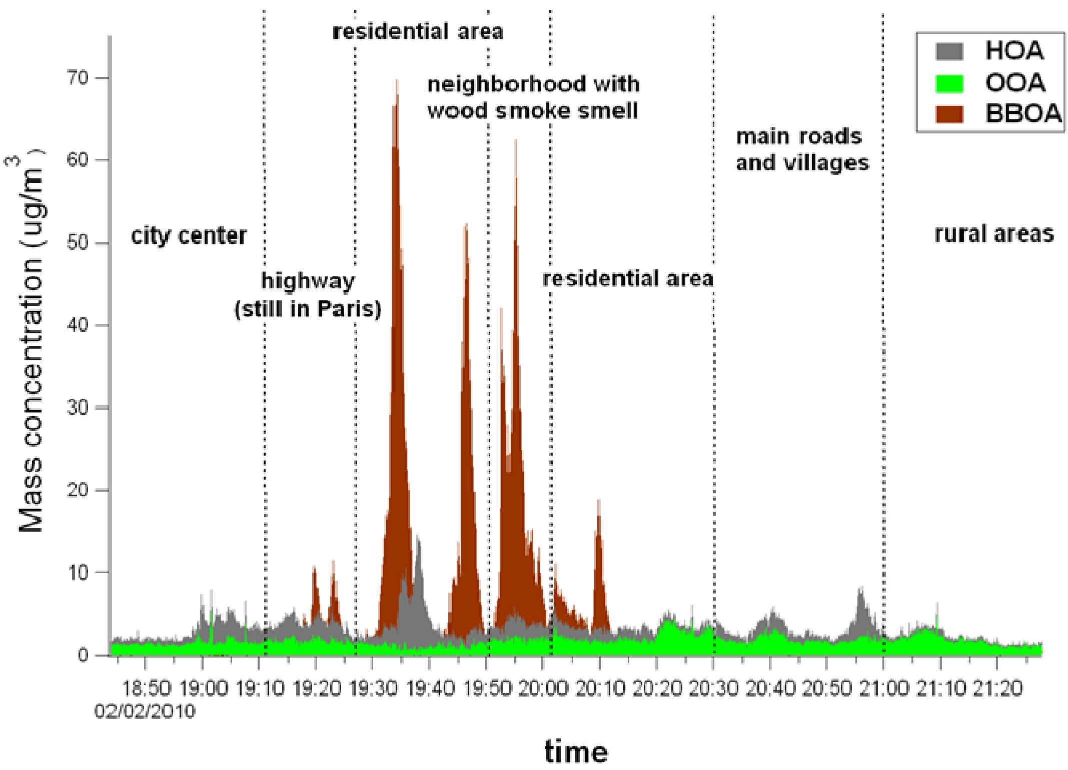
MEGAPOLI Scientific Report 11-05

Source Apportionment of Major Urban Aerosol Components Including Primary and Secondary PM Sources

MEGAPOLI Deliverable D3.2

Urs Baltensperger, Matthias Beekmann
and the MEGAPOLI campaign team



Example of the source apportionment of the organic aerosol as determined from PMF of AMS measurements

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Abstract

Two intensive measurement campaigns were performed in the Ile-de-France region during a one-month summer and a one-month winter period (July 1–31, 2009 and January 15 to February 15, 2010, respectively). The campaigns have aimed at better quantifying primary and secondary organic aerosol sources on an example of a big European megacity (the Paris conglomeration) according to the MEGAPOLI WP3 core objectives:

- O3.1 - To characterize atmospheric aerosol and relevant precursors at two urban and suburban sites in Greater Paris area;
- O3.2 - To provide a source apportionment of PM (separately for ultrafine particles, PM 1, and the coarse mode);
- O3.3 - To examine the evolution of aerosols and gas-aerosol interactions in the urban outflow of Paris;
- O3.4 - To provide additional data for the evaluation of Chemical Transport Models.

The campaign design included 3 primary and 7 secondary fixed ground measurement sites, an aircraft and 5 mobile platforms. This set-up was much bigger than initially planned and funded by the European Commission, due to a large number of additional volunteering contributions by the MEGAPOLI partners and other research groups, and due to additional national (French) funding.

This MEGAPOLI Deliverable D3.1 "*Source Apportionment of Major Urban Aerosol Components Including Primary and Secondary PM Sources*" describes the methods applied to determine contributions of various sources (both primary and secondary) to the Paris aerosol, as well as the major results. Two campaigns seasons (winter and summer) are covered.

1. Introduction

Two intensive measurement campaigns were performed in the Ile de France region during a one-month summer and a one-month winter period (July 1 – 31, 2009 and January 15 to February 15, 2010, respectively). The campaigns aimed at better quantifying primary and secondary organic aerosol sources for a big European Megacity (the Paris region) according to the MEGAPOLI WP3 core objectives O3.1-O3.4. The campaign design included 3 primary and 7 secondary fixed ground measurement sites, an aircraft and 5 mobile platforms (see Deliverables D3.1 and D3.4). This set-up was much bigger than initially planned and funded by the Commission, due to a large number of additional voluntary contributions by the MEGAPOLI partners and other research groups and due to additional national (French) funding.

This Deliverable describes the methods applied to determine contributions of various sources (both primary and secondary) to the Paris aerosol, as well as the major results. Two seasons (winter and summer) are covered.

2. Description of the sites and methods

2.1. Spatial set-up of fixed campaign sites

The campaign design included 3 primary and 7 secondary fixed ground measurement sites. Primary sites included measurements of the concentration of gas phase pollutants, aerosol size-resolved composition, and of aerosol physical properties. In addition, at one site (SIRTA), at the south-western edge of the agglomeration, dynamical meteorological measurements were performed (on a routine basis, independent of the campaign, but real-time measurements and data access was activated during the campaign). Among these three sites, the central urban site (LHVP) is located near the agglomeration centre, in the 13th Arrondissement of Paris in the south-east of the town. The SIRTA site is located about 20 km south-west of the central site, and the Golf de la Poudrerie site about 20 km in the North-East. The two latter sites are suburban and located at the edge of the agglomeration. The three primary sites were thus aligned with respect to a south-west / north-east axis which correspond to the major two wind axes prevailing for the agglomeration. Thus in the case of NE or SW winds, one site was upwind, another central, and the third one downwind with respect to the agglomeration. In addition, secondary sites were devoted to vertical measurements, in general by Lidar, and for two sites also spectroscopic column measurements. They are located within Paris and close suburbs and outside or at the edge of the agglomeration in four different sectors. This allowed derivation of spatial gradients in aerosol load or in boundary layer height. Only measurements at two of the primary sites (LHVP and SIRTA) were contractual in the frame of the FP7 project.

2.2. Description of the aerosol instrumentation

At the three primary sites, a complete set-up of instruments was deployed (Table 1) allowing a detailed characterization of aerosol properties, including their size distribution, volatility, hygroscopicity, optical properties, and chemical composition. This included fast measurements of aerosol composition by Aerosol Mass Spectrometry (AMS with time resolution of a few minutes and single particle composition measurements) and wet chromatographic measurements by a PILS system (at LHVP and SIRTA sites). These measurements give the mass concentration of inorganic ions, and of primary and secondary organic particulate matter, respectively for PM_1 and $PM_{2.5}$ aerosol. In addition, filter measurements were taken to allow for individual analysis of up to 100 individual organic compounds, as well as for carbon-14 analysis.

Table 1 shows that most of the measurements were performed with a high temporal frequency, giving access to the hourly (or even better) variation of pollution. This is an important achievement of this campaign. In addition, the fact that measurements have been performed over a whole summer and a whole winter month allows us to investigate the seasonal variation of gas and particle concentrations.

2.3. Methods for source apportionment

The fine and coarse mode of the atmospheric aerosol exhibit quite different chemical compositions. The cut between the two modes is roughly between 1 and 2.5 µm. Some instruments are mainly sensitive to particles with an aerodynamic diameter d < 1 µm (e.g., the AMS), while other instruments are operated behind a 2.5 µm size cut. In the following it is assumed that there is negligible aerosol mass between 1 and 2.5 µm, such that the discrimination is made either at 1 or 2.5 µm, based on the availability of data.

The chemical composition was used to determine the sources of the Paris aerosol as follows. Nitrate, sulphate and ammonium are formed from the precursor gases NO_x, SO_2, and NH_3, respectively. It is assumed that the contributions from the various sources for nitrate, sulphate and ammonium are identical to the ones of the precursor gases. The relative contributions of the inorganic precursor gases as well as the black carbon concentrations are based on the SNAP inventory of a region approximately delimited by Hamburg at North, Wien at East, Milan at South, and Brest at West.

The black carbon concentration will also be apportioned to diesel and wood combustion (BC_{ff} and BC_{wb}, respectively) based on two different methods. First, the aethalometer method (*Sandrdewi et al., 2008*) will be used, which takes advantage of the fact that the light absorption coefficients of BC from diesel and wood combustion have different wavelength dependences. Second, the carbon-14 method will be applied (*Szidat et al., 2006*, and references therein). This method uses the fact that EC from wood combustion has a contemporary value of C-14, while EC from diesel exhaust contains zero C-14. Due to still ongoing quality assurance of the discrimination between OC and EC in the C-14 method these results are not yet available.

This source apportionment focuses on PM_1, as not enough analyses for a source apportionment of the coarse mode were performed.

Table 1: *Set-up of aerosol measurement at the three primary measurement sites.*

Parameter	Instrument	Time resol.	Urban site LHVP	SOUTH-WEST Suburban site SIRTA	NORTH-EAST Suburban site GOLF
Number and Mass Concentration					
Size distribution - submicron aerosols	DMPS/SMPS	5 min	IfT	UHEL	MPI
CN	AIS	5 min		UHEL	
Size distribution after TD	V-DMPS/SMPS	5 min		FORTH	
Size distribution	APS	5 min	FORTH	PSI	MPI
Size distribution	GRIMM	1min	LSCE	LSCE	MPI
Hygroscopic growth factor	HTDMA	5 min	IfT	PSI	
Cloud condensation nuclei (CCN)	CCN counter	5 min		FORTH/PSI	
PM_1	TEOM-FDMS	15 min			MPI
$PM_{2.5}$	TEOM-FDMS	15 min	INERIS	LSCE	
Fast chemistry (<1 h)					
size resolved chemistry in PM_1 OA, sulfate, nitrate, ammonium, chloride	AMS	7 sec	IfT	PSI	MPI
PMF analysis from above AMS data			IfT	PSI	MPI
size resolved chemistry in PM_1 after TD	AMS	2.5 min		PSI/UHEL	
Inorganic salts in $PM_{2.5}$	PILS-IC	15 min	LSCE	LSCE	
WSOC in $PM_{2.5}$	PILS-TOC	4 min	LSCE		
EC-OC in $PM_{2.5}$	Sunset Field Inst.	60 min	LSCE		
$PM_{2.5}$	TEOM-FDMS	6 min	LSCE		
Size resolved elements in $PM_{2.5}$	RDI / SRXFR	120 min	PSI	PSI	
Individual particles	ATOF-MS or SPLAT	Some min	U CORK ATOF-MS (winter)		MPI SPLAT
Integrated chemistry (1-24 h)					
EC-OC + WSOC + ions + sugars in $PM_{2.5}$	Filters (Partisol)	3 h	LSCE	LSCE	
EC-OC + ions in PM_1	Filters	12h			LSCE
Carbon-14 in PM_1	Filter (daily sampling)	48h	PSI	PSI	
Organic tracers in PM2.5 (up to 100 individual compounds, for primary emission sources and secondary formation pathways – biogenic and anthropogenic,),	Filter (DA80 in $PM_{2.5}$)	12h	LCP - LGGE	LCP - LGGE	
Size resolved chemistry	13-stage Cascade Impactor	24-48h	LSCE	LCP - LGGE	
Optical properties					
Absorption coefficient (BC)	Aethalometer 7-L	5 min	LSCE	PSI/LSCE	
Absorption coefficient (BC)	MAAP	1-5 min	IfT	PSI	MPI
Black carbon	SP2	1 sec		PSI	
Light scattering coefficient	TSI 3wavelength	5 min	IfT	PSI	

The source apportionment of the organic carbon was performed based on positive matrix factorization (PMF) of the AMS data (*Lanz et al., 2007, 2008*). This allowed for separating the total OA into its underlying components, such as oxygenated (mostly secondary) organic aerosol (OOA), hydrocarbon-like organic aerosol (HOA, attributed to traffic), as well as OA from biomass burning (BBOA, attributed to residential emissions).

3. Results

3.1. Source apportionment for summer 2009

3.1.1. GOLF site

Figure 1 shows the chemical composition of PM_1 at the GOLF site as determined from AMS and BC measurements. The organic fraction was separated by PMF into oxygenated (= photochemically aged) organic aerosol (OOA) and hydrocarbon-like organic aerosol (HOA), indicating fresh emissions from fossil fuel burning. Biomass burning aerosol was found to be negligible during the summer campaign.

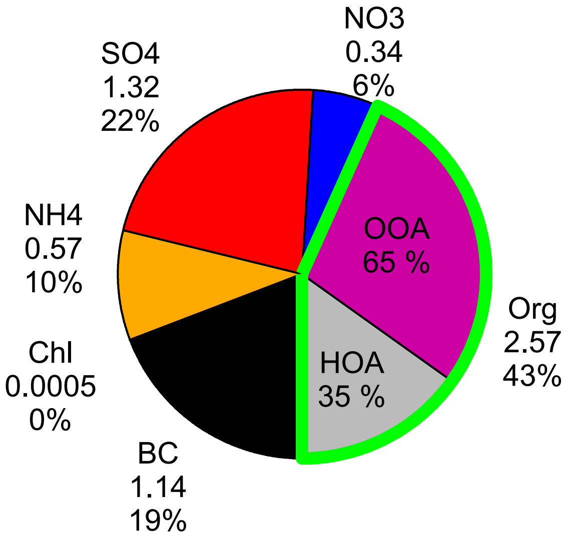

Figure 1: *Chemical composition of PM_1 at the Golf site as determined from AMS and BC measurements. The organic fraction is split into two different factors based on PMF (see 2.3). The absolute numbers refer to mass concentrations in $\mu g\ m^{-3}$.*

3.1.2. LHVP Site

Figure 2 shows the chemical composition of PM_1 at LHVP as determined from AMS and BC measurements. The separation of the organic fraction by PMF is still work in progress.

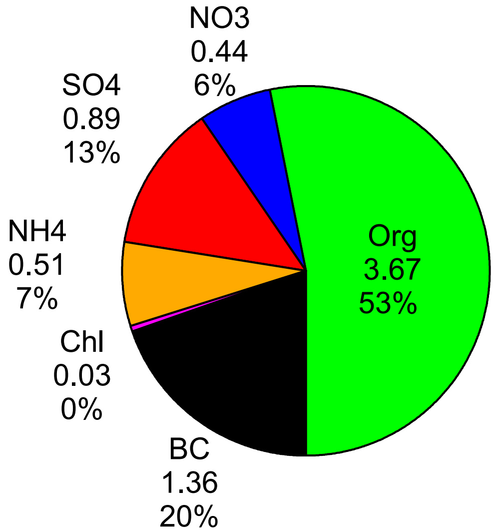

Figure 2: *Chemical composition of PM_1 at the LHVP site as determined from AMS and BC measurements. The absolute numbers refer to mass concentrations in µg m^{-3}.*

3.1.3. SIRTA Site

Figure 3 shows the chemical composition of PM_1 at SIRTA as determined from AMS and BC measurements. The separation of the organic fraction by PMF is still work in progress.

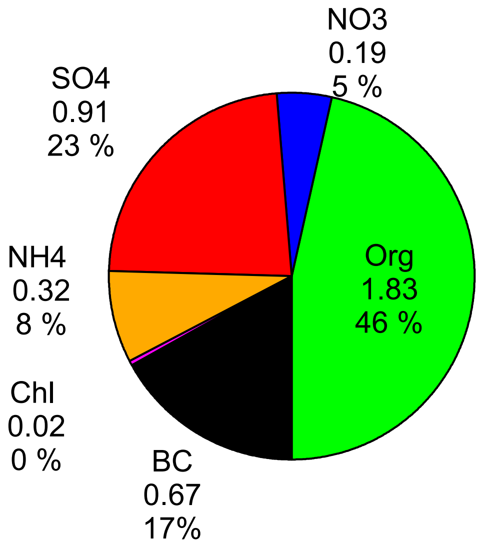

Figure 3: *Chemical composition of PM_1 at the SIRTA site as determined from AMS and BC measurements. The absolute numbers refer to mass concentrations in µg m^{-3}.*

3.1.4. ATR-42 Measurements within pollution sector

Figure 4 shows the chemical composition of PM_1 measured within the ATR-42 research aircraft as determined from AMS and BC measurements. The separation of the organic fraction by PMF is still work in progress. The data have been averaged over all flights.

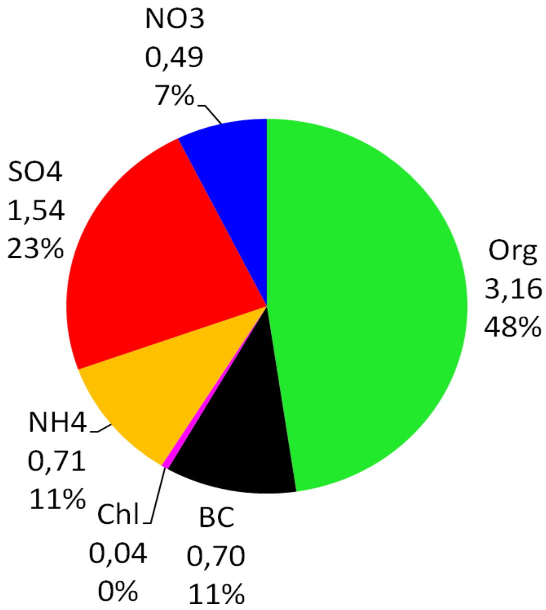

Figure 4: *Averaged chemical composition of PM_1 measured on the ATR-42 aircraft flying principally in the pollution sector. The absolute numbers refer to mass concentrations in $\mu g\ m^{-3}$ as determined from AMS and BC measurements*

3.2. Source apportionment for winter 2010

3.2.1. GOLF site

Figure 5 shows the chemical composition of PM_1 at the GOLF site as determined from AMS and BC measurements.

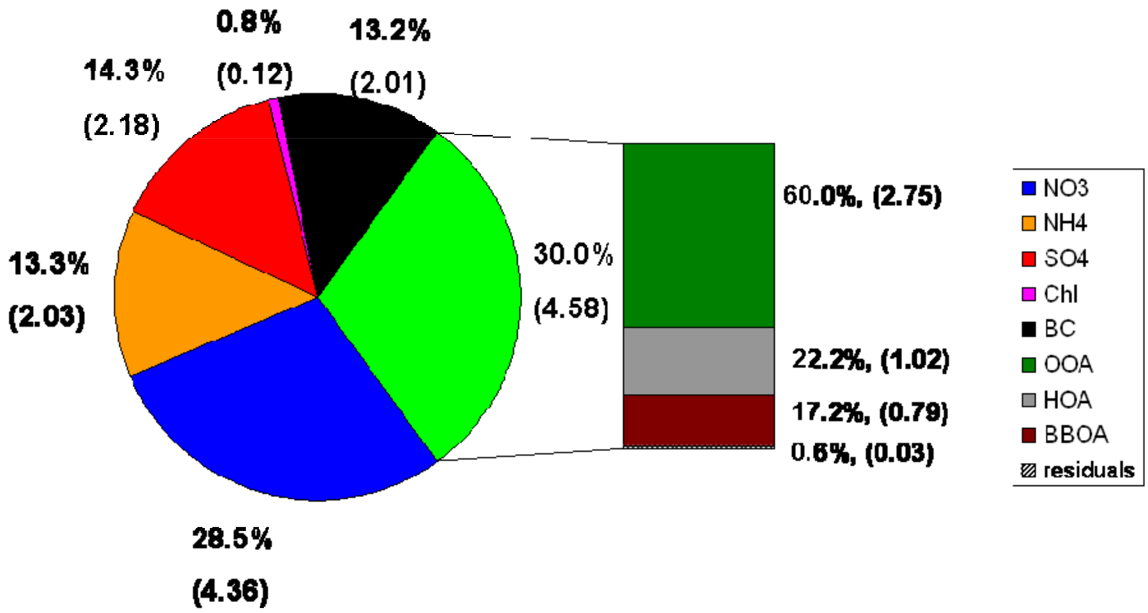

Figure 5: *Chemical composition of PM_1 at the Golf site as determined from AMS and BC measurements. The organic fraction (green) is split into three different factors based on PMF (see 2.3).*

3.2.2. LHVP Site

Figure 6 shows the chemical composition of PM_1 at LHVP as determined from AMS and BC measurements. In contrast to the other two sites, where only HOA, OOA and BBOA were found contributing to OM, here a 4th urban factor could be probably identified but the attribution of the various factors to the corresponding sources is still ongoing.

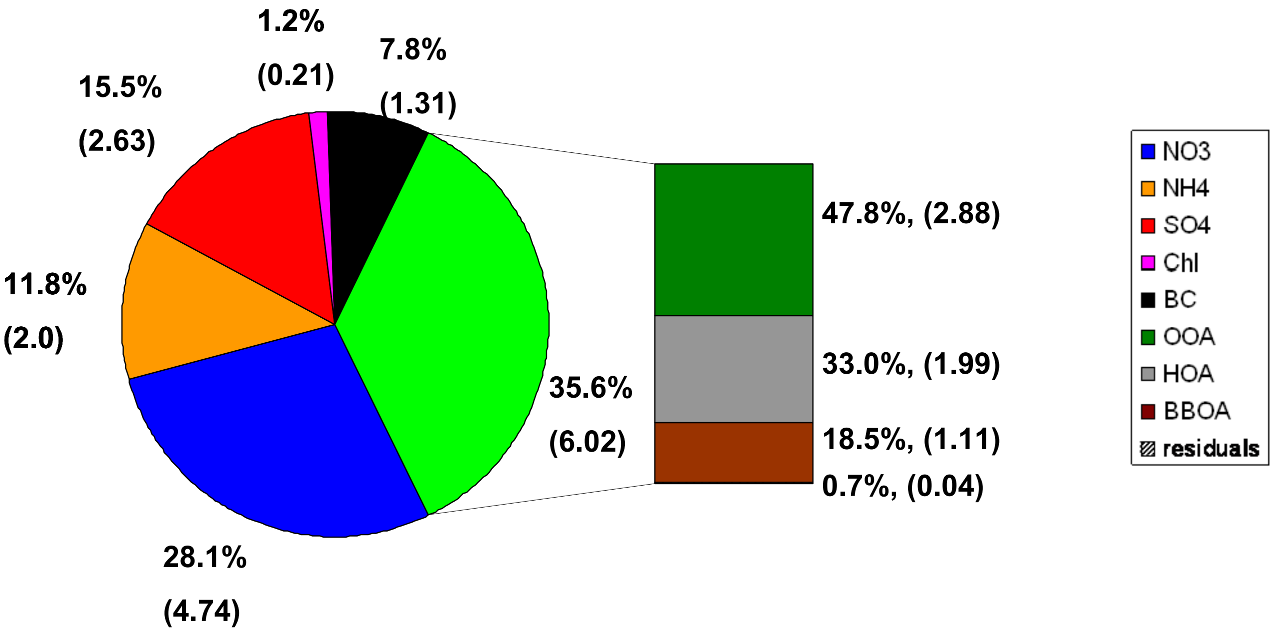

Figure 6: *Chemical composition of PM_1 at the LHVP site as determined from AMS and BC measurements. The organic fraction (green) is split into three different factors based on PMF (see 2.3).*

3.2.3. SIRTA Site

Figure 7 shows the chemical composition of PM_1 at SIRTA as determined from AMS and BC measurements.

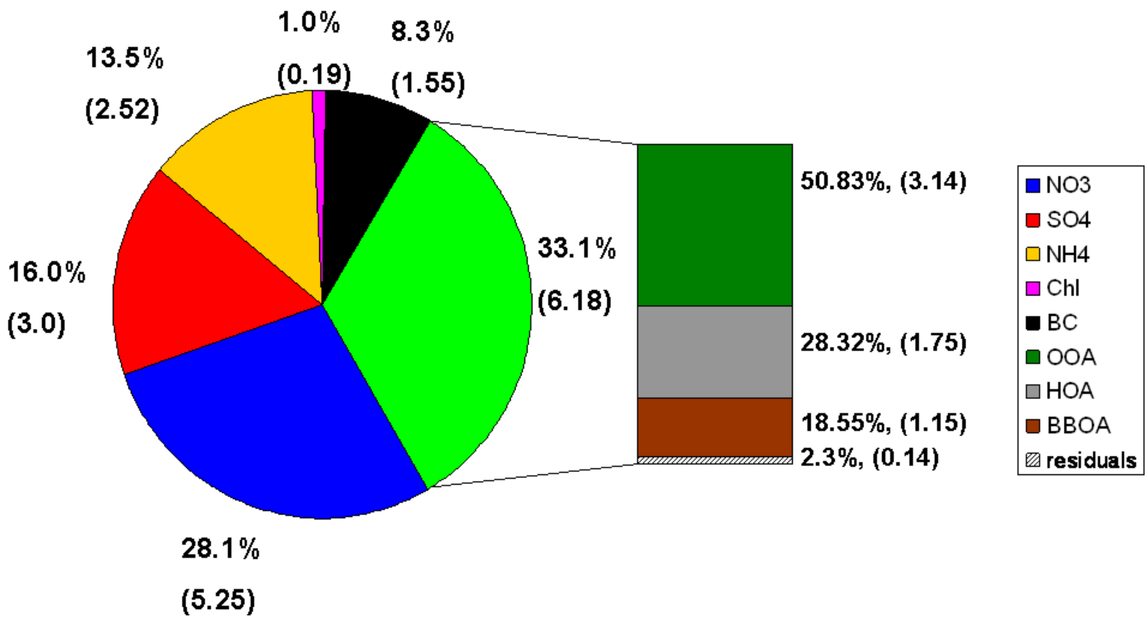

Figure 7: *Chemical composition of PM_1 at the LHVP site as determined from AMS and BC measurements. The organic fraction (green) is split into three different factors based on PMF (see 2.3).*

3.3. Example of mobile measurements

Figure 8 shows an example of the source apportionment of the organic aerosol as determined from PMF of AMS measurements. The high variability of biomass burning organic aerosol resulting in occasionally very high concentrations in residential areas is striking.

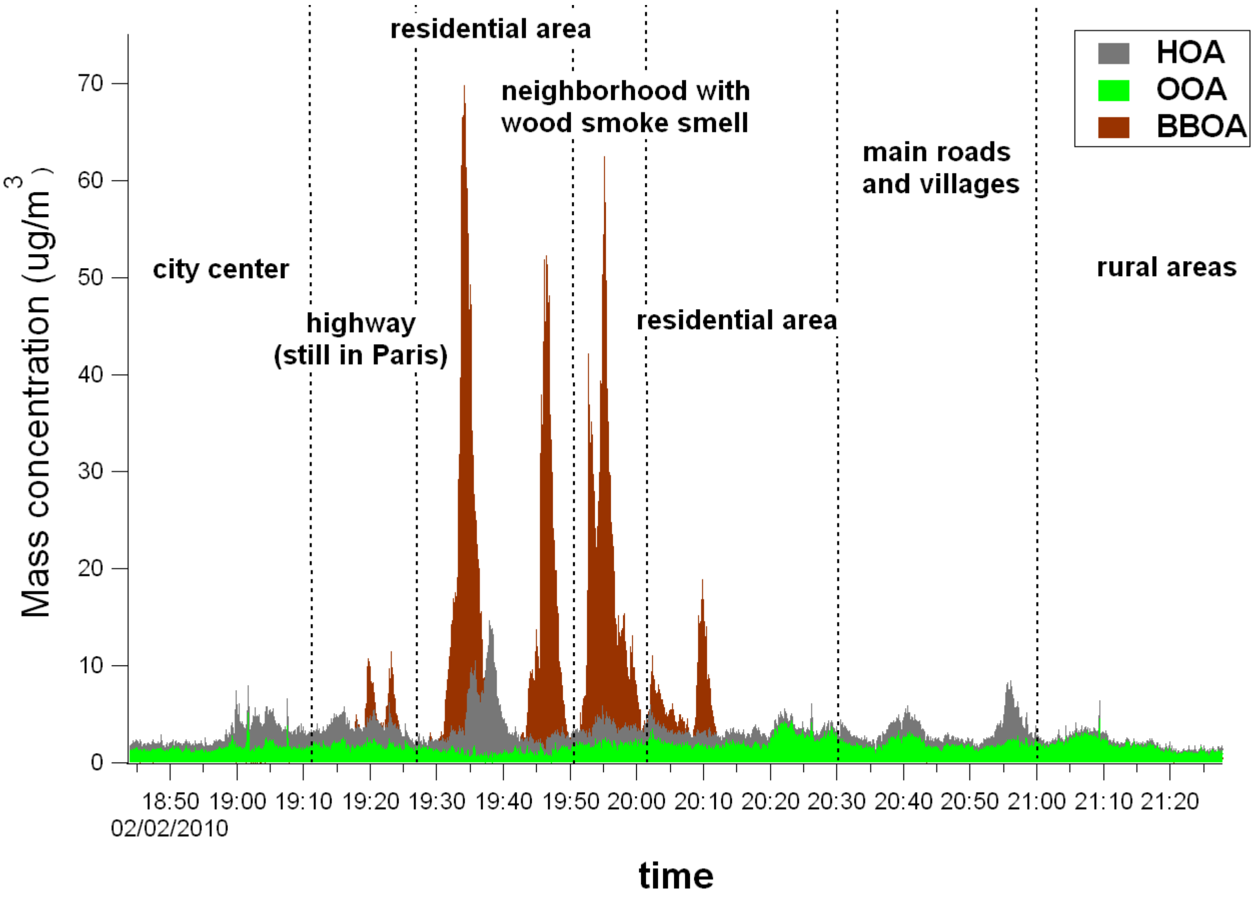

Figure 8: *Example of the source apportionment of the organic aerosol as determined from PMF of AMS measurements.*

3.4. Overview of data

Tables 2 and 3 present an overview on the mass concentrations in PM_1 from different sectors during summer and winter, respectively. It should be noted that these data are still preliminary. In particular, the secondary organic aerosol (oxidized organic aerosol, OOA from the PMF analysis) is not yet discriminated into different sources. This discrimination is subject of ongoing research worldwide.

Table 2: *Mass concentrations (μg/m³) in PM_1 from different sectors during summer.*

Species	**Total**	**Traffic**	**Industry**	**Agriculture**	**Residential (incl. wood combustion)**	**Other (including secondary organic aerosol)**
GOLF						
Sulfate	1.32	0.43	0.83	0.00	0.06	0.00
Nitrate	0.34	0.26	0.06	0.01	0.01	0.00
Ammonium	0.57	0.01	0.02	0.54	0.00	0.00
Black carbon	1.14	0.90	0.16	0.00	0.07	0.00
Organic carbon	2.57	1.18	0.00	0.00	0.00	1.39
Total	5.94	2.78	1.08	0.55	0.14	1.39
LHVP						
Sulfate	0.89	0.29	0.56	0.00	0.04	0.00
Nitrate	0.44	0.33	0.08	0.02	0.01	0.00
Ammonium	0.51	0.01	0.02	0.48	0.00	0.00
Black carbon	1.36	1.08	0.20	0.00	0.08	0.00
Organic carbon	3.67	1.76	0.00	0.00	0.00	1.91
Total	6.87	3.47	0.85	0.50	0.13	1.91
SIRTA						
Sulfate	0.91	0.29	0.57	0.00	0.04	0.00
Nitrate	0.19	0.14	0.03	0.01	0.01	0.00
Ammonium	0.32	0.01	0.01	0.30	0.00	0.00
Black carbon	0.67	0.53	0.10	0.00	0.04	0.00
Organic carbon	1.83	0.73	0.00	0.00	0.00	1.10
Total	3.92	1.71	0.72	0.31	0.09	1.10

For sulphate, nitrate, ammonium and BC: data based on SNAP inventory of a region approximately delimited by Hamburg at North, Wien at East, Milan at South, and Brest at West.

Table 3: *Mass concentrations in PM_1 from different sectors during winter.*

Species	Total	Traffic	Industry	Agriculture	Residential (incl. wood combustion)	Other (including secondary organic aerosol)
GOLF						
Sulfate	2.18	0.25	1.62	0.00	0.31	0.00
Nitrate	4.36	2.01	1.68	0.07	0.60	0.00
Ammonium	2.03	0.05	0.06	1.92	0.00	0.00
Black carbon	2.01	0.98	0.27	0.00	0.76	0.00
Organic carbon	4.58	0.60	0.00	0.00	0.58	3.40
Total	15.16	3.89	3.63	1.99	2.25	3.40
LHVP						
Sulfate	2.63	0.30	1.96	0.00	0.38	0.00
Nitrate	4.74	2.19	1.83	0.07	0.65	0.00
Ammonium	2.00	0.05	0.06	1.89	0.00	0.00
Black carbon	1.31	0.64	0.17	0.00	0.49	0.00
Organic carbon	6.02	0.69	0.00	0.00	0.88	4.45
Total	16.70	3.86	4.02	1.96	2.40	4.45
SIRTA						
Sulfate	3.00	0.34	2.23	0.00	0.43	0.00
Nitrate	5.25	2.42	2.02	0.08	0.72	0.00
Ammonium	2.52	0.06	0.08	2.38	0.01	0.00
Black carbon	1.55	0.76	0.21	0.00	0.58	0.00
Organic carbon	6.18	0.90	0.00	0.00	0.98	4.31
Total	18.50	4.47	4.54	2.46	2.72	4.31

For sulphate, nitrate, ammonium, and BC: data based on SNAP inventory of a region approximately delimited by Hamburg at North, Wien at East, Milan at South, and Brest at West.

4. Conclusions

This MEGAPOLI Deliverable D3.1 "*Source Apportionment of Major Urban Aerosol Components Including Primary and Secondary PM Sources*" describes the methods applied to determine contributions of various sources (both primary and secondary) to the Paris aerosol, as well as the major results of the source apportionment of the Paris aerosol during the MEGAPOLI intensive campaigns.

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EC FP7 Collaborative Project

2008-2011

Theme 6: Environment (including climate change)
Sub-Area: ENV-2007.1.1.2.1:
Megacities and regional hot-spots air quality and climate

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WP4: Megacity air quality
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