Milestone M4 (M1.4)

Existing single pollutant SA at selected EU urban background sites

RI-URBANS

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By

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Work package (WP)	WP1 Novel AQ metrics and advanced source apportionment STs for PM, and nanoparticles
Milestone	M4 (M1.4)
Lead beneficiary	CSIC
Means of verification	Existing and new source apportionment (SA) at selected EU supersites. Source apportionment implemented for EU urban supersites with data available for this type of studies. New SA done, including multitime SA. Data sets made available.
Estimated delivery deadline	M28 (31/01/2024)
Actual delivery deadline	31/01/2024
Version	Final
Reviewed by	WP1 leaders
Accepted by	Project Coordination Team
Comments	This report offers a literature review of source apportionment studies at several EU urban background sites.

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1. About this document

In WP1, T1.2 focuses on developing and implementing advanced source apportionment STs (M02-M36). This task aims at providing STs, based on best procedures and methodologies, to apportion novel health-related AQ metrics. Have evaluated and applied the most suited source apportionment receptor models for operational applications, taking into account previous work in FAIRMODE, EMEP and COLOSSAL (COST Action: Chemical On-Line cOmpoSition and Source Apportionment of fine aerosoL). State-of-the-art source apportionment methodologies have been applied on nanoparticle PNSD, online and offline PM chemical speciation, BC and VOCs datasets from T1.1 to provide the source contributions to each of these in a harmonized way. This offered a pan-European centralized source apportionment database (D1.3) required for compiling new exposure indicators for health effect studies (WP2) and datasets for inter-comparison with chemical transport models (WP3). This task is providing recommendations on best procedures and methodologies to apportion novel AQ variables.

This report offers a literature review of source apportionment studies at several EU urban background sites for atmospheric particulate matter (PM) and emerging air quality metrics such as off-line PM chemistry, non-refractory PM, equivalent Black Carbon (eBC), Particle Number Size Distribution (PNSD), on-line elements and off-line Volatile Organic Compounds (VOCs). Based on such review, main conclusions on source contributions at the European scale are drawn.

This is a public document that will be distributed to all RI-URBANS partners for their use and submitted to European Commission as a RI-URBANS Milestone M4 (M1.4). This document can be downloaded at https://riurbans.eu/work-package-1/#milestones-wp1.

2. Off-line PM chemistry

A review of the studies of source apportionment studies of PM10 and PM2.5 using off-line chemical characterization carried out since 2009 in European urban areas has been conducted updating the review carried out by Hopke et al. (2020). Among the 106 European urban studies 85 presented PM10 data, and 56 presented PM2.5 data (32 combined PM10 and PM2.5).

The 87% of these studies were performed with positive matrix factorization (PMF), 6% with chemical mass balance (CMB), 5% with principal component analysis (PCA) with and without multiple linear regression (PCA-MLR), and the others with UNMIX.

In most studies the major sources/factors of PM were found to be secondary inorganic aerosols (SIA), traffic, dust, sea salt, biomass burning, industry, coal or oil combustion. Other sources, such as biogenic SOA and anthropogenic SOA, which require molecular tracers for identification (Amato et al., 2024) have been found only in a small proportion of these studies. In some cases, there is a source that is not identified, so it is labelled by the main tracer (as in Visser et al., 2015). A summary of the results could be found in Figure 1. The mean values can be found in Table 1 (as %) and Table 2 (in μg m^{-3}), while Tables 3 and 4 present these values grouped according to the region where the study was conducted.

Averaging sources contributions to PM10 among all sites, 27% was attributed to mixed SIA, 18% to traffic, 18% to dust, 16% to nitrates,15% to sulphates, 13% to sea salt, 13% to biomass burning, 8% to industry and 8% to coal or oil combustion and 10% to other sources (mixed factors, sometimes well identified, sometimes not, and also the

unaccounted mass), but these proportions might change widely according the sites. From the averages of sources contributions to PM2.5, 32% was attributed to mixed SIA, 25% to sulphate, 24% to traffic, 17% to biomass burning, 16% to nitrate, 13% to coal or oil combustion, 11% to dust, 9% to sea salt, 7% to industry and 11% to "Other sources". In case of mixed sources (e.g. traffic + industry) these were included in the "other sources" category. It is important to note that part of the secondary organic and inorganic aerosols might have the origin in the emissions of precursors from road traffic and industrial. This is a very important limitation of the source receptor modelling: Inorganic and organic secondary organic aerosols are two type of atmospheric PM formation processes, but not sources. To disentangle the source contribution to these direct modelling is required by using chemical models.

Secondary inorganic aerosols result from the atmospheric oxidation of inorganic gaseous precursors to the particulate phase. Secondary inorganic aerosol (SIA) include three type of factors: nitrate, sulphate and/or a mixed SIA. In PM10, 22% of the studies identified a mixed source of SIA, of which only two studies additionally found a nitrate or sulphate source. Instead, 49% of studies distinguished between nitrate and sulphate factors, 11% identified only the sulphate source, and 8.5% found no SIA-related source. In PM2.5, 30% determined a mixed source of SIA, of which 5% additionally identified the nitrate source. 37% distinguished between nitrate and sulphate, 12% identified only the sulphate source, and 16% found no SIA-related sources. In the studies which include mixed SIA, this factor contributed on average the 28% of PM10 and 31% PM2.5. In PM2.5, the contribution of sulphate (25%) was on average higher than nitrate (16%), while in PM10 those means were similar (16%). Sulphates and nitrates are formed through the oxidation of SO_2 and NO_2, respectively. During the summer, sulphate concentrations tend to peak as a result of increased photochemical activity, being up to 6 times higher than during winter due to higher photochemistry. As residential heating usage and vehicular traffic increase in winter, nitrates may increase due to increased nitrogen oxide (NOx) emissions. In addition, winter meteorological conditions, such as reduced pollutant dispersion due to atmospheric stability, can contribute to an accumulation of nitrates in the atmosphere. However, it must be considered that not all studies cover a whole year. It is important to notice that particulate ammonium nitrate, under warm conditions, is dissociated into gaseous NH_3 and HNO_3. Thus, in many regions of Europe PM nitrate concentrations are lower in summer and higher in winter due to this instability in warm conditions. This account, in many sites, for an opposite seasonal pattern of sulphate and nitrate, but this is more related with the faster SO_2 summer oxidation and nitrate instability. It is also very relevant to note, that in many cases, the SIA profiles contain a relevant contribution from OC (organic carbon) due to the fact that both SIA and secondary organic aerosols (SOA) tend to increase under similar atmospheric conditions. There is a need to use organic tracers in the receptor modelling to quantify SOA contributions.

Secondary organic aerosols (SOA) is a relevant contributing source to PM concentrations, but this source is mostly identified and the contribution apportioned only when organic speciation of PM is available and the specific carboxylic tracers are analysed. This can be both generated from biogenic and anthropogenic volatile organic compounds (VOVs) and the contributions tend to be higher in summer due to higher biogenic VOCs and higher photochemistry. If no organic speciation is available the OC attributable to this SOA contribution is apportioned among other sources such as SIA and road traffic. Se below organic tracers for SOA.
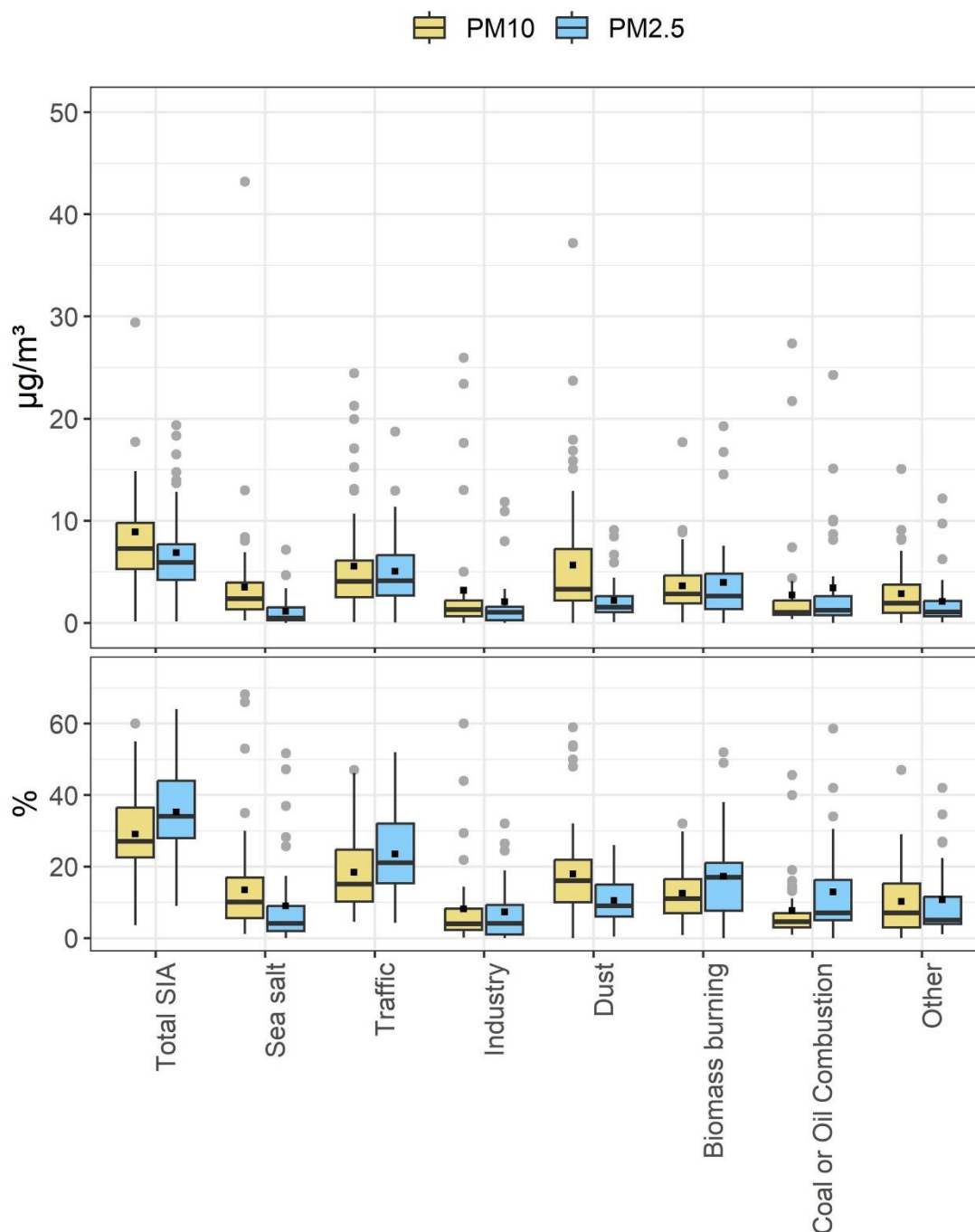
Figure 1. *Variations of source absolute (top) and relative (bottom) contributions of urban European studies from 2010 to 2020. Boxplot represents the median value (line inside the box), the mean value (black square), the Q1 and Q3 (lower and upper boundary of the box, respectively), the maximum and minimum (vertical line) and the outliers (grey dots).*

Road Traffic sources encompass a wide range of emissions, including primary PM from exhausts and non-exhaust (including road dust resuspension) and secondary aerosols mostly from exhaust gaseous organic and inorganic precursors. Vehicle exhaust primarily contains carbon compounds (OC, EC), while non-exhaust sources are traced by trace elements (Ba, Cu, Mn, Pb, Zn) and mineral species (Al, Ca, Fe, Mn, Si, Ti) (Viana et al., 2008). Typically, traffic factors are combined in one source, but sometimes exhaust and non-exhaust are reported separately (Hopke

et al., 2020). Sometimes this source is mixed with other anthropogenic contributions, such as industry (Samek et al.,2019; Salameh et al., 2018). The 91% of the studies analysed the PM10 fraction (mean contribution of 18%), and 84% of the studies the PM2.5 (mean contribution of 24%) included at least one traffic-related source. The regions with major contributions of this source are Southern Europe (with an average of 24% in PM10 and 26% in PM2.5) and Eastern Europe (19% and 26%) while the Western (15% and 21%) and Northern (11 and 13%) regions presented on average lower values (Table 3). The contribution of road traffic tends to be underestimated by receptor modelling because a relevant proportion of SIA (nitrate) and SOA, might arise from the oxidation of primary traffic gaseous pollutants, and these are attributed to SIA or SOA contributions, and not to road traffic. There are organic tracers, such as hopene that can help to include more organic pollutants to this traffic contribution.

Mineral dust is identified either as a natural and/or anthropogenic source traced by Al, Fe, Mn, Si, and Ti, or Ca/Ca^{2+} (Amato et al., 2024) among others. In some studies, some trace elements (Cr, V, Pb, Cu, Sb, Zn, Fe) are enriched in the source profiles, due to the impact of the anthropogenic emissions such as traffic (road dust) or industry (Contini et al., 2016; Samek et al., 2018). It is also frequent to include in this factor the contribution of desert dust (Amato et al., 2016). 87% of the studies of PM10 identified this factor, with a mean contribution of 18%, and 80% of the studies of PM2.5, with a mean contribution of 11%. In PM10 important differences in contribution by regions are not observed (19, 18, 16 and 12% in Southern, Eastern, Northern, and Western Europe, respectively). In PM2.5, these differences are more pronounced (17, 8.8, 8.4 and 3.7 for Eastern, Northern, Southern and Western Europe, respectively).

Biomass burning contributions are generally related to residential heating and/or agricultural burning. In some areas with a mix of biomass and coal in residential heating these sources are included in one factor (Hopke et al., 2020). The main tracers for the identification of this factor are the high concentration of K and levoglucosan, galactosan and manosan, which are crucial in many cases to identify and apportion this contribution (Amato et al., 2024). Because in PM10 a relevant fraction of K is due to mineral dust (clay minerals and K-feldspars), it is very important to include organic tracers in the receptor modelling of this PM fraction. The biomass burning source exhibits seasonality, with peaks occurring in winter, primarily attributed to emissions from fireplaces and woodstoves used for residential heating (Almeida et al., 2020). 77% of the studies of PM10 included this factor, with a mean contribution of 13%, and 77% of the studies of PM2.5, with a mean contribution of 17%. On average, the relative contribution is significantly lower in Northern Europe (Table 1) for both PM10 and PM2.5 (2.2% and 9.0%, respectively). However, looking at the concentration, the highest levels were detected in Eastern Europe (5.9 µg m^{-3} and 6.5 µg m^{-3}, respectively).

Coal or oil Combustion sources encompass fossil fuel, coal or heavy oil combustion (Hopke et al., 2020) from residential and industrial activities. Coal is generally traced by Cl, As and Se, and Oil combustion is traced by V and Ni. Given the inclusion of residential heating, contributions during winter are higher compared to those observed in other seasons. Some of the studies identified this source a shipping-related heavy oil combustion (Kfoury et al., 2016; Salameh et al., 2018). As expected, contributions of this factor are markedly higher in Eastern Europe for PM10 (42%, more than 5 times higher than the next value) and PM2.5 (34%, 2 times higher than the next value). In this region, efforts have been made to reduce the extent of coal-burning in order to reduce both air pollutants and greenhouse gas emission (Hopke,2020). If petrochemical PM emissions are expected to contribute to increase Pm levels, it is also relevant to measure concentrations of La and Ce. Usually La has a mineral origin with the mass Ce/La tending to be close to 2. If relevant petrochemical emissions affect PM levels, the emissions from La increase (La is used in petrochemical processes) and Ce/La decreases a lot.

Industry is a source with high variability in chemical profiles depending of the specific industrial processes taking place in a given region, but some common tracers are Pb, Ni, Br, Zn, Mn, Cd and As (Amato et al., 2024) for cement, smelting, among others, but Zn, Pb, As, Tl, Se, Cd have been identified as tracers of ceramic industries and V, Ni and

La from petrochemical emissions, by other studies. It generally includes a mix of contributions from different industrial processes, although some studies separated a specific industrial process (Kfoury et al., 2016; Koukoulakis et al., 2019). Some studies identified a mixed source including industrial processes and secondary sulphate, coal and fuel oil combustion, or traffic-related (Almeida et al., 2020). 49% of the studies analysed the PM10 (mean contribution of 8.1%), and 43% of the studies the PM2.5 (mean contribution of 7.3%). Eastern Europe was the region with the highest contributions from this source on average (Table 1, 12% in PM10 and 18% in PM2.5).

Fresh sea salt is traced either by ions (Na^+, Cl^-, NO_3^-, Mg^{2+}) and elements (Na, Cl, Mg, S). Some studies differentiate between fresh and aged sea salt (Maenhaut et al., 2016; Weber et al., 2019; Amato et al., 2024). If the mineral contribution is high, Mg may be associated to mineral matter in PM10. **Aged sea salt** is identified by the displacement of Cl^- by NO_3^- and/or SO_4^{2-} (Hopke et al., 2020). In some cases, this factor includes other contributions such as road salt (Amato et al., 2016; Pandolfi et al., 2020). 88% of the studies of PM10 included this factor (mean contribution of 13%), with 32% distinguishing between aged and fresh salt. Similarly, 64% of the studies of PM2.5 (mean contribution of 9.0%) included this factor, some of them differentiating between sources in fresh and aged salt. On average, the contribution is higher in Northern Europe for both PM10 and PM2.5 (Table 3).

The "Others" category encompasses all identified sources that do not fit into any of the previous categories, along with the unaccounted mass. Shipping emissions (traced by V and Ni) are sometimes classified as a source (Manousakas et al., 2017), while others are included in other factors such as sea salt (Saraga et al., 2019) or an oil source (Kfoury et al., 2019). In PM10, 63% of the studies incorporate at least one source characterized by the aforementioned features, with a mean contribution of 10%. Similarly, in PM2.5, 54% of the studies include one of these sources with an average contribution of 11%.

Table 1. *Relative source contributions (%) in European urban areas for PM10 and PM2.5 by regions.*

Source	PM10			PM2.5		
	Mean	SD	N (%)	Mean	SD	N (%)
Total SIA	29	12	91	35	15	84
Mixed SIA	28	9.4	22	31	12	30
Nitrate	16	8.7	60	16	14	38
Sulfate	15	7.4	71	25	10	55
Sea salt	13	12.8	88	9.0	13	64
Traffic	18	10.7	91	24	11	84
Industry	8.1	11.9	49	7.3	9.1	43
Dust	18	12	87	11	6.4	80
Biomass burning	13	7.7	77	17	11	77
Coal or Oil Combustion	7.7	9.3	50	13	13	59
Other	10	9.3	63	11	10	54

Table 2. *Absolute source contributions (μg m^{-3}) in European urban areas for PM10 and PM2.5.*

Source	PM10			PM2.5		
	Mean	SD	N (%)	Mean	SD	N (%)
Total SIA	8.9	9.8	91	6.9	4.4	82
Mixed SIA	8.9	4.2	22	6.3	3.1	29
Nitrate	5.3	8.2	60	3.8	4.3	38
Sulfate	4.3	3.6	71	4.5	2.5	54
Sea salt	3.5	5.2	88	1.1	1.5	63
Traffic	5.5	4.9	91	5.0	3.8	82
Industry	3.2	6.0	49	2.1	3.3	43
Dust	5.6	6.0	87	2.2	2.0	80
Biomass burning	3.6	2.9	77	3.9	4.2	75
Coal or Oil Combustion	2.7	5.2	50	3.4	5.3	57
Other	2.8	2.8	63	2.1	2.8	52

Table 3. *Relative source contributions (%) in European urban areas for PM10 and PM2.5 by regions, with # indicating the number of studies included.*

Size	Source	Eastern Europe		Northern Europe		Southern Europe		Western Europe	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
PM10	Total SIA	49	9.2	17	15	26	8.3	34	11
	Mixed SIA	42	-	25		24	6.5	30	11
	Nitrate	12	-	28		13	6.2	18	9.7
	Sulfate	43	-	8.6	4.6	15	6.3	16	6.7
	Sea salt	8.0	-	45	25	9.1	5.9	13	8.8
	Traffic	19	18	11	8.2	24	9.7	15	9.0
	Industry	12	15	0.85	1.1	11	15	4.6	3.4
	Dust	18	9.5	16	6.0	19	10	12	6.1
	Biomass burning	14		2.2	1.3	14	8.2	13	7.3
	Coal or Oil Combustion	43	3.9	3.4	3.5	6.3	4.7	6.0	5.6
	Other	9.3	4.9	7.1	9.8	6.7	7.5	13	10
PM2.5	Total SIA	39	22	27	18	34	12	48	17
	Mixed SIA	31	13	29	28	32	8.9	-	-
	Nitrate	19	15	37	-	12	11	37	18
	Sulfate	-	-	18	7.1	25	9.7	23	9.0
	Sea salt	27	1.8	36	20	4.7	4.8	4.5	1.5
	Traffic	26	14	13	10	26	10	21	8.3
	Industry	18	14	4.7	9.4	5.1	3.7	1.3	0.35
	Dust	17	7.4	8.8	4.7	8.4	4.7	3.7	4.7
	Biomass burning	22	19	9.0	10	17	8.9	18	7.8
	Coal or Oil Combustion	34	13	12	15	5.5	2.4	12	9.5
	Other	25	18	14	11	7.8	7.3	-	-

Table 4. *Absolute source contributions (µg m^{-3}) in European urban areas for PM10 and PM2.5 by regions, with # indicating the number of studies included.*

Size	Source	Eastern Europe		Northern Europe		Southern Europe		Western Europe	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
PM10	Total SIA	16	2.1	18	37	7.6	3.1	8.7	4.7
	Mixed SIA	18	-	4.4	-	8.6	2.8	8.7	4.9
	Nitrate	3.2	-	58	-	3.7	2.4	4.6	3.0
	Sulfate	12	-	6.6	13	3.9	1.5	3.8	1.7
	Sea salt	2.2	-	10	18	2.8	2.7	3.2	2.1
	Traffic	9.3	10	3.9	7.4	7.4	4.4	4.1	3.7
	Industry	6.8	9.4	1.3	2.5	4.3	7.3	1.1	0.8
	Dust	8.0	4.7	8.5	16	6.0	4.8	3.1	2.1
	Biomass burning	5.9	-	1.8	3.5	4.8	4.1	3.2	1.8
	Coal or Oil Combustion	25	4.0	1.5	0.70	1.8	1.7	1.4	1.3
	Other	4.2	3.4	3.9	6.5	2.0	2.4	3.0	2.0
PM2.5	Total SIA	8.9	3.9	2.0	3.1	7.3	4.2	10.5	6.8
	Mixed SIA	7.4	3.6	1.9	1.5	6.4	1.9	-	-
	Nitrate	3.3	2.6	5.8	-	3.0	4.0	9.0	7.8
	Sulfate	-		0.66	1.0	5.2	2.4	4.5	1.0
	Sea salt	5.9	1.8	0.57	0.37	0.97	1.0	0.91	0.36
	Traffic	7.7	5.5	0.66	0.77	5.7	3.1	4.8	3.5
	Industry	6.4	5.4	0.29	0.57	1.2	0.92	0.29	0.20
	Dust	5.0	2.7	0.21	0.15	1.8	1.2	0.71	0.88
	Biomass burning	6.5	6.7	0.67	1.1	4.1	3.5	3.1	1.8
	Coal or Oil Combustion	12	6.4	1.1	1.4	0.93	0.55	2.0	1.7
	Other	8.1	5.1	1.3	1.7	1.4	1.3	-	-

The use of organic speciation is necessary to detect the source contribution of specific sources. The ideal scenario is using the combination of both inorganic and organic tracers, but increasing the list of tracers require a marked increase of the samples to obtain data matrices that are adequate for multivariate receptor models. In many cases studies are based only on inorganic or in organic speciation. The organic tracers used for source apportionment studies are (van Drooge et al., 2016, 2018):

Biomass burning: Levoglucosan, galactosan and manosan, with the ratios among these allowing to deduce the type of biomass being burned. Some PAHs might help also to identify this source and obtain the contribution to PM.

Road traffic: Hopanes and other tracers (see below).

Primary organic aerosol (POA): polycyclic aromatic hidrocarbons (PAHs), hopanes and anhidrosugars. PAHs are primary products of incomplete combustion of fossil fuel and biomass. The ratio between PAHs provides information about the source (Tobiszeski and Namiesnik, 2012). Hopanes are molecular markers for mineral oils. They are related to unburned lubricating oil residues from traffic, although they are also emitted during coal combustion (van Drooge et al., 2018). Anhydrosugars (levoglucosan, mannosan and galactosan) are monosaccharide anhydriudes generated by thermal alteration of cellulose and hemicellulose. They are extensively used as tracers of biomass burning, and their ratios may also provide information about the emission source.

Soil: Primary saccharides and polyols (mannitol, glucose, arabitol) are constituents of plant tissues and microorganisms that are related to soil content and biological dust resuspension (Medeiros and Simoneit, 2007).

Secondary organic aerosols: Dicarboxylic acids are related to the SOA formation, primarily formed by photochemical alteration of VOCs, although they can also be emitted from a variety of primary sources (such as traffic exhaust fumes or meat cooking). SOA sources are generally traced by the products of isoprene and α-pinene (van Drooge et al., 2018).

Only the 19% of the source apportionment analysis identified primary and secondary biogenic sources (POA and SOA). Weber et al. (2019) identified these sources in PM10 at 15 urban locations in France between 2012 and 2016. They include in the analysis major water-soluble inorganic contents, methanelsulfonic acid (MSA), many metals or trace elements and various organic molecular markers (including levoglucosan, mannosan, arabitol, sorbitol, and mannitol). They identified primary biogenic particles, traced by polyols, and secondary organic aerosols (SOA) from marine biogenic emissions (marine SOA), traced by MSA. These factors represent represent on average 1.2 ± 0.5 µg m^{-3} (7 ± 3%) and 0.6 ± 0.2 µg m^{-3} (4 ± 1%). van Pinxteren et al. (2016) included levoglucosan, arabitol, C22–C34 n-alkanes and a selection of PAHs and hopanes in the analysis carried out in Germany. They identified a source of spores traced by arabitol, which represented 1.2% of the PM10 fraction. Organic molecular markers traced other sources such as traffic exhaust (hopanes, n-alkanes), coal combustion (PAHs), coal combustion (hopanes), biomass burning (levoglucosan), photochemistry (oxalate) and cooking (odd n-alkanes). In a source apportionment analysis of PM2.5 in London, Yin et al. (2015) include organic markers (n-alkanes C24–C35, hopanes, PAHs, sterols (cholesterol and levoglucosan), fatty acids and secondary biogenic molecular markers). They identified a SOA source (10%) and they related an unidentified source (24%) with vegetative detritus, woodsmoke, natural gas, coal, dust/soil and food cooking. Srivastava et al. (2018) performed a PM10 source apportionment using specific primary and secondary organic molecular tracers in Grenoble (France). They distinguished between two categories of primary biogenic organic aerosols (fungal spores and plant debris), and biogenic and anthropogenic secondary organic aerosols (SOA). Primary and secondary biogenic contributions together accounted for at least 68% of the total OC in summer and anthropogenic primary and secondary sources represented at least 71% of OC in winter.

Those studies serve as examples to conclude that a source apportionment analysis combining organic molecular tracers with OC/EC and the inorganic tracers lead to an integrated understanding of the different sources obtained, allowing for the identification of both natural and anthropogenic biogenic sources.

3. Non-refractory PM1 (NR-PM1)

There is a growing interest on source apportionment studies in the recent years, particularly for the organic fraction of the submicron aerosol. In the last decade, many articles have reported site-specific source apportionment results but there are also some intercomparison studies in Europe both for AMS and ACSM (Crippa et al., 2014 and Chen et al., 2022, respectively). Taking only the results from urban sites of these studies, the main conclusions drawn are these that follow:

- The most widely used approach to perform source apportionment of the organic aerosol (OA) is the Positive Matrix Factorisation algorithm.
- OA is the main component of the fine aerosol (PM_1), ranging from a 21-64% of the total PM_1.
- The sources of OA which are identified at all stations are: hydrocarbon-like OA (HOA, related to traffic), biomass burning OA (BBOA, in all sites except Helsinki), Less-Oxidised Oxigenated OA (LO-OOA) and More-Oxidised OOA (MO-OOA). The two first and the two second sources are of primary and secondary origin, respectively. The differentiation of the secondary OA (SOA) into LO-OOA and MO-OOA is achieved in all the datasets independently and the mean ratio LO-OOA-to-MO-OOA is 1.1 ± 0.6 (adim.).
- Other sources identified in European sites are cooking-like OA (COA), mostly at low-latitude sites, cigarette-smoke OA (CSOA, in Zurich), Coal Combustion OA (CCOA, in Krakow) and shipping + industry (SHINDOA in Marseille).
- The highest contributions are those from the SOA factors, which in sum represents a 67% in median, being MO-OOA (34% median) slightly above the LO-OOA (32% median). SOA is in all the cases the main constituent of OA.
- Primary factors contribute in median a 13% for HOA, a 12% for COA, a 14% of BBOA, a 7% of CSOA, and a 4% of ShINDOA.
- During the highly-polluted stagnation events, concentrations of the primary OA sources grow significantly. In particular, all factors related to the combustion of solid fuel (i.e., BBOA, SFOA, and CCOA) show the most pronounced enhancement when the OA mass loading increases, especially during winter seasons. However, in all cases, more than a half of the OA is still of secondary origin.
- HOA shows a distinct pattern at urban sites with characteristic morning and evening rush-hour peaks. COA shows distinct noon and evening peaks with minor standard deviations, which suggests small spatial variabilities of cooking emissions. BBOA has a diel cycle with reduced values during the day and a marked evening peak, which indicates that most likely, (residential) heating emissions are the main contributor to BBOA. The diel cycle of LO-OOA reveals night-time maxima with a slight decrease at noon, suggesting local production or enhanced vapour partitioning onto pre-existing aerosol in the shallow nocturnal boundary layer. The MO-OOA diel trend shows the most stable pattern.

4. Equivalent Black Carbon

An increasing number of source apportionment studies have investigated the proportion of Black Carbon (BC) that stems from the burning of fossil fuels in vehicular exhaust emissions (road traffic; BC_{LF}) and solid fuel combustion (mainly from residential and commercial wood and coal burning; BC_{SF}) in specific urban, traffic, and regional background environments. BC-source apportionment research attracted a significant amount of attention due to the potential health and climate impacts of soot (i.e., combustion particles made of a core of BC and coating of organic and sulfur compounds) of which BC is a tracer (Blanco-Donado et al., 2022; Grange et al., 2020; Helin et al., 2018; Kumar et al., 2020; Liakakou et al., 2020; Milinković et al., 2021; Mousavi et al., 2018, 2019; Sandradewi et al., 2008; Favez et al., 2009; Segersson et al., 2017; Titos et al., 2017).

After reviewing about 50 BC source apportionment studies that have been conducted in either Europe or the US in the last decade, different source apportionment methods were found to be used across different regions (see Table 5). Among them, Aethalometer model and Positive Matrix Factorization (PMF) methods were the most commonly used techniques demonstrating the significance of different combustion sources, such as road traffic (i.e., liquid fuel) (BC_{LF}) and solid fuel combustion (BC_{SF}) in Europe and the US. Other complementary methods as Chemical Mass Balance (CMB), Radiocarbon, Specialized air quality models, and Macro-tracer have been also used in combination for particulate matter (PM) and black carbon source apportionment (Belis et al., 2013; Hopke, 2016; Viana et al., 2008). Each method has its own strengths, limitations, and associated uncertainties, leading to variations in the results obtained when comparing different BC source apportionment studies.

Among the existing recent literature, aethalometer model is commonly used for source apportionment of BC (see Table 5). This method can provide insights into the temporal and spatial variations of BC sources. This bilinear model typically involves the use of source-specific absorption Ångström exponents (AAEs) obtained from experimental measurements or values from literature (Sandradewi et al., 2008; Zotter et al., 2017). AAEs spectra are derived for specific types of BC sources, such as vehicle emissions, biomass burning, or industrial emissions. By applying these spectra to the measured absorption coefficients, the contributions of different sources to the observed BC concentrations can be estimated. However, there are certain limitations to consider when using aethalometer model. The output of this model is highly sensitive to the input of AAEs. The method relies on accurate calibration factors and representative AAEs for different sources, which may introduce uncertainties. This bilinear model assumes that the absorption properties of BC remain constant, regardless of aging or mixing with other aerosol components. However, variations in BC mixing state and aging processes can affect the accuracy of source apportionment results due to changes of characteristics of optical properties of a specific source over time (Harrison et al., 2013; Helin et al., 2021; Luo et al., 2023; Virkkula, 2021).

There are evidences from a number of experimental studies that demonstrate challenges on the reliability of the estimations derived from aethalometer model. In fact, this model has been developed in a region which only accounted for the presence of two predominant sources of BC: biomass burning and fossil fuel combustion (linked to traffic) (Sandradewi et al., 2008). The most widely used AAE values are the ones recommended by Sandradewi et al. (2008) and Zotter et al. (2017). These values (AAE_{ff} = 1.0 or 0.9 and AAE_{bb} = 1.68 or 2) have been obtained in Switzerland with the significant contribution from biomass burning emissions. However, a robust AAE value may vary over location and time depending on the type of fuel burnt and the combustion conditions and efficiency including emissions from shipping, coal, traffic and residential wood burning in a specific area (Helin et al., 2021). In addition AAEs are sensitive to both particle geometry, morphology and size distribution (Li et al., 2016; Liu et al., 2018). For example, Liu et al. (2022) reported the determination of AAE pairs (αff and αwb) with associated uncertainties of 4.63% and 3.32%, respectively, derived from the dynamic optimization of AAE values. Helin et al., (2021) has been presented a broad estimation of AAE (470-950 nm) with relative uncertainty in the different emission measurements with the high uncertainty observed for the sauna-stove emissions. In a numerical investigation conducted by Liu et al. (2018), it was observed that the AAE of BC exhibits a variable range of 0.5 to 1.5. Furthermore, Garg et al. (2016) demonstrated that the overall uncertainty in determining BC mass values for source apportionment studies can be influenced by factors such as attenuation measurement at different wavelengths, uncertainty associated with the flow controller, and the selection of suitable mass absorption cross-section (MAC) values. In addition, the post-processing of aethalometer data, taking into account measurement noises as discussed by Backman et al. (2017), has been represented an additional substantial source of uncertainty in aethalometer measurements and the utilization of aethalometer models for source apportionment.

Therefore, the source apportionment outcome of black carbon obtained through the use of aethalometer models can exhibit bias, attributable to AAE values that are relatively higher or lower in magnitude. There are studies that have recommended the use of AAE frequency distributions as an alternative method to estimate site-specific AAE

values (Tobler et al., 2021). Alternatively, the incorporation of time-dependent AAEs into the aethalometer model can be proposed, with the aim of evaluating its performance.

Another common approach, PMF is a widely used technique for PM and BC source apportionment in air pollution studies (Grange et al., 2020; Liu et al., 2022; Masiol et al., 2019; Massabò and Prati, 2021; Rivas et al., 2020; Squizzato et al., 2018; Tobler et al., 2021; Wang et al., 2021). PMF is a receptor model that aims to identify and quantify the contributions of different emission sources to ambient PM and BC concentrations. This method uses the measured BC concentration data along with additional information such as chemical composition, particle size distribution, and meteorological data. The advantages of PMF for BC source apportionment include its ability to identify distinct emission sources, even in complex atmospheric environments with multiple source contributions. It can provide quantitative estimates of source contributions and help in understanding the spatial and temporal variations of BC pollution. PMF is also flexible and can be applied to various types of BC measurements, such as filter-based sampling or online instrumentation. However, like any modeling technique, PMF has certain limitations. The accuracy of the results depends on the quality and representativeness of the input data, including the measurement data and ancillary information. Uncertainties in the source profiles, model assumptions, and parameter selection can influence the results. Interpretation of the factors requires expert knowledge and careful consideration of potential confounding factors (Hopke, 2016; Hopke et al., 2023).

In this context, an ongoing novel implementation of the PMF approach is being conducted as part of the EMEP intensive measurement campaign to enhance the accuracy of eBC source apportionment results (Platt, S. M., et al., 2022, in preparation). Additionally, previous studies have employed various methodologies to compare the outcomes of the aethalometer model with source apportionment results using offline auxiliary measurements such as trace chemical levoglucosan (LG), Elemental Carbon (EC), radiocarbon (^{14}C), or a combination of different source apportionment methods to effectively attribute carbonaceous matter. For instance, Bibi et al. (2021); and Zheng et al. (2021) have recently introduced novel methodologies to enhance the accuracy of BC source apportionment by employing a combination of PMF and aethalometer modeling techniques. These innovative approaches offer improved discrimination of various BC sources, leading to more reliable and robust identification of BC origins.

The findings derived from a diverse array of studies on BC source apportionment demonstrate the heterogeneity of sources across different regions, leading to variations in the relative contributions of fossil fuel and biomass burning sources (e.g., FF, BB) that cannot be precisely categorized within a specific range. Notably, factors such as atmospheric aging conditions, morphologies, and size distribution play significant roles in shaping the fractions of fossil and biomass sources, further introducing statistically significant biases in source characterization (Ferrero et al., 2021; Luo et al., 2023). It is important to acknowledge that BC source apportionment uncertainties may vary in their quantifiability. While some uncertainties can be quantified through rigorous sensitivity analyses and uncertainty propagation methods, others may be more challenging to quantify due to the limitations in data availability. To enhance the consistency and comparability of BC source apportionment studies, several recommendations have been put forth in the literature. Firstly, it is recommended to adopt a multi-tracer approach or macro-tracer method that combines BC measurements with other complementary measurements, such as EC, OC, and other chemical species (Briggs and Long, 2016; Fourtziou et al., 2017). This comprehensive approach provides additional information for distinguishing between primary and secondary BC sources and helps identify specific emission sources. Nevertheless, these types of measurements are not extensively accessible at monitoring sites.

Secondly, the use of high-resolution time series measurements is encouraged to capture the temporal variability of BC sources to enable the identification of diurnal, weekly, and seasonal patterns, facilitating the identification and quantification of different source contributions especially for human exposure assessments (Gregorič et al., 2020; Hamilton and Harley, 2021). In addition, advanced source apportionment methods are recommended to enhance

the analysis and interpretation of BC source apportionment results (Hopke et al., 2023). In addition, to ensure the comparability of results across different studies, it is essential to report detailed information on filter absorption measurement techniques, instrument calibration, data quality control, and uncertainty estimation. Transparent reporting enables better understanding and assessment of the reliability and robustness of the BC source apportionment results.

Table 5. Overview of PM and BC source apportionment studies.

No	Author(s)	Year	Location	Source apportionment method /AAE
1	Viana et al.	2008	Europe	Source apportionment of particulate matter in Europe: A review of methods and results
2	Sandradewi et al.	2008	Switzerland	Aethalometer model
3	Favez et al.	2009	France	Aethalometer model
4	Favez et al.	2010	France	Chemical Mass Balance (CMB, performed with off-line filter measurements) and (PMF, applied to Aerosol Mass Spectrometer measurements)
5	Herich et al.	2011	Switzerland	Aethalometer model
6	Yttri et al.	2011	North Europe	Levels of source specific tracers, i.e. cellulose, levoglucosan, mannitol and the 14C/12C ratio of total carbon (TC) used as input for source apportionment
7	Wang et al.	2012	USA	PMF-Delta C
8	Močnik et al.	2012	Slovenia	Aethalometer model
9	Harrison et al.	2013	UK	Aethalometer model
10	Belis at al.	2013	Europe	source apportionment using receptor models
11	Jeong et al.	2013	Canada	PMF
12	Liu et al.	2014	UK	SP2 attribution, BC–AMS–PMF and Aethalometer method
13	Belis at al.	2015	Global	Performance and uncertainty of source apportionment models in intercomparison exercises
14	Philip K. Hopke	2016	Global	Review of receptor modeling methods for source apportionment
15	Briggs at al.	2016	EU-USA	Critical review of black carbon and elemental carbon source apportionment
16	Ulevicius et al.	2016	Europe	PMF results and radiocarbon (14C) measurements of the elemental (EC) and
17	Martinsson et al.	2017	Sweden	Aethalometer model – evaluation by radiocarbon and levoglucosan analysis
18	Zotter et al.	2017	Switzerland	Aethalometer model using radiocarbon measurements
19	Becerril-Valle	2017	Spain	Aethalometer model
20	Diapouli et al.	2017	Greece	Aethalometer model
21	Titos et al.	2017	Spain	Levoglucosan tracer and online real- time Aethalometer methods
22	Mousavi et al.	2018	USA	Aethalometer model
23	Helin et al.	2018	Finland	Aethalometer model
24	Irena et al.	2018	Slovenia	Aethalometer model
25	Mousavi et al.	2019	Italy	Aethalometer model
26	Thunis et al.	2019	Global	Source apportionment- Strengths and weaknesses of existing approaches
27	Miller et al.	2019	USA	EPA's Positive Matrix Factorization 5.0
28	Yunjiang et al.	2019	France	Aethalometer model

29	Yang et al.	2020	Europe	Trends and source apportionment of aerosols in Europe
30	Grange et al.	2020	Switzerland	Aethalometer model
31	Mbengue et al.	2020	Czech	Aethalometer model
32	Almeida et al.	2020	European and Central Asia, urban	EPA PMF 5.0
33	Gregorič et al.	2020	Slovenia	Aethalometer model
34	Liakakou et al.	2020	Greece	Aethalometer model+ Levogluocosan
35	Moschos et al.	2021	Switzerland	Aethalometer model
36	Ferrero et al.	2021	Italy	Aethalometer model
37	Saarikoski et al.	2021	Finland	The results from the PMF analysis were compared with the Aethalometer model
38	Bibi et al.	2021	UK	PMF- A new approach
39	Kaskaoutis et al.	2021	Greece	Combination of Brown Carbon and ethalometer models
40	Aki Virkkula	2021	Finland	Modeled source apportionment (Aethalometer model evaluated with a core–shell Mie model)
41	Bernardoni et al.	2021	Italy	Aethalometer model
42	Milinković et al.	2021	Croatia	Aethalometer model, optimized by using levoglucosan measurements
43	Hopke et al.	2022	Global	PMF
44	Hopke et al.	2022	Global	Source apportionment of particle number concentrations: A global review
45	Popovicheva et al.	2022	Norway	Aethalometer model
46	Blanco-Donado et al.	2022	USA	Aethalometer model
47	Alfoldy et al.	2022	Slovenia	Aethalometer model
48	Blanco-Alegre et al.	2022	Spain	Coupling tracers with the aethalometer model
49	Hopke et al.	2023	USA	PMF-Big data
50	Savadkoohi et al.	2023	Europe	Aethalometer model

5. Particle Number Size Distribution

Ultrafine particles (UFP) have significant impacts on health. They can be present in atmosphere through direct emissions such as from motor vehicles or through new particle formation events (secondary particles), and it is possible to perform source apportionment analysis using particle number size distributions (PNSD).   However, there are only a few studies on source apportionment of UFP compared to larger particles. Hopke et al. (2022) identified 55 peer-review journal papers as reporting source apportionments of PNSD in 102 locations/time periods. Almost all of these apportionment analyses were performed with Positive Matrix Factorization (PMF), but other methodologies were also used in previous studies such as Principal component analysis (PCA) and k-means clustering.

Pey et al. (2009) performed PCA using STATISTICA v4.2 software on datasets composed by number size distribution of aerosols, meteorological parameters, gaseous pollutants and chemical speciation of PM$_{2.5}$, following the methodology proposed by Thurston and Spengler (1985). PCA was also used in other studies (Wehner and Wiedensohler, 2003; Chan and Mozurkewich, 2007; Oliveira et al., 2009; Cusack et al., 2013; Khan et al., 2015). Beddows et al. (2009) analyzed the data using k-means cluster analysis, in which the particle size distributions are generalized by cluster types (characteristic of an emission or formation process) which facilitate an understanding of the temporal and spatial trends of the size distribution. This method was used in several studies (Dall'Osto et al., 2011; Wegner et al., 2012; Brines et al., 2014; Brines et al., 2015; Dall'Osto et al., 2019; Chen et al., 2021; among others).

PMF is the most recently used data analysis method to identify and apportion the sources of PNSD. But different methodologies have been applied in the analysis by PMF. EPA PMF 3.0 (Gu et al., 2011; Wang et al., 2013; Liu et al., 2014; among others) and EPA PMF 5.0 (Squizzato et al., 2019; Dai et al., 2020; Pokorná et al., 2020; among others) are the last versions of PMF Model software using graphical user interfaces that ease data input, visualization of model diagnostics and exporting of results. It uses the underlying program Multilinear Engine 2 (ME-2, Paatero, 1999) as the factor analytic problem solver. But this software accepts a limited number of observations, then PMF can also be performed using ME-2 without EPA software (Ioar et al., 2019). Hopke et al. (2023) has developed a protocol to use the ME-2 for larger datasets.

Hopke et al. (2022) searched the literature for all papers and source categories have been generally characterized as Nucleation, Traffic 1, Traffic 2, Heating, O_3-Rich, Secondary Inorganic Aerosol (SIA) and Other that include biomass burning and unidentified sources.

- Nucleation has a single mode with its peak size ranging from <10 to 25 nm, and there are commonly diel peaks in the contributions of this source associated with traffic temporal patterns during rush hours and a mid-afternoon peak when high photochemical activity leads to new particle formation events.
- Two traffic factors are commonly reported with major number modes with peaks around 30-35 nm, commonly ascribed to spark-ignition vehicles, and 60-80 nm, associated with diesel vehicle emissions. It is also suggested that the smaller mode particles represented freshly emitted traffic particles and larger mode is due to coagulation of the aerosol as it moves from the source area.
- Building heating can be accomplished through the combustion of a variety of fuels that produce very small particles including natural gas, liquid petroleum gas and No. 2 oil, or fuels that produce a distribution with its mode around 90 to 100 nm, such as residual oil and solid fuels including coal, wood and other biomass.
- Ozone rich factors are also resolved, with multiple modes and diel patterns that typically peaks in the early to mid-afternoon, which suggest that particles have grown through condensation of secondary material.
- SIA is typically attributed to factors with a mode above 100 nm. When its maximum is in winter, it is typically labelled as secondary nitrate, with a mode cantered at 250 nm, while in summer, it is called secondary sulphate, with a mode at ~ 100 nm. The seasonal patterns of these factors fits with the lower production of sulphate in winter given the lower levels of photochemical activity and the thermal dissociation of ammonium nitrate at the higher temperatures in summer.
- Other sources include biomass burning, urban background, industrial emissions, mixed sources, dust, and unknown. Most of these size distributions have their primary mode above 100 nm.

6. Volatile Organic Compounds

This section of the report will focus on the literature review and recommendation on the source apportionment of VOCs in urban areas. The authors only found 4 studies that performed a VOC source apportionment in urban areas in Europe. To aid in this literature review 9 other studies were added to the literature review that either were performed in rural areas in Europe, or urban areas in the U.S. or Asia. This section will focus on the summary of these studies and highlight their conclusions in bullet points. After careful research the following conclusions were made:

- The studies used both automated gas chromatography with flame ionization detector (GC-FID), using a VOC analyzer, and proton transfer reaction mass spectrometry (PTR-MS), using either a quadruple or time of flight mass detector to measured ambient VOC concentrations.

- o The automated GC-FID were the best apparatus to measure C_2-C_{11} alkanes, alkenes, and alkynes, in addition to isoprene and benzene.
  - o The PTR-MS was the best machine to measure aromatic VOCs and oxygenated VOCs with high resolution.
- Positive Matrix Factorization model (PMF) using the EPA 5.0 software was used in all papers to perform the source apportionment. The software is excellent as it does not require previous knowledge on the source factors to perform the PMF, and it has built-in bootstrapping and error estimations for the model.
- A variety of factors were identified in the urban environment. Over the 13 papers a total of 84 unique VOCs were measured. Below all found sources are described with their common tracers.
  - o Natural gas source was mostly identified by high contributions of ethane and propane, with minor contributions from butane.
  - o Wood-burning source was traced by acetonitrile, acetylene, ethylene, and benzene.
  - o Biogenic source was identified by isoprene and its oxidation products (MVK+MACR). Although isoprene can have a biogenic origin, its anthropogenic origin is often not described. The addition of monoterpenes can aid in identifying its biogenic origin.
  - o Motor vehicle exhaust was characterized by high contributions from alkanes, especially butane and pentane, and BTEX (benzene, toluene, ethylbenzene, and xylenes). The exact tracers highly depend on the type of traffic and engines most common in the urban area, and depending on the VOCs included in the study, this source was separated or a combination of traffic sources.
  - o Solvent source is often traced by high contributions of OVOCs and BTEX.
  - o Some specific industry sources were found, but this highly depends on the type of industry what VOCs were used to identify this source.
  - o Various studies found a secondary source compromised of OVOCs, although some studies identified this source as long-lifetime VOCs.
- The addition of wind speed and wind direction data gave the possibility to create a conditional probability function (CPF) plots, which could aid in identifying the sources of various factors, such as specific industries, given their specific location.
- Correlations with NO_x (NO_2 + NO), O_3, CO, and SO_2 could further aid in the identification off various sources. For instance, NO_x and CO are clear indicators of anthropogenic activities, while one study showed that a correlation with NO_2 and O_3 could show secondary formation.
- Including temperature and solar radiation correlation aided in the identification of secondary and biogenic sources.
- It must be mentioned that most of these VOC source apportionments are in regards to the observed VOC concentrations and not the initial VOC concentrations. Many VOCs, especially alkenes and aromatics, have a substantial decrease from photochemical loss, and therefore its contribution will be underestimated. Although, this did not hinder the source identification.
- None but one study used a dispersion-normalized PMF (DN-PMF) used to decrease the impact of atmospheric dispersion. To be able to perform a DN-PMF, the mixed layer height and mean wind speed is required.

Table 6 summarizes the papers used as a source for this report. Their reference name and title are presented, together with the station type, location, VOC measurement method, and source apportionment model.

Table 6. Overview of VOCs source apportionment studies.

	Reference	Title	Station	Location	Method	Apportionment
European Urban	Baudic et al. (2016)	Seasonal variability and source apportionment of volatile organic compounds (VOCs) in the Paris megacity (France).	Urban	Paris, FR	PTR-QMS GC-FID	EPA PMF 5.0
	Gaimoz et al. (2011)	Volatile organic compounds sources in Paris in spring 2007. Part II: Source apportionment using positive matrix factorization.	Urban	Paris, FR	GC-FID PTR-QMS	PMF2
	In 't Veld et al. (2023)	Identification of volatile organic compounds and their sources driving ozone and secondary organic aerosol formation in NE Spain.	Urban Rural	Barcelona, ES NE Spain	PTR-QMS PTR-ToF-MS	EPA PMF 5.0
	Lanz et al. (2008)	Receptor modelling of $C_2 - C_7$ hydrocarbon sources at an urban background site in Zurich, Switzerland: changes between 1993 – 1994 and 2005 – 2006.	Urban	Zurich, CH	GC-FID	EPA PMF 5.0
European Rural	Yáñez-Serrano et al. (2021)	Dynamics of volatile organic compounds in a western Mediterranean oak forest.	Rural	NE Spain	PTR-QMS	EPA PMF 5.0
	Leuchner et al. (2015)	Can positive matrix factorization help to understand patterns of organic trace gases at the continental Global Atmosphere Watch site Hohenpeissenberg.	Rural	Germany	GC-FID	EPA PMF 3.0
U.S./Asia Urban	Brown et al. (2007)	Source apportionment of VOCs in the Los Angeles area using positive matrix factorization.	Urban	LA, U.S.	GC-FID	Multivariate factor analysis tool
	Healy et al. (2022)	Spatially resolved source apportionment of industrial VOCs using a mobile monitoring platform.	Industrial	Ontario, CA	PTR-ToF-MS	EPA PMF 5.0
	Liu et al. (2023)	Effect of photochemical losses of ambient volatile organic compounds on their source apportionment.	Urban	Tianjin, CN	GC-FID	EPA 5.0
	Pallavi et al. (2019)	Source apportionment of volatile organic compounds in the northwest Indo-Gangetic Plain using a positive matrix factorization model.	(Sub)urban	India	PTR-ToF-MS	EPA 5.0
	Song et al. (2019)	Source apportionment of VOCs and their impact on air quality and health in the megacity of Seoul.	Urban	Seoul, KR	GC-FID	EPA 5.0

	Tan et al. (2021)	Characteristics and source apportionment of volatile organic compounds (VOCs) at a coastal site in Hong Kong.	Urban	Hong Kong, CN	PTR-QMS	EPA 5.0
	Kuo et al. (2014)	Source apportionment of particulate matter and selected volatile organic compounds with multiple time resolution data.	Urban	Taipei, Taiwan	GC-FID	Mass Balance

7. On-line elements

The first published study (Furger et al. 2017), was about the evaluation of the data quality by intercomparison with other independent measurements and test its applicability for aerosol source quantification. The field campaign was performed at the permanent station Härkingen, of the Swiss national monitoring network from 23 July until 13 August 2015. The Xact was configured to measure 24 elements in PM10 with 1 h time resolution. Data quality was evaluated for 10 24 h averages of Xact data by intercomparison with 24 h PM10 filter data analysed with ICP-OES for major elements, ICP-MS for trace elements, and gold amalgamation atomic absorption spectrometry for Hg. The slopes of the regressions between Xact 625 and ICP data varied from 0.97 to 1.8 (average 1.28) and thus indicated generally higher Xact elemental concentrations than ICP for these elements. It must be noted here that those results refer to the first generation of Xact instruments (Xact 625). Improvements have been done since then.

Another study on the evaluation of the instrument has been published by (Tremper et al. 2018). This study had three goals: a) Laboratory evaluation using a novel mass-based calibration technique to independently assess the accuracy of the XRF against laboratory generated aerosols, b) filed evaluation in three contrasting field deployments; a heavily trafficked roadside site (PM10 and PM2.5, Jul 2014 to Mar 2015), an industrial location downwind of a nickel refinery (PM10, Nov 2015 to Dec 2015)) and an urban background location influenced by nearby industries and motorways (PM10, Jan 2017 to Mar 2017), and c) offline analysis of filter samples using the Xact. Results from the implementation of the new calibration technique revealed that that generated particles can indeed serve as an alternative calibration method for this instrument. The comparison to ICP-MS showed a good agreement to the measurements of daily filter samples in all cases with a median R^2 of 0.93 and a median slope of 1.07. Finally, filters analysed off-line with the Xact compared well to in situ Xact measurements with a median R2 of 0.96 and median slope of 1.07.

The first source apportionment study in Europe using Xact data was the one from (Rai et al. 2019). This study used the same data as (Furger et al. 2017) for the identification of the sources of the elemental component of PM. Eight different sources were identified in PM10el (elemental PM10) mass driven by the sum of 14 elements (notable elements in brackets): Fireworks-I (K, S, Ba and Cl), Fireworks- II (K), sea salt (Cl), secondary sulfate (S), background dust (Si, Ti), road dust (Ca), non-exhaust traffic-related elements (Fe) and industrial elements (Zn and Pb). The major components were secondary sulfate and non-exhaust traffic-related elements followed by background dust and road dust factors, explaining 21 %, 20 %, 18% and 16% of the analysed PM10 elemental mass, respectively, with the factor mass not corrected for oxygen content. Further, there were minor contributions (on the order of a few percent) of sea salt and industrial sources. To get to these results the authors had to use a novel source apportionment approach to go around the mixing the very high contribution of fireworks had during the period of study.

An interesting study by (Belis et al. 2019), consists the first published attempted of combining Xact data with that of other high-time resolution instruments. Parallel hourly online measurements were made using the Xact 625 (CES LLC) XRF analyser and the Q-ACSM (Aerodyne Research Inc.) spectrometer. The sampling campaign was carried out in January–February 2015 in a background monitoring site located in a small town (Veggiano) near Padua, Italy. To ensure the consistency of the final PMF results, a multistep approach was adopted. In the first step PMF was run with only the offline dataset, in the second step only the online organic data were used and in a third step the PMF run was executed using only the online inorganic species. The sources identified were: biomass burning, aged biomass burning, secondary ammonium nitrate and ammonium sulphate, traffic, steel industry and waste thermal treatment.

A study dedicated to assist towards the attribution and quantification of atmospheric nickel concentrations in an industrial area in the UK, was published by (Font et al. 2022). A campaign measuring metals concentrations in PM10 at an hourly resolution was undertaken using the Mobile Atmospheric Research PLatform (MARPL) in Pontardawe (27 Nov - Dec 24, 2015) alongside the long-running UK Heavy Metals monitoring site (Defra). PMF was used to identify the sources and those industrial processes contributing to the ambient concentrations based on the chemical composition. Bivariate polar plots for the factor time series containing Ni in the chemical profile were built and cluster analysis applied to pinpoint and quantify the contribution of each industry to each source process. Two sources were identified to contribute to Ni concentrations, stainless-steel (which contributed to 10% of the Ni burden) and the Ni refinery (contributing 90%). From the stainless-steel process, melting activities were responsible for 66% of the stainless-steel factor contribution.

Another source apportionment study was conducted by (Manousakas et al. 2022). This study utilized the longest available size segregated elemental composition dataset until today in Europe. Measurements were performed at an urban background site in Zürich (Zürich-Kaserne), during May 2019 to May 2020. For data collection, we have used an ambient metals monitor, Xact 625i, equipped with a sampling inlet alternating between PM2.5 and PM10. By implementing interpolation and a newly proposed uncertainty estimation methodology, it was possible to obtain and use in PMF a combined dataset of PM2.5 and PMcoarse (PM10-2.5) having data from only one instrument. The combination of the inlet switching system, the instrument's high time resolution, and the use of advanced source apportionment approaches yielded improved source apportionment results in terms of the number of identified sources, as the model, additionally to the diurnal and seasonal variation of the dataset, also utilizes the variation from the size segregated data. Thirteen sources of elements were identified, i.e., sea salt (5.4%), biomass burning (7.2%), construction (4.3%), industrial (3.3%), light-duty vehicles (5.4%), Pb (0.7%), Zn (0.7%), dust (22.1%), transported dust (9.5%), sulfates (15.4%), heavy-duty vehicles (17%), railway (6.6%) and fireworks (2.4%).

Finally, there is a study on oxidative potential apportionment of atmospheric PM1 using a new approach combining high-sensitive online analysers (including the Xact) for chemical composition and offline OP measurement technique, submitted to Atmos. Chem. Phys., (Camman et al, 2023), but more information is not yet available.

Table 7. Overview of published Xact related studies in Europe.

Site	Country	Period start	Period end	Reference
Härkingen	Switzerland	23 Jul 2015	13 Aug 15	(Furger et al. 2017)
Marylebone Road, London	UK	PM10: 1 Jul 2014 PM2.5: 14 Oct 2014	PM10: 11 Mar 2015 PM2.5: 1 Dec 2014	(Tremper et al. 2018)
Tawe Terrace, Pontardawe	UK	25 Nov 2015	24 Dec 2015	(Tremper et al. 2018)
Tinsley, Sheffiel	UK	19 Jan 2017	27 Mar 2017	(Tremper et al. 2018)
Härkingen	Switzerland	23 Jul 2015	13 Aug 15	(Rai et al. 2019)
Veggiano	Italy	Jan 2015	Feb 2015	(Belis et al. 2019)

Tawe Terrace, Pontardawe	UK	25 Nov 2015	24 Dec 2015	(Font et al. 2022)
Kaserne, Zurich	Switzerland	May 2019	May 2020	(Manousakas et al. 2022)

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