apply PMCAMx-UF over the inner domain with 1x1 km^2 grid resolution.
3. Then, we subtract the results of step 2 from the results of step 1 in order to get the boundary conditions to use for simulating the particle number over the city of interest.

Figure 4 displays the average particle number concentration over Athens for July 2019 and January 2019. For the summer, high number concentrations of particles are predicted over the port of Piraeus and the Saronic gulf. For winter, the higher particle number levels are over the urban core according to PMCAMx-UF. Figure 5 shows the number concentration due to diesel engines. Diesel emissions affect more the city center where most of the traffic is observed. In Figure 6 the diesel percent contribution to particle number is presented. For July 2019, diesel is predicted to contribute approximately 25% to the number concentration for particles with diameter above 10 nm (N_{10}) over the city center of Athens. For January 2019, diesel contributes approximately 40% of N_{10} over the Athens center.

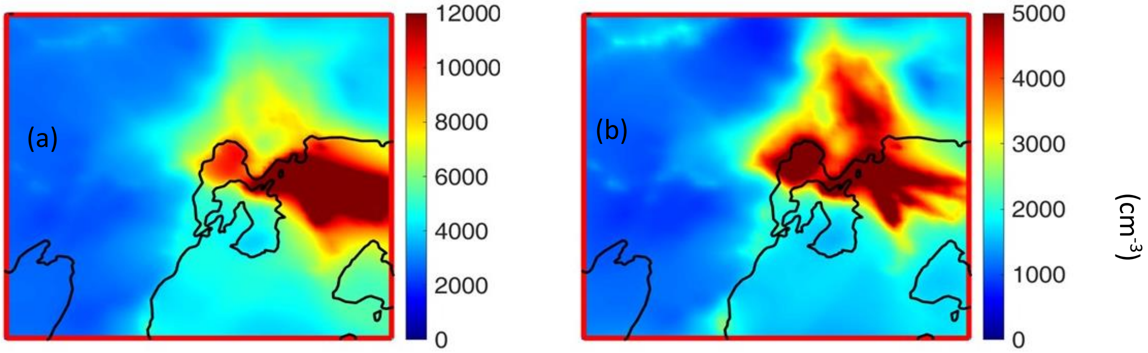

Figure 4. *Predicted ground-level N_{10} concentrations for the city of Athens for (a) July of 2019 and (b) January of 2019.*

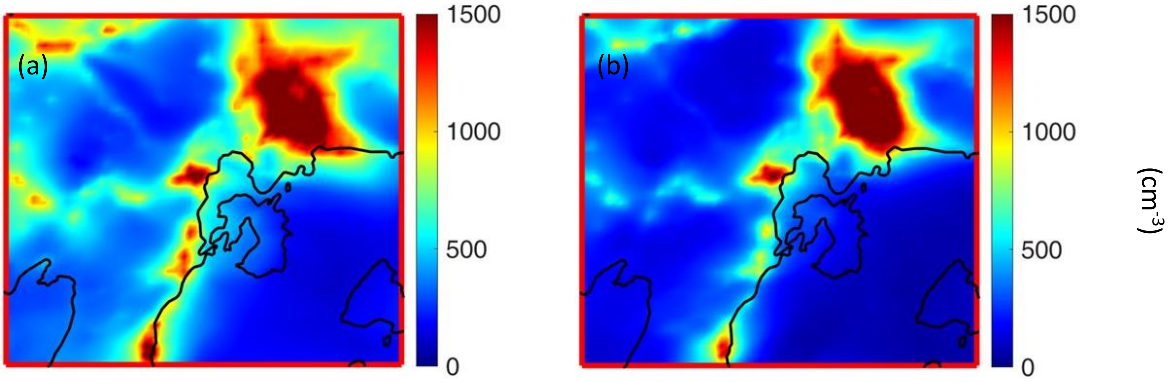

Figure 5. Predicted diesel absolute contribution to N_{10} concentration for the city of Athens for (a) July of 2019 and (b) January of 2019.

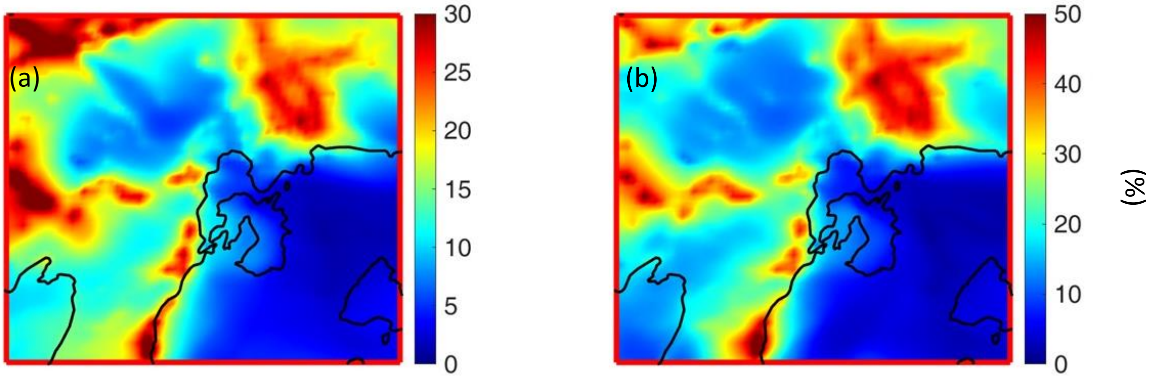

Figure 6. *Predicted diesel contribution in N_{10} concentration as a percentage for the city of Athens for (a) July of 2019 and (b) January of 2019.*

RI-URBANS (www.RIURBANS.eu) is supported by the European Commission under the Horizon 2020 – Research and Innovation Framework Programme, H2020-GD-2020, Grant Agreement number: 101036245

6. SOURCE APPORTIONMENT WITH THE LOCAL FRACTION METHODOLOGY AND APPLICATION FOR OP

The Local Fractions (LF) method was originally developed as a practical method to give the fraction of pollutants that have a local origin, e.g. originating from emissions from the same grid as the relevant concentrations (Wind et al. 2020). It can efficiently track pollutants from a large number of sources, over large distances. Fundamentally the LF method can give the same results as a direct ("brute force") method for small emission changes. The advantage of the LF method is that it can do so at a lower computational cost when a large number of scenarios are to be computed and the scenarios only differ by small changes in emissions. This makes it possible to consider using the LF method as an alternative to the classical brute force (BF) for the calculation of source receptor relationships. In its first formulation, the method could only be applied to inert pollutants. Also, pollutants that have a non-linear dependence on emission intensity could be described, as long as the non-linear dependency can be neglected in a first approximation. However, many important pollutants have a non-linear dependency on emissions. Last year, the LF method was developed to allow for tracking such pollutants through the full complexity of the chemical processes, allowing a description of fundamentally non-linear species such as ozone (EMEP Status Report 1/2023). Recently, all the secondary inorganic aerosols (SIA), $PM_{2.5}$ (and not just the primary fraction of $PM_{2.5}$), wet and dry deposition of pollutants have been included in the LF methodology (EMEP Status Report 1/2024).

In RI-URBANs, the Local Fraction methodology has been used to track primary road traffic PM for each country (and other sources such as e.g. biomass burning emissions). The modelled data from the relevant sources have been combined with intrinsic OP values to generate model results for OP. First results were presented as part of section 2.2.7 of D19 (D3.4). In RP2, the approach has been extended by combining EMEP modelled source apportionment data with TNO road transport emission data (split into brake wear, tyre wear, exhaust, dust) for 1990-2022 to calculate historical exhaust fraction of total road traffic PM and then combine that with the primary HOA fraction from exhaust based on TNO chemical speciation data. Furthermore, the brake wear fraction from total (modeled) road traffic PM is used together with the assumption on the fraction of Fe+Cu. The new results will be presented and discussed as a part of D3.5.

7. BC SOURCE APPORTIONMENT

For BC, the methodology is mature enough to propose a fair comparison between models and observation, considering the uncertainty of the emission fluxes and observations in eBC and quantifying the source apportionment of liquid and solid fuels, using aethalometer wavelength dependent observations as discussed in ST11. While emissions from liquid fuel sources contain predominantly pure eBC dominate at IR wavelengths and exhibit only a weak wavelength dependence, solid fuel combustions contain light absorbing organic substances, show an enhanced absorption in the N-UV range and are strongly wavelength dependent. The "Aethalometer model" (Sandradewi et al., 2008) based on source-specific Absorption Angstrom Exponents (AAE) is a method for separating these two combustion types when only two eBC sources are present.

CAMS Ensemble model performance scores for the average attribution of eBC to fossil fuels for 18 European sites were assessed for the year 2018. Solid fuels are mainly related to the residential sector and considered as the complementary source to traffic for a total eBC. The scores of the ensemble of CAMS models and further details are presented in D19 (D3.4).

The model performance is sensitive to the AAE parameters used for the two types of BC sources. D19 (D3.4) presents the impact of changing AAE from what is suggested by Sandradewi et al. (2008) (AAE of 2 and 1) to the values proposed by Zotter et al. (2017) (1.68 and 0.9). We tested the values of Zotter et al. (2017) on several stations (Figure 7 shows the example of Athens NOA). While the temporal correlation coefficient hardly varies (because the variability of the response does not change), the RMSE score can increase or decrease by a factor of two depending on the station (because the fraction of eBC attributed to traffic changes). This result reinforces the need for robuts observationally driven source apportionment modeling.

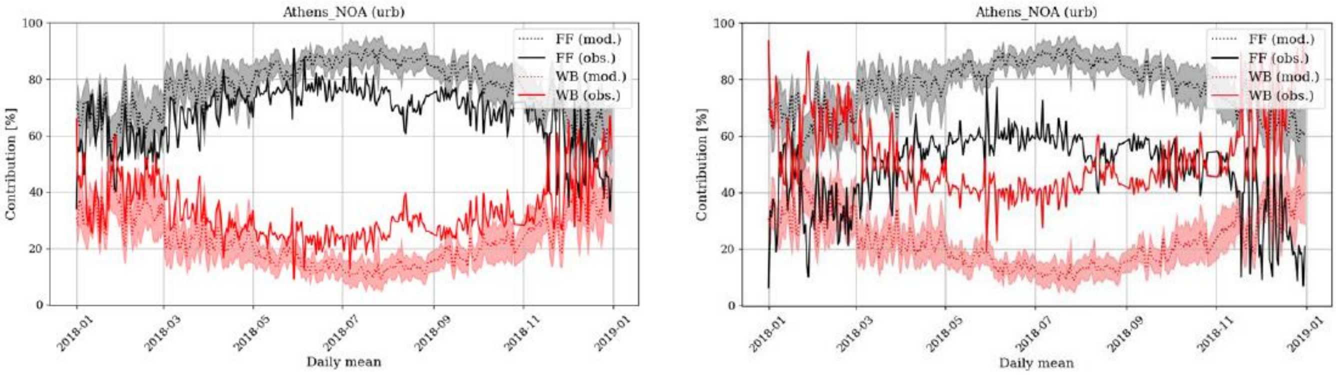

Figure 7. *Source distribution of eBC based on aethalometers using the AAE values of Sandradewi et al. (2008) (left panel) and Zotter et al. (2017) (right panel) and compared with the model ENSEMBLE for the "Athens NOA" station in 2018 (figure from D19 (D3.4)).*

A first demonstrator was developed to perform near real time QA/QC of the 11 operational CAMS models retrieved from the Atmosphere Data Store against 11 stations reporting NRT data from the RI-Urbans pilot cities in 2024. This demonstrator will be proposed to CAMS for future uptake in an operational ST.

8. OA AND OP SOURCE APPORTIONMENT

Both for OA and OP there are significant challenges to provide a common methodology for source apportionment methodologies due to the differences in the way that the models are simulating them and to the correspondence to the observed source apportionment variables, which are themselves weakly constrained. The differences in the way that the models represent OAs as well as the OP leads to significant differences between the models, which is sometimes even larger than that between models and observations.

8.1 Organic aerosol

For OA, the methodology proposed in WP3 to compare models with ACSM source apportionment data (OOA, HOA and BBOA) from 31 sites could be tested with the advanced version of 4 different CTMs (CHIMERE, CAMx, EMEP and LOTOS EUROS). The provision of apportioned OA is not currently implemented in the regular CAMS service. Table 3 in D19 (D3.4) compares the main features of the different RI-Urban models with regard to the treatment of different OA types and reveals significant differences. Therefore, the OA ST for model evaluation will be focused on the methodological matching between models and measurement. While NRT ACSM data will soon become available, strong constrain on the source apportionment variables (OOA, HOA and BBOA) are required, and it is only when advanced OA model parameterisation is implemented in a regular service such as CAMS that routine evaluation will be possible.

8.2 Oxidative Potential

As for OA, the OP simulations among models differ significantly. For OPo, the robustness of the methodology was strengthened by aligning the approach of 3 different modelling teams (CHIMERE, LOTOS-EUROS and EMEP). The modelled source apportionment was explored by implementing the OP data obtained from 2 observation providers in WP2 (IGE and PSI, themselves relying also on data collected by CSIC, EMPA, and NABEL). The description of local fraction and its application to OP in the EMEP model has been presented in section 3. The comparison of model parameterisations and results and thorough discussion can be also found in section 2.2.7 of D19 (D3.4). Although the maturity of the source apportionment was significantly improved, the matching with observed OP which is a critical step to propose a ST for potential downstream user still requires further research work.

9. REFERENCES

Colette, A., Rouïl, L., Meleux, F., Lemaire, V., & Raux, B. (2022). Air Control Toolbox (ACT_v1. 0): a flexible surrogate model to explore mitigation scenarios in air quality forecasts. *Geoscientific Model Development*, *15*(4), 1441-1465.

EMEP Status Report 1/2023, ISSN 1504-6192 (on line) Aug. 2023

EMEP Status Report 1/2024, ISSN 1504-6192 (on-line) Aug. 2024

Fountoukis, C., Riipinen, I., Van Der Gon, H. D., Charalampidis, P. E., Pilinis, C., Wiedensohler, A., O'Dowd, C., Putaud, J. P., Moerman, M., & Pandis, S. N. (2012). Simulating ultrafine particle formation in Europe using a regional CTM: contribution of primary emissions versus secondary formation to aerosol number concentrations. Atmospheric Chemistry and Physics, 12, 8663– 8677.

Gaydos, T. M., R. Pinder, B. Koo, K. M. Fahey, G. Yarwood, and S. N. Pandis (2007), Development and application of a three-dimensional aerosol chemical transport model, PMCAMx, Atmos. Environ., 41, 2594– 2611.

Jung, J. G., Fountoukis, C., Adams, P. J., & Pandis, S. N. (2010). Simulation of in situ ultrafine particle formation in the eastern United States using PMCAMx-UF. Journal of Geophysical Research, 115.

Karydis, V. A., A. P. Tsimpidi, and S. N. Pandis (2007), Evaluation of a three-dimensional chemical transport model (PMCAMx) in the eastern United States for all four seasons, J. Geophys. Res., 112, D14211.

Kranenburg, R., Segers, A.J.J., Hendriks, C., Schaap, M. (2013). Source apportionment using LOTOS-EUROS: module description and evaluation. Geosci. Model Dev. 6, 721–733. https://doi.org/10.5194/gmd-6-721-2013

Patoulias, D., Fountoukis, C., Riipinen, I., & Pandis, S. N. (2015). The role of organic condensation on ultrafine particle growth during nucleation events. Atmospheric Chemistry and Physics, 15(11), 6337–6350.

Pommier, M., Fagerli, H., Schulz, M., Valdebenito, A., Kranenburg, R., & Schaap, M. (2020). Prediction of source contributions to urban background PM 10 concentrations in European cities: a case study for an episode in December 2016 using EMEP/MSC-W rv4. 15 and LOTOS-EUROS v2. 0–Part 1: The country contributions. *Geoscientific Model Development*, *13*(4), 1787-1807.

Posner, L. N., & Pandis, S. N. (2015b). Sources of ultrafine particles in the Eastern United States. Atmospheric Environment, 111, 103–112.

Sandradewi, J., Prévôt, A. S., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E. & Baltensperger, U. R. S. (2008) Using aerosol light absorption measurements for the quantitative determination of wood burning and traffic emission contributions to particulate matter. Environmental science & technology, 42(9), 3316-3323, https://doi.org/10.1021/es702253m.

Wagstrom, K. M., Pandis, S. N., Yarwood, G., Wilson, G. M., & Morris, R. E. (2008). Development and application of a computationally efficient particulate matter apportionment algorithm in a three-dimensional chemical transport model. Atmospheric Environment, 42(22), 5650-5659.

Wind, P., Rolstad Denby, B. and Gauss, M. (2020) Local fractions -a method for the calculation of local source contributions to air pollution, illustrated by examples using the EMEP MSC-W model rv4_33 Geoscientific Model Development, 13 (3), 1623-1634, https://doi.org/10.5194/gmd-13-1623-2020.

Zotter, P., Herich, H., Gysel, M., El-Haddad, I., Zhang, Y., Močnik, G., Hüglin, C., Baltensperger, U., Szidat, S., and Prévôt, A. S. H. (2017) Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol, Atmos. Chem. Phys., 17, 4229–4249, https://doi.org/10.5194/acp-17-4229-2017.