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Full project title:

Integrated Climate forcing and Air pollution Reduction in Urban Systems

D3.4 Report on results of source apportionment in all participating cities

WP3 Integrated atmospheric modelling for connecting pressures to the environment to concentrations at the regional and urban scales

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1 Scope

The scope of the present report is to present the results from the source apportionment methods application on PM2.5 data collected in four European cities (Athens, Brno, Ljubljana and Thessaloniki) in the frame of ICARUS project. In particular, PM2.5 samples collected from three different sites in each city (traffic, urban background and rural, see Table 1.1) were chemically analyzed for ions, metals, organic/elemental carbon (OC/EC) and Polycyclic Aromatic Compounds (PAHs). The chemical composition data were inserted in PMF (Positive Matrix Factorization) and PCA (Principal Component Analysis) models with the scope of identifying the main groups of sources and estimating their contribution to PM2.5 concentrations.

Table1.1. ICARUS monitoring sites in Athens, Brno, Ljubljana, Madrid and Thessaloniki.

	Monitoring sites		
Cities (responsible partner)	**Traffic**	**Urban Background**	**Regional**
Athens (NCSRD)	**Marousi**	**Ag. Paraskevi**	**Aliartos** Distance from city center: 101 Km
Brno (MU)	**Brno-Svatoplukova**	**Brno-Lány**	**Košetice** Distance from city centre: 130 km
Ljubljana (JSI)	**MOL – Vosnjakova**	**ARSO – Bezigrad**	**TETOL – Zadobrova** Distance from city center: 7 km
Madrid (ISCIII)	**E. Aguirre station**	**Farolillo station**	**Casa de Campo station**
Thessaloniki (AUTH)	**1) Egnatia** **2) University campus**	**1) Stavroupoli** **2) Eptapyrgio**	**Neochorouda** Distance from city center: 15 km

2 Introduction

Source Apportionment (SA) is the practice of deriving information about pollution sources and the amount they contribute to ambient air pollution levels based on the composition or fingerprints of the sources. One of the SA main approaches is receptor modeling. Receptor-oriented models (also known as receptor models (RMs)) apportion the measured mass of an atmospheric pollutant at a given site, called the receptor, to its emission sources by using multivariate analysis to solve a mass balance equation. These tools have the advantage of providing information derived from real-world measurements, including estimations of output uncertainty. Depending on the knowledge about emission sources there is a wide range of receptor models available from multivariate models like PCA (principal component analysis) and PMF (positive matrix factor), if the knowledge about the emission source is limited, up to regression models and CBM (chemical mass balance) models, if the knowledge about the emission sources is complete.

3 Positive Matrix Factorization (PMF)

3.1 PMF Mathematical background

PMF introduces a weighting scheme taking into account errors of the data points, which are used as point-by-point weights. Adjustment of the corresponding error estimates also allows it to handle missing and below detection limit data. Moreover, non-negative constraints are implemented in order to obtain more physically meaningful factors. The latest PMF version available by USEPA (PMF 5.0) has been used in the present task. PMF analysis background is described in detail by Paatero, 1997. In brief, the factor model can be written as

$$X = GF + E, \qquad (1)$$

where X is the known $n \times m$ matrix of the m measured chemical species in n samples. G is an $n \times p$ matrix of the p sources contributions to the samples (time variations). F is a $p \times m$ matrix of source compositions (source profiles). Both G and F are factor matrices to be determined. E is defined as a residual matrix, i.e., the difference between the measurement X and the model Y as a function of factors G and F.

$$e_{ij} = x_{ij} - y_{ij} = x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj} \, (i = 1,....,n;\ j = 1,...,m; k = 1,..., p) \qquad (2)$$

The objective of PMF is to minimize the sum of the squares of the residuals weighted inversely with error estimates of the data points. Furthermore, PMF constrains all of the elements of G and F to be non-negative; meaning that sources cannot have negative species concentration ($f_{kj} \geq 0$) and samples cannot have a negative source contribution ($g_{ik} \geq 0$). The task of PMF analysis can thus be described as to minimize Q, which is defined as

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{m} (e_{ij} / s_{ij})^2 \qquad (3)$$

with $f_{kj} \geq 0$; $g_{ik} \geq 0$ and s_{ij} is the error estimate for x_{ij}.

In some cases other auxiliary equations can be added in order to include a priori information such as well-known chemical profiles for certain sources (Paatero and Hopke, 2009). The auxiliary equations can be applied to the selected solution in the form of constraints, which can lead to a free rotation of the solution with better physical meaning than the original one. Further, a number of rotations blocking zero values can be introduced to the matrix increasing the rotational stability of the solution.

3.2 Data pre-treatment, analysis of input data and model runs

EPA PMFv.5 model was run for the three sampling sites (rural, traffic and urban background) of Athens, Thessaloniki, Brno and Ljubljana cities. Data from the winter sampling periods were used. The model application on Madrid's data has not been completed yet because samples chemical analysis is in progress.

PM2.5 samples collected during the ICARUS measurement campaigns were analyzed for 27 PAHs, 24 trace elements, anions (Cl^-, NO_3^-, SO_4^{2-}), elemental and organic carbon (EC, OC). Cations and PAHs from some cities were missing. Table 3.1 presents the available data which were used to run models for each city.

Table 3.1. Data availability for source apportionment models' application

city	site	period	PM mass concentration	anions	cations	metals	PAHs	OCEC
Athens	regional	winter	✓	✓		✓	partly	✓
	traffic	winter	✓	✓		✓	✓	✓
	urban backgr.	winter	✓	✓		✓	✓	✓
Brno	regional	winter	✓	✓		✓	partly	✓
	traffic	winter	✓	✓		✓	✓	✓
	urban backgr.	winter	✓	✓		✓	✓	✓
Lubliana	regional	winter	✓	✓		✓		✓
	traffic	winter	✓	✓		✓	✓	✓
	urban backgr.	winter	✓	✓		✓	✓	✓
Madrid	regional	winter	✓	✓				✓
	traffic	winter	✓	✓				✓
	urban backgr.	winter	✓	✓				✓
Thessaloniki	regional	winter	✓	✓		✓		✓
	traffic	winter	✓	✓		✓		✓
	urban backgr.	winter	✓	✓		✓		✓

However, for the input to the model matrix, lighter PAHS were excluded due to their volatilization and a sum of the heaviest (Benzo[b]fluoranthene, Benzo[k]fluoranthene, Benzo[e]pyrene, Benzo[a]pyrene, Indeno[1,2,3-cd]pyrene, Benzo[ghi]perylene) was inserted as one specie, 'SPAHs'. For avoiding double mass counting, either S or SO_4^{2-} was excluded from the analysis. Depending on the case, 'bad' species were excluded from the analysis due to the high percentage of missing values. On the other hand, depending on the case, some species were set as 'weak' due to their low signal/noise ratio (S/N) or/and bad scaled residuals (d-matrix). Finally, outliers were excluded from the analysis. Concentration data below the detection limit (the maximum reported detection limit was used as a conservative limit for all samples) was substituted with one-half of the detection limit and missing concentration data were substituted with the median value (Polissar et al., 2001). The modeling extra uncertainty was adjusted to 8-10%.

Thirty runs were performed for each factor, in order to obtain Q-value stability. All runs converged and Q values ranged between ±1.2%. In each case, the Q-robust value was lower than 1.5 times the Q-true value, indicating that outliers are not significantly impacting the Q value. The optimal number of factors was determined by examining the Q values for PMF solutions resulting from a range of the -number of factors- values without excluding the solution's physical validity. A range of solutions were examined with different number of factors (3-8) in each case, and the solution of meaningful sources was selected. A limitation of PMF is that if factors number increases from the optimal, some factor profiles are split to profiles with no physical meaning, while the rotational instability of the solution increases significantly. The two final steps were the bootstrap and Fpeak runs in order to examine the stability and the rotational ambiguity of the solution, respectively.

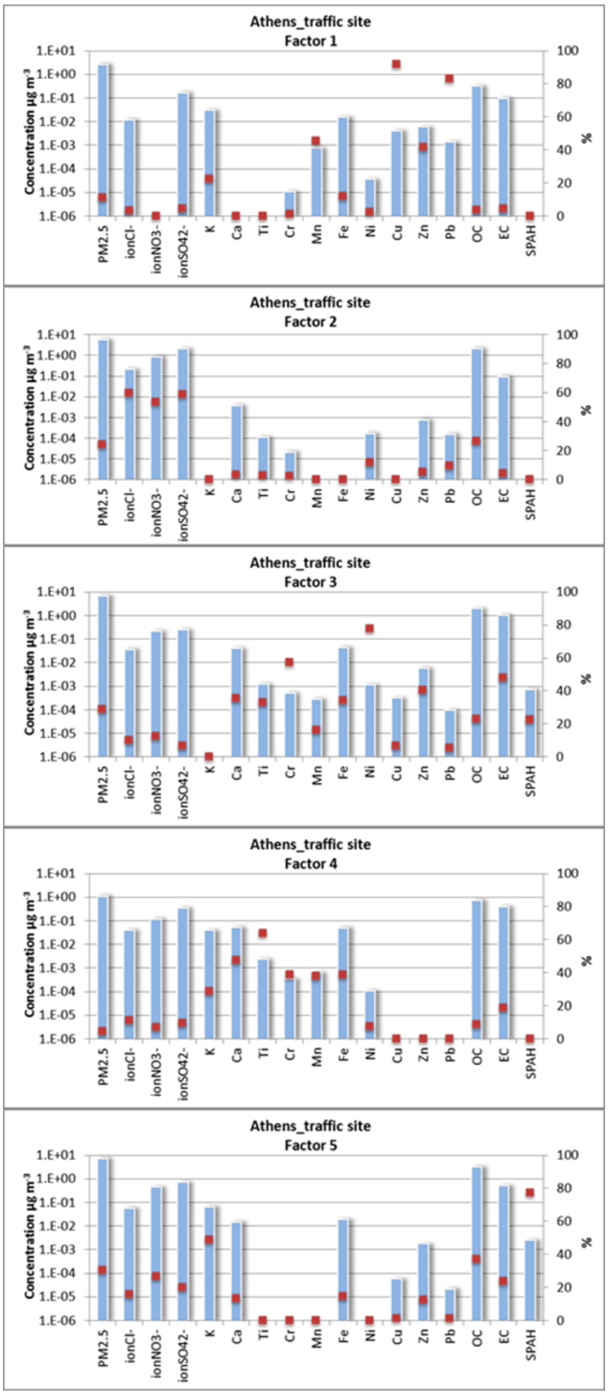
3.3 PMF Results

There was a good correlation between the model-predicted and the real PM2.5 mass (r>0.8) in all cases. In the following the results from PMF application for each site/city are presented.

3.3.1 PMF results for Athens

3.3.1.a Athens traffic site

PMF resulted in five sources/groups of sources for the traffic site in Athens. Figures 3.1a-e present the factor profiles in μg m^{-3} and % contribution. Figure 3.2 presents the % contribution of the five factors/sources to the measured PM2.5 concentration.



Figures 3.1.a-e. Factor profiles in μg m^{-3} and % contribution for the traffic site in Athens.

Factor 1 is strongly associated with **Cu, Zn, Pb** which are tracers of **industrial emissions o**r/and **non-exhaust traffic emissions** (e.g. brake and tire wear, combustion of lubricating oil). This factor contributes up to **11.4 %** of the measured PM2.5 at the traffic site of Athens.

Factor 2 is characterized by high percentages (>50%) of **chloride**, **nitrate** and **sulfate ions** as well as an amount of **organic carbon** (25%). which reveal a **secondary aerosol** source probably mixed with a **sea salt** source. In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for **24.5%** of the measured PM2.5 at the traffic site of Athens.

The high shares of **Ca, Fe, Ti** in **Factor 3** imply the crustal source origin of particles (natural source), combined with anthropogenic dust sources such as elemental materials emitted from vehicles brake pads, tires and mechanical parts and comprises the **mineral-road dust** source (Waked et al., 2014; Lucarelli et al., 2004). The ratio of OC/EC is lower than 0.7, which accordingly to previous studies (El Haddad et al., 2009; Amato et al., 2011; Waked et al., 2014) reveals traffic exhausts emissions. Thus, this **traffic-related factor** contributes to **28.8 %** of the measured PM2.5 at the traffic site of Athens.

F**actor 4** reveals a natural-origin source of **mineral dust** as it is traced by significant percentages of **Ca, K, Ti, Fe**. The contribution of this source to the measured PM2.5 levels at the traffic site is low (**4.8%**).

Finally, **Factor 5** is strongly associated with **PAHs, K and OC** which are characteristic **biomass burning/combustion** tracers. While carbonaceous fractions are major components in combustion-related sources, the OC/EC ratio is considered as a robust diagnostic of biomass burning. Indeed, the observed ratio value lies within the range (2-6) found in other Mediterranean cities (Amato et al., 2016) while also being observed in wood burning emissions (Fine et al., 2001). It is worth mentioning that since 2010, Greek citizens have used alternative heating fuels, such as wood, due to the economic crisis and the increased price of diesel oil. This factor accounts for the quite high percentage of **30.5%** of the measured PM2.5, which is expected during winter time (domestic heating) (Sarigiannis et al., 2014; Saraga et al., 2015).

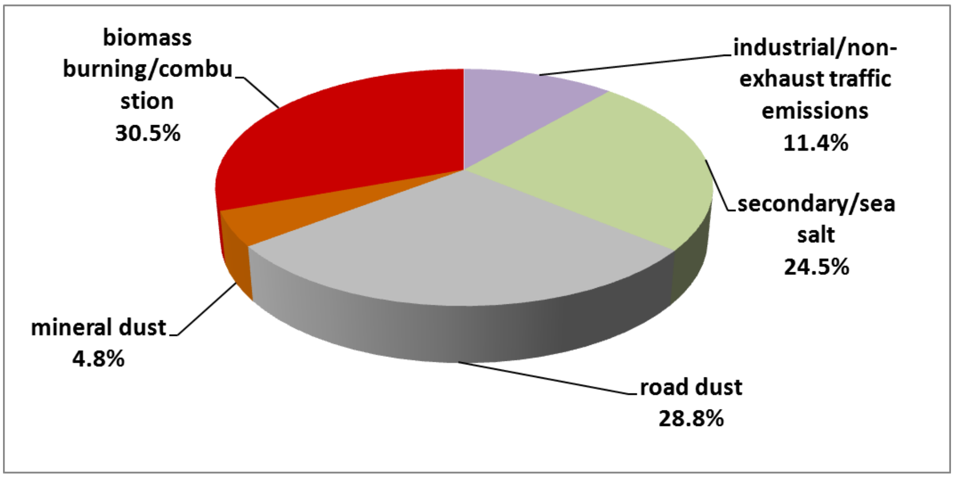
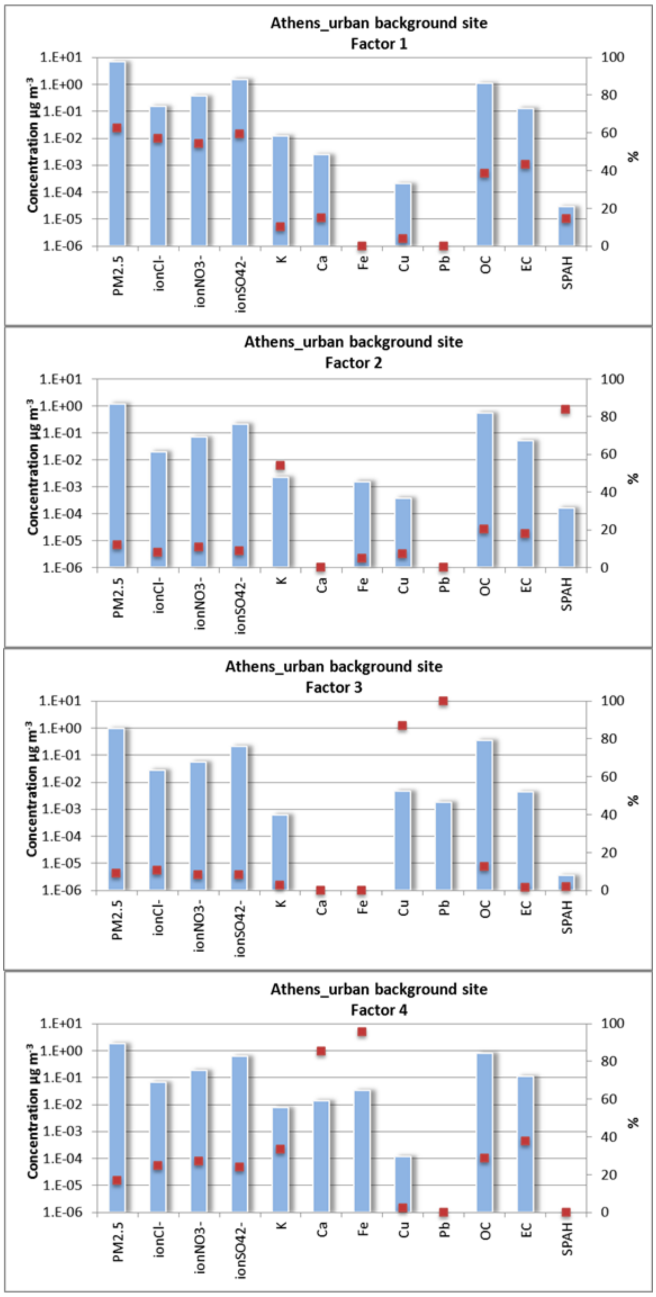


Figure 3.2. % contribution of the five factors/sources to the measured PM2.5 concentration for the traffic site in Athens.

3.3.1.b Athens urban background site

PMF resulted in four sources/groups of sources for the urban background site in Athens. Figures 3.3a-d present the factor profiles in μg m^{-3} and % contribution. Figure 3.4 presents the % contribution of the four factors/sources to the measured PM2.5 concentration.



Figures 3.3a-d. Factor profiles in μg m^{-3} and % contribution for the urban background site in Athens.

Factor 1 corresponds to a mixed traffic: the high percentages (>50%) of **chloride**, **nitrate** and **sulfate ions** reveal a **secondary aerosol** source mixed with a **sea salt** source. On the other hand, the presence of the two carbonaceous fractions (OC, EC) can be associated with **traffic** emissions. This mixed factor accounts for the quite high percentage of **60.2%** of the measured PM2.5 at the urban background site of Athens.

Factor 2 is associated with **PAHs, K** (84% and 54% respectively) and **OC** which are characteristic **biomass burning/combustion** tracers. This factor accounts for **11.9%** of the measured PM2.5 at the urban background site of Athens.

Factor 3 is characterized by high percentages of **Cu, Pb** and could correspond to **non-exhaust traffic emissions**. Its contribution to PM2.5 is **8.1**%.

Factor 4 reveals a source of crustal origin (**Ca, K, Ti**) enriched with traffic tracers (**EC, OC**) and therefore is corresponded to **road dust** (Waked et al., 2014; Lucarelli et al., 2004). This factor contributes to **19.5 %** of the measured PM2.5 at the urban background site of Athens.

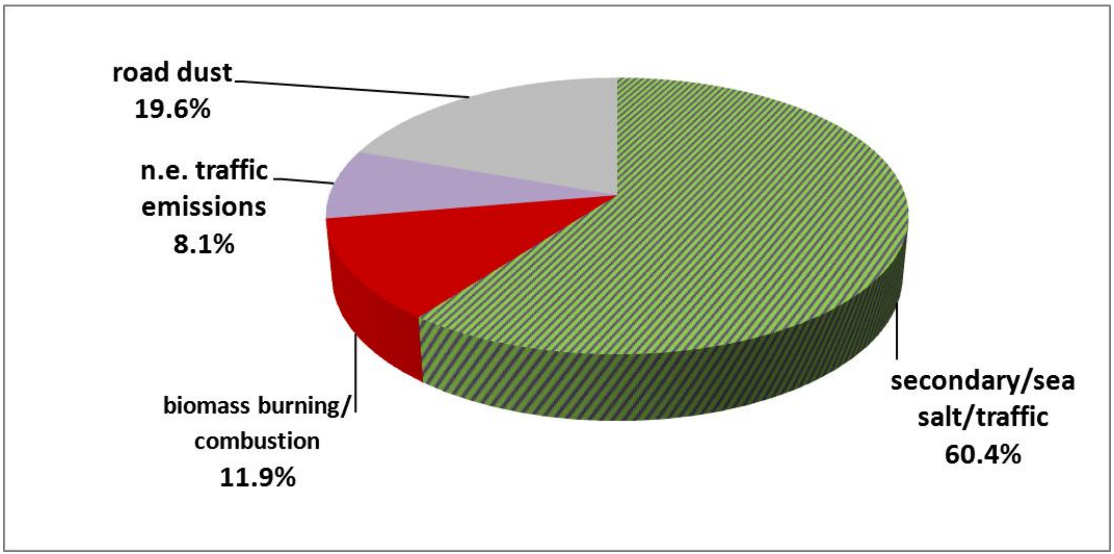
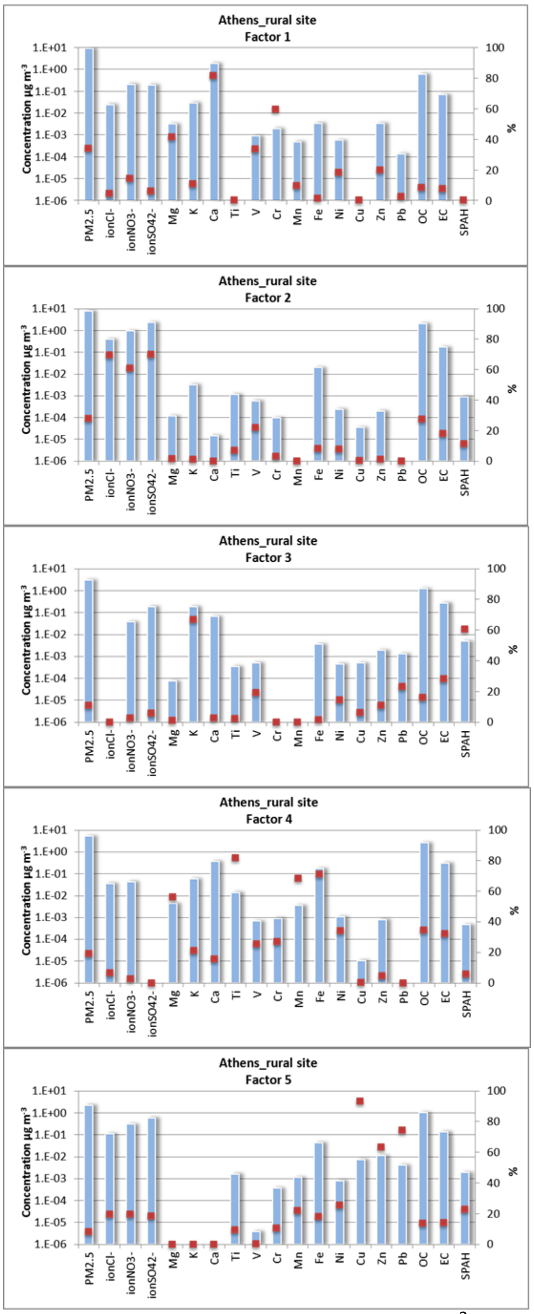


Figure 3.4. % contribution of the four factors/sources to the measured PM2.5 concentration for the urban background site in Athens.

3.3.1.c Athens rural site

PMF resulted in **five sources/groups of sources** for the **rural** site in Athens. Figures 3.5a-e present the factor profiles in μgm^{-3} and % contribution. Figure 3.6 presents the % contribution of the five factors/sources to the measured PM2.5 concentration.



Figures 3.5a-d. Factor profiles in μg m^{-3} and % contribution for the rural site in Athens.

Factor 1 reveals a natural-origin source of **mineral dust** as it is traced by significant percentages of **Ca and Mg**. The contribution of this source to the measured PM2.5 levels at the rural site is **19%**. It should be noticed that the crustal source appears to be split into two dust-related sources: one of the natural mineral source (Factor 1) and another of road dust (Factor 4).

Factor 2 is characterized by high percentages (>50%) of **chloride**, **nitrate** and **sulfate ions** as well as an amount of **organic carbon** (25%). which reveal a **secondary aerosol** source probably mixed with a **sea salt** source. In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for the **28%** of the measured PM2.5 at the rural site of Athens.

F**actor 3** is strongly associated with **PAHs and K** which are characteristic **biomass burning/combustion** tracers. This factor accounts for the **10.9%** of the measured PM2.5 and can be both related to domestic heating and agricultural activities.

Factor 4 reveals the crustal origin of the source (**Ca, Ti, Mg**) enriched with traffic/industrial emission tracers (**Fe, EC, OC, Mn, Ni**) and therefore is matched to **road dust** (Waked et al., 2014; Lucarelli et al., 2004). This factor contributes for **34.1 %** of the measured PM2.5 at the rural site of Athens.

Finally, **Factor 5** is strongly associated with **Cu, Zn, Pb** which are tracers of **industrial emissions o**r/and **non-exhaust traffic emissions** (e.g. brake and tire wear, combustion of lubricating oil). This factor contributes the **8.1 %** of the measured PM2.5 at the rural site of Athens.

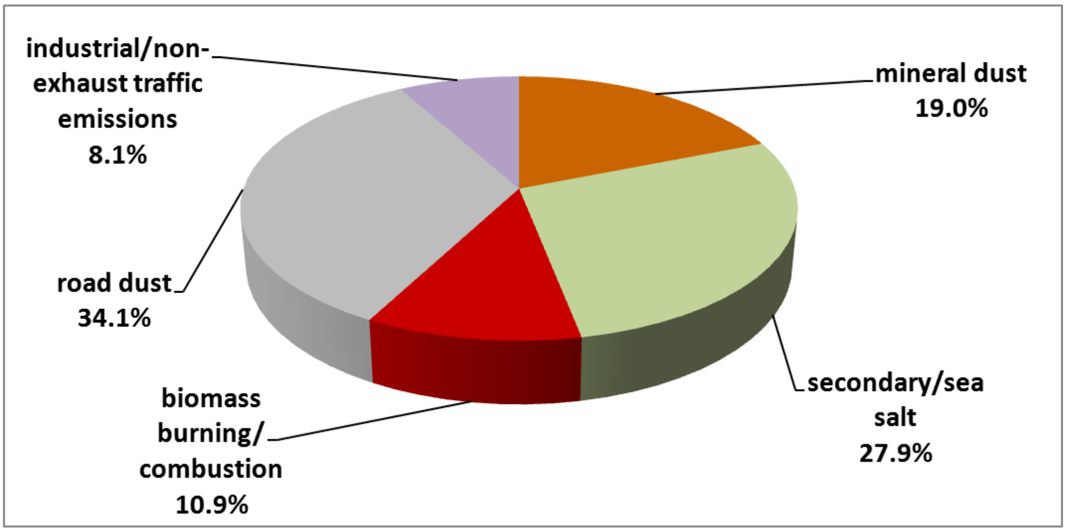
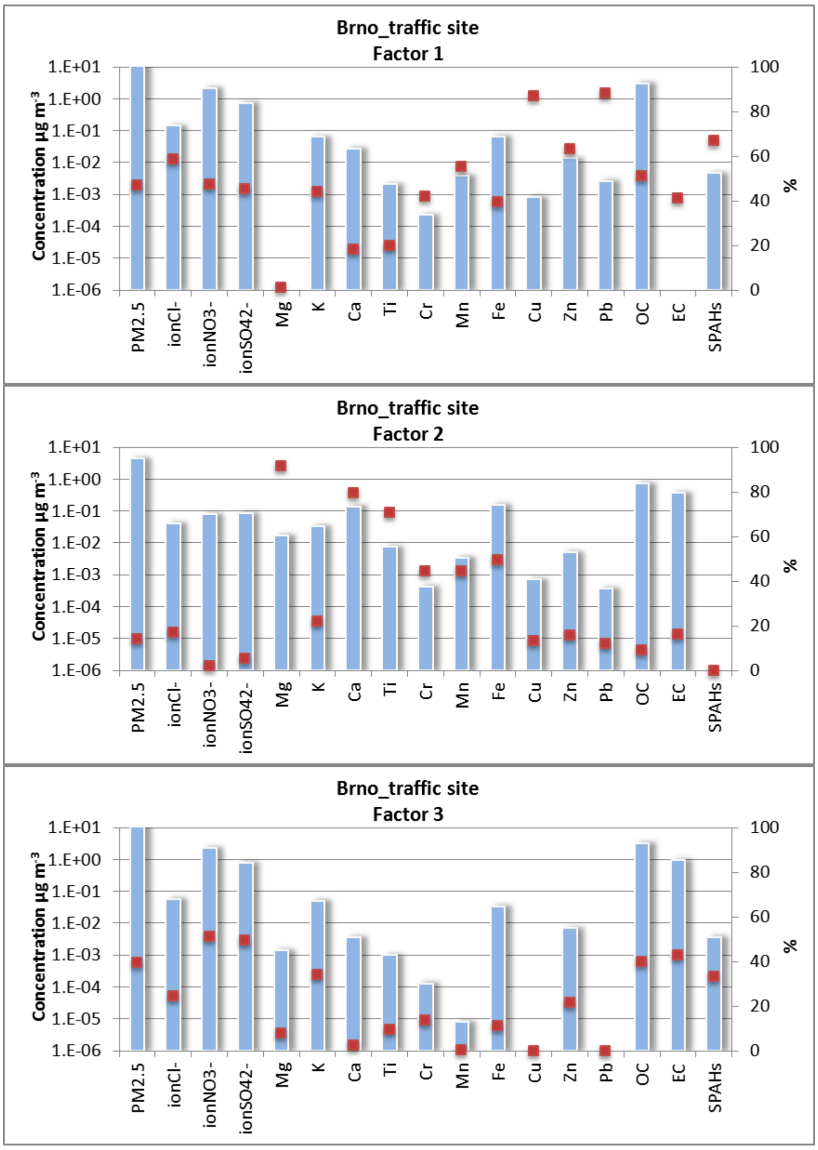


Figure 3.6. % contribution of the five factors/sources to the measured PM2.5 concentration for the rural site in Athens.

3.3.2 PMF results for Brno

3.3.2.a Brno traffic site

PMF resulted in three sources/groups of sources for the traffic site in Brno. Figures 3.7a-c present the factor profiles in μg m^{-3} and % contribution. Figure 3.8 presents the % contribution of the three factors/sources to the measured PM2.5 concentration.



Figures 3.7a-c. Factor profiles in μg m^{-3} and % contribution for the traffic site in Brno.

Factor 1 is strongly associated with **Cu, Zn, Pb**, **OC, EC, Fe, Mn, K** which are tracers of **exhaust** and **non-exhaust traffic emissions**. The presence of **nitrate** and **sulfate** ions reveals that this factor may also include and **secondary aerosol source.** In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for the **46.7%** of the measured PM2.5 at the traffic site of Brno.

Factor 2 reveals the crustal origin of the source (**Ca, Ti, Mg, K, Fe**) and corresponds to **mineral dust**. This factor contributes for **13.9 %** of the measured PM2.5 at the traffic site of Brno.

Finally, **Factor 3** is associated with **PAHs, K, EC and OC** which are characteristic **biomass burning/combustion** tracers. It also includes a part of a secondary aerosol source (**nitrate, sulfate**). This factor accounts for the quite high percentage of **39.4%** of the measured PM2.5, which is expected during winter time (domestic heating).

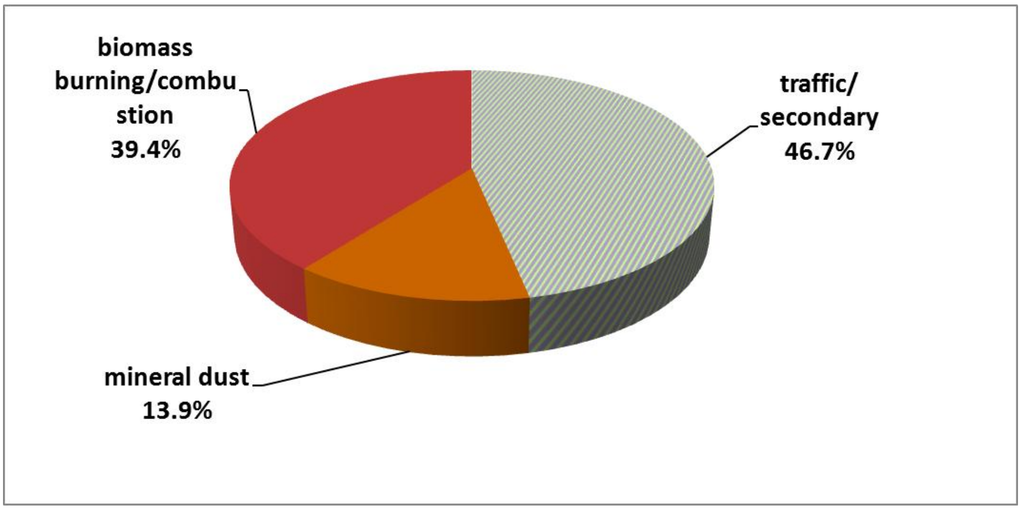
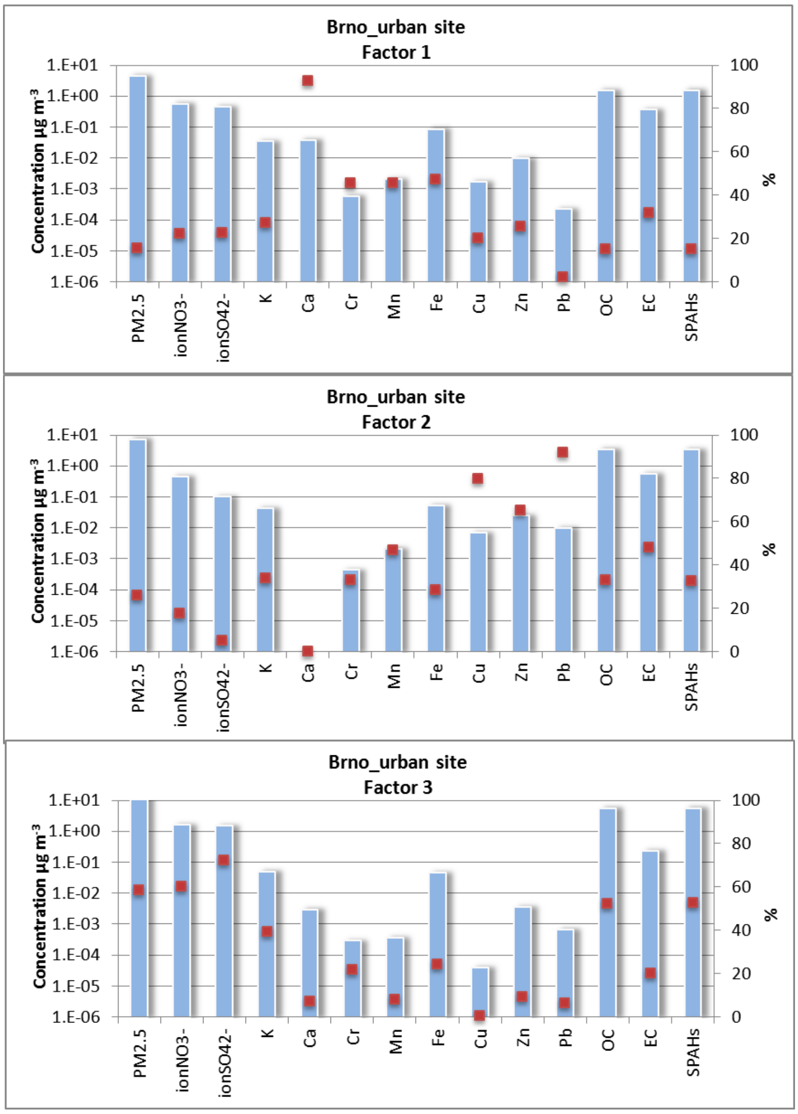


Figure 3.8. % contribution of the three factors/sources to the measured PM2.5 concentration for the traffic site in Brno.

3.3.2.b Brno urban background site

PMF resulted in **three sources/groups of sources** for the **urban** background site in Brno. Figures 3.9a-c present the factor profiles in μg m^{-3} and % contribution. Figure 3.10 presents the % contribution of the three factors/sources to the measured PM2.5 concentration.



Figures 3.9a-c. Factor profiles in μg m^{-3} and % contribution for the urban background site in Brno.

Factor 1 reveals the crustal origin of the source (**Ca, Ti, Mg**) enriched with traffic/industrial emission tracers (**Fe, EC, Zn, Cu**) and therefore is matched to **road dust** (Waked et al., 2014; Lucarelli et al., 2004). This factor contributes for **15.6 %** of the measured PM2.5 at the urban background site of Brno.

Factor 2 is strongly associated with **Zn, Pb**, **OC, EC, Fe, Mn, PAHs, K** which are tracers of **exhaust** and **non-exhaust traffic emissions**. This factor accounts for the **25.7%** of the measured PM2.5 at the urban background site of Brno.

Finally, **Factor 3** is associated with **PAHs, K, EC and OC** which are characteristic **biomass burning/combustion** tracers (Amato et al., 2016; Fine et al., 2001). It also includes a part of a secondary aerosol source (**nitrate, sulfate**). This factor accounts for the quite high percentage of **58.7%** of the measured PM2.5, which is expected during winter time (domestic heating).

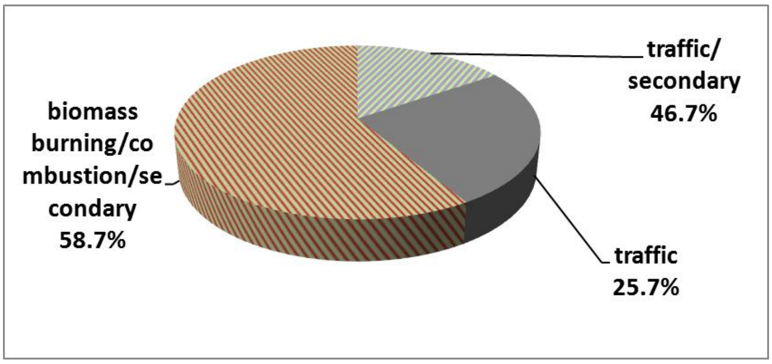
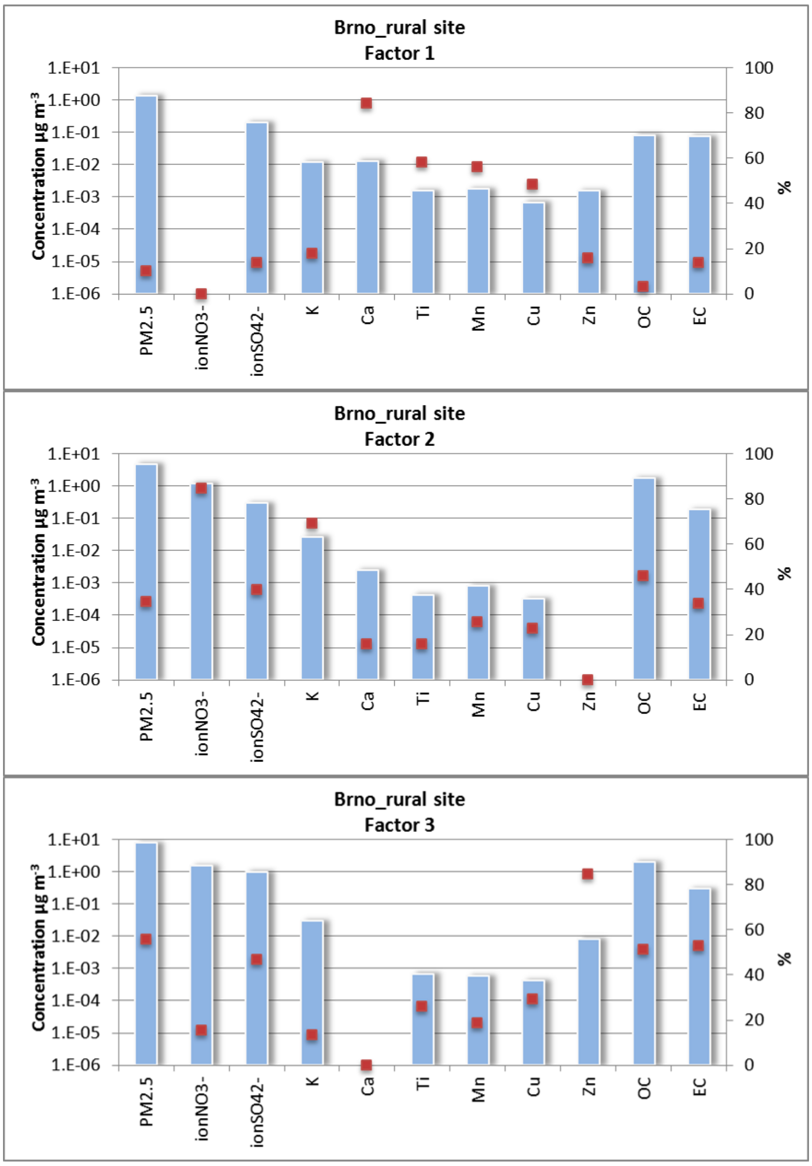


Figure 3.10. % contribution of the three factors/sources to the measured PM2.5 concentration for the urban background site in Brno.

3.3.2.c Brno rural site

PMF resulted in three sources/groups of sources for the rural site in Brno. Figures 3.11a-c present the factor profiles in μgm^{-3} and % contribution. Figure 3.12 presents the % contribution of the three factors/sources to the measured PM2.5 concentration.



Figures 3.11a-c. Factor profiles in μg m^{-3} and % contribution for the rural site in Brno.

Factor 1 reveals the crustal origin of the source (**Ca, Ti**) and corresponds to **mineral dust** combined with industrial emissions (**Mn, Cu**). This factor contributes for **9.9%** of the measured PM2.5 at the rural site of Brno.

Factor 2 is associated with **K, EC and OC** which are characteristic **combustion** tracers. It also includes a part of a secondary aerosol source (**nitrate, sulfate**). This factor accounts for the **34.3%** of the measured PM2.5.

Factor 3 is strongly associated with **Zn, OC, EC, Cu, sulfate** which link to a **traffic-related source** (El Haddad et al., 2009; Amato et al., 2011; Waked et al., 2014). This factor accounts for the **55.8%** of the measured PM2.5 at the rural site of Brno.

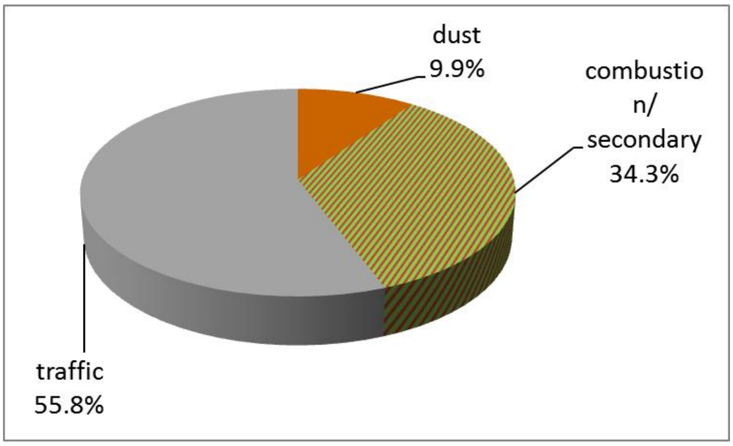
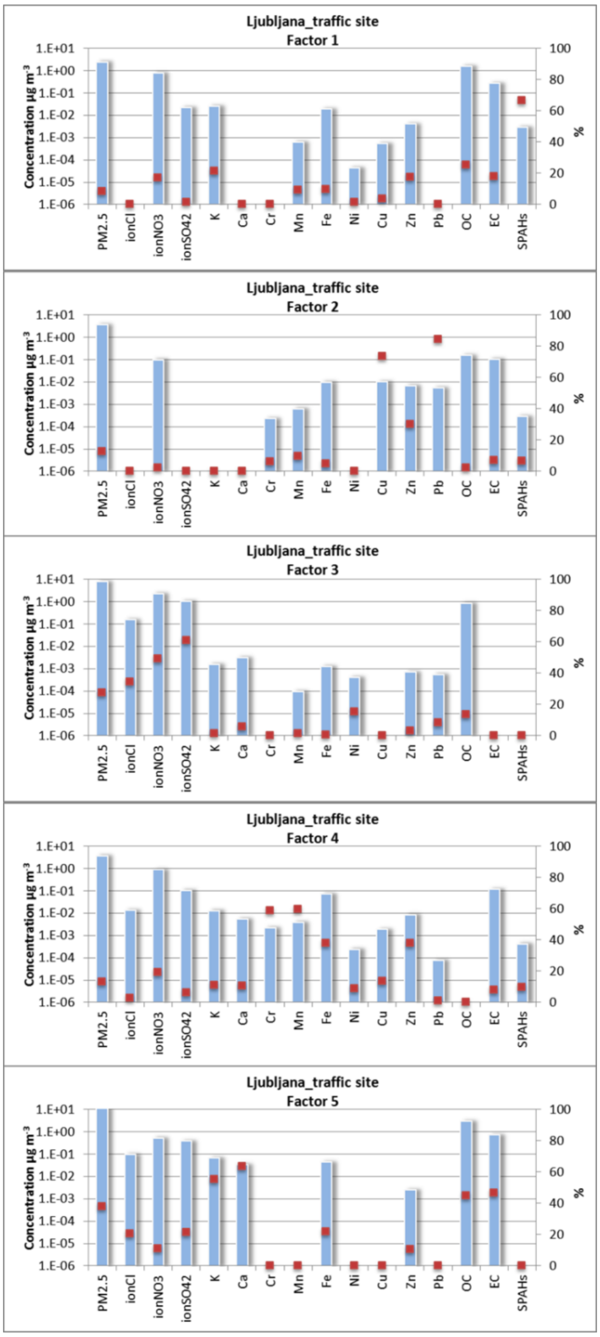


Figure 3.12. % contribution of the three factors/sources to the measured PM2.5 concentration for the rural site in Brno.

3.3.3 PMF results for Ljubljana

3.3.3.a Ljubljana traffic site

PMF resulted in five sources/groups of sources for the traffic site in Ljubljana. Figures 13 a-e present the factor profiles in μg m^{-3} and % contribution. Figure 3.14 presents the % contribution of the five factors/sources to the measured PM2.5 concentration.



Figures 3.13a-e. Factor profiles in μg m^{-3} and % contribution for the traffic site in Ljubljana.

Factor 1 is strongly associated with **PAHs**, as well as with **K, EC** and **OC** which are characteristic **biomass burning/combustion** tracers (Amato et al., 2016; Fine et al., 2001). This factor accounts for **8.6%** of the measured PM2.5.

Factor 2 is strongly associated with **Cu, Zn, Pb** which are tracers of **industrial emissions** or/and **non-exhaust traffic emissions** (e.g. brake and tire wear, combustion of lubricating oil). This factor contributes the **12.6 %** of the measured PM2.5 at the traffic site of Ljubljana.

Factor 3 is characterized by significant percentages of **chloride**, **nitrate** and **sulfate ions** which reveal a **secondary aerosol** source probably mixed with a **sea salt** source. In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for the **27.5%** of the measured PM2.5 at the traffic site of Ljubljana.

Factor 4, similarly to factor 2, is related with tracers of **industrial emissions/ fuel oil**/ **non-exhaust traffic emissions** and accounts for **13.2%** of the measured PM2.5.

Factor 5 reveals the crustal origin of the source (**Ca, K**) enriched with traffic tracers (**Fe, EC, OC**) and therefore is matched to **road dust** (Waked et al., 2014; Lucarelli et al., 2004). This factor contributes for **38.1 %** of the measured PM2.5 at the traffic site of Ljubljana.

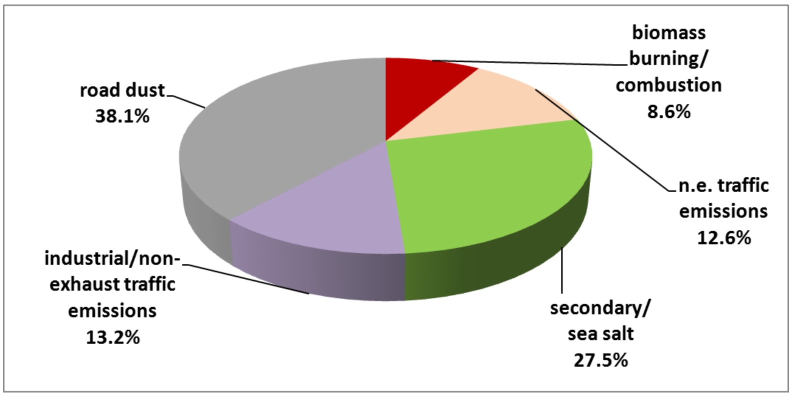
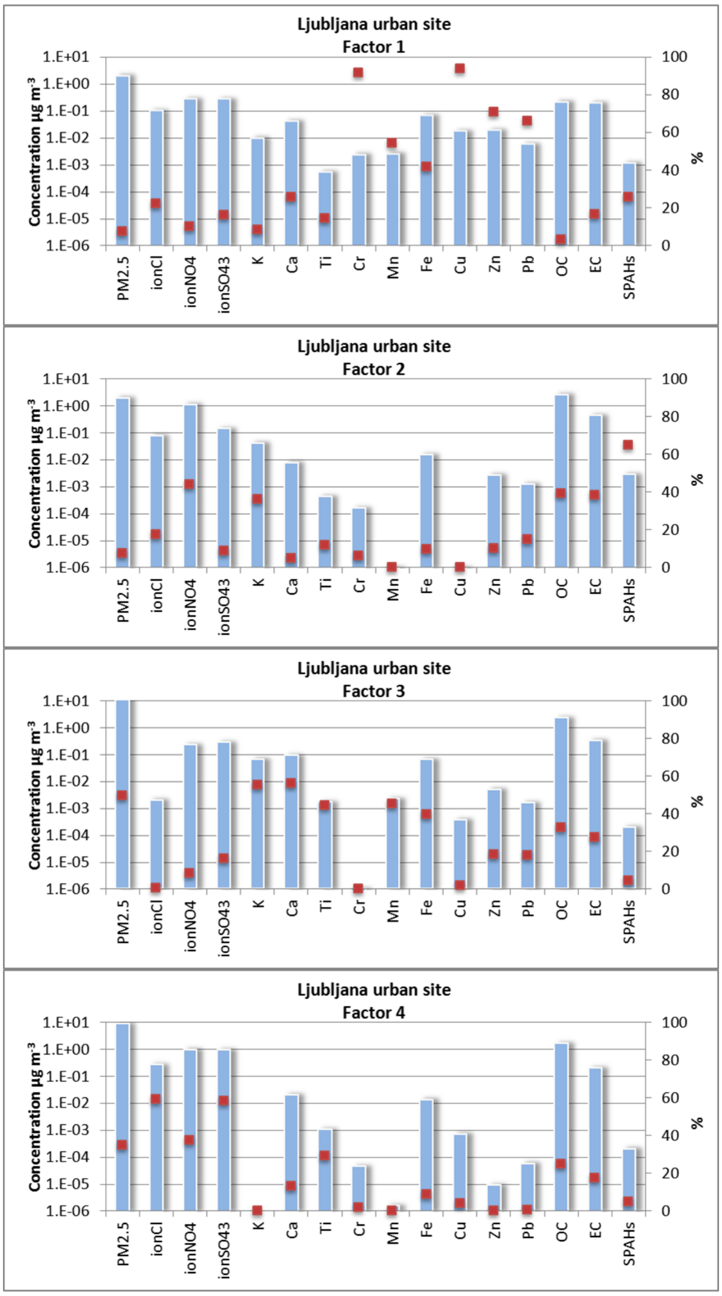


Figure 3.14. % contribution of the five factors/sources to the measured PM2.5 concentration for the traffic site in Ljubljana.

3.3.3.b Ljubljana background site

PMF resulted in **four sources/groups of sources** for the urban background site in Ljubljana. Figures 3.15a-d present the factor profiles in μg m^{-3} and % contribution. Figure 3.16 presents the % contribution of the four factors/sources to the measured PM2.5 concentration.



Figures 3.15a-d. Factor profiles in μg m^{-3} and % contribution for the urban background site in Ljubljana.

Factor 1 is related with tracers of **industrial emissions/ non-exhaust traffic emissions (Cr, Mn, Fe, Cu, Zn , Pb)** and accounts for **7.7 %** of the measured PM2.5.

Factor 2 is strongly associated with **PAHs**, as well as with **K, EC** and **OC** which are characteristic **biomass burning/combustion** tracers. This factor accounts for **7.6%** of the measured PM2.5.

Factor 3 reveals the crustal origin of the source (**Ca, K**) enriched with traffic tracers (**Fe, EC, OC**) and therefore is matched to **road dust** (Waked et al., 2014; Lucarelli et al., 2004). This factor contributes for **49.9%** of the measured PM2.5 at the urban background site of Ljubljana.

Factor 4 is characterized by significant percentages of **chloride**, **nitrate** and **sulfate ions** which reveal a **secondary aerosol** source probably mixed with a **sea salt** source. In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for the **34.8%** of the measured PM2.5 at the urban background site of Ljubljana.

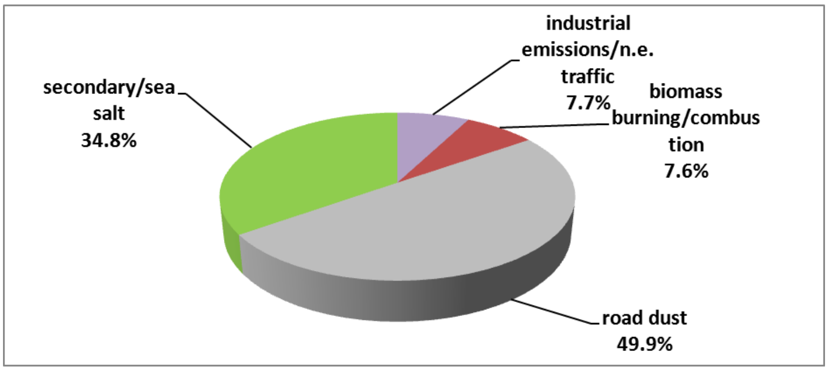
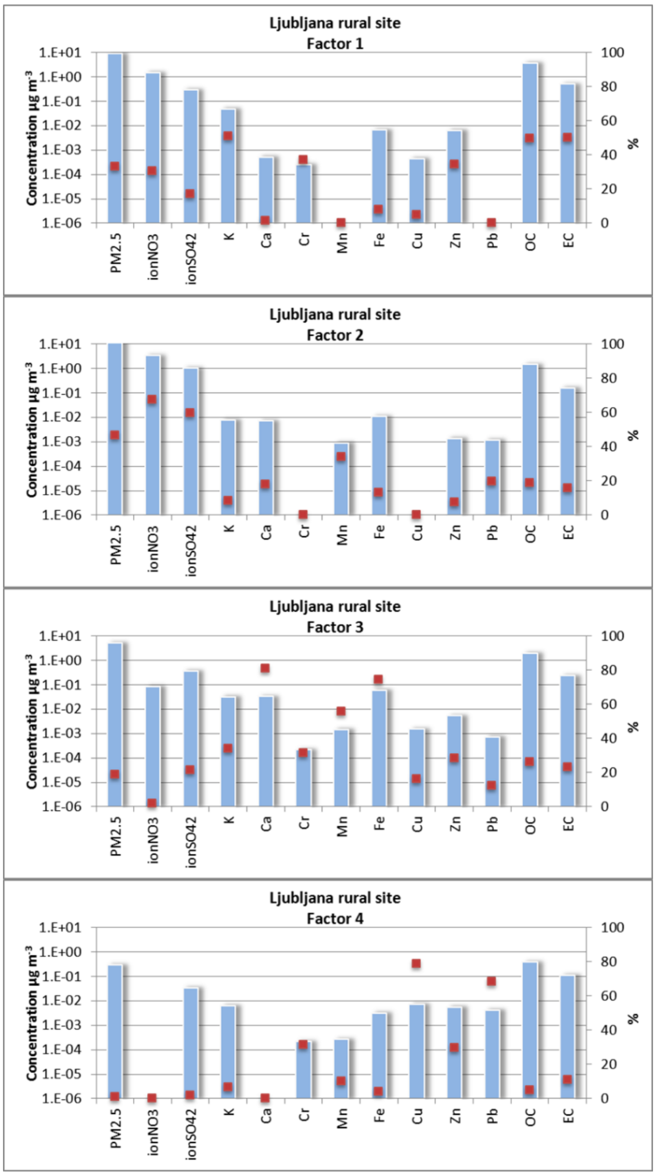


Figure 3.16. % contribution of the four factors/sources to the measured PM2.5 concentration for the urban background site in Ljubljana.

3.3.3.c Ljubljana rural site

PMF resulted in **four sources/groups of sources** for the **rural** site in Ljubljana. Figures 3.17a-d present the factor profiles in μg m^{-3} and % contribution. Figure 3.18 presents the % contribution of the four factors/sources to the measured PM2.5 concentration.



Figures 3.17a-d. Factor profiles in μg m^{-3} and % contribution for the rural site in Ljubljana.

Factor 1 is strongly characterized by significant percentages of **K, EC** and **OC** as well as contribution of **Zn** and **Cr**. This factor corresponds to **combustion-related** source or group of sources (including **domestic heating, traffic** and **agricultural activities**) while accounting for **45.6%** of the measured PM2.5.

Factor 2 is characterized by significant percentages of **nitrate** and **sulfate ions** which reveal a **secondary aerosol** source probably mixed with a **sea salt** source. In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for the **2.3%** of the measured PM2.5 at the rural site of Ljubljana.

Factor 3 reveals the crustal origin of the source (**Ca, K, Mn**) enriched with traffic tracers (**Fe, EC, OC**) and therefore is matched to **road dust** (Waked et al., 2014; Lucarelli et al., 2004). This factor contributes for **41.9%** of the measured PM2.5 at the rural site of Ljubljana.

Factor 4 is related with tracers of **industrial emissions/ non-exhaust traffic emissions (Cr, Cu, Zn , Pb)** and accounts for **10.2 %** of the measured PM2.5.

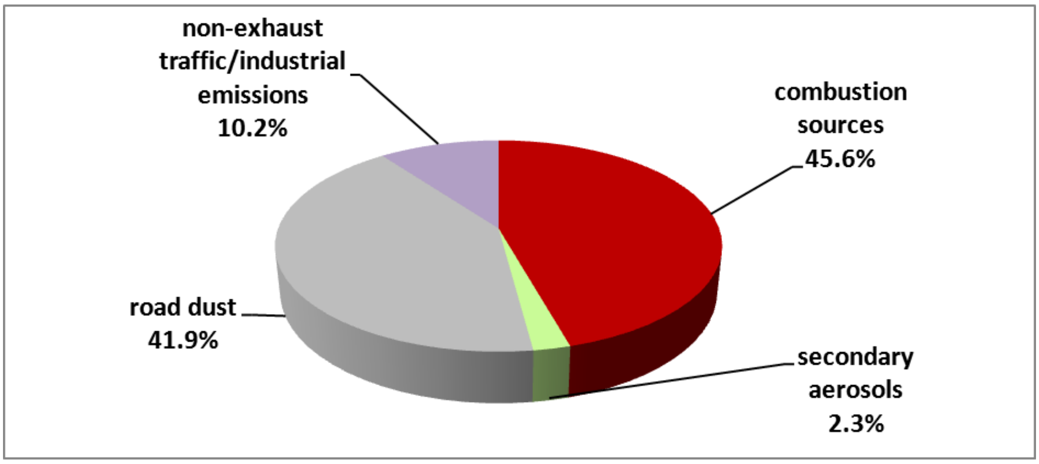
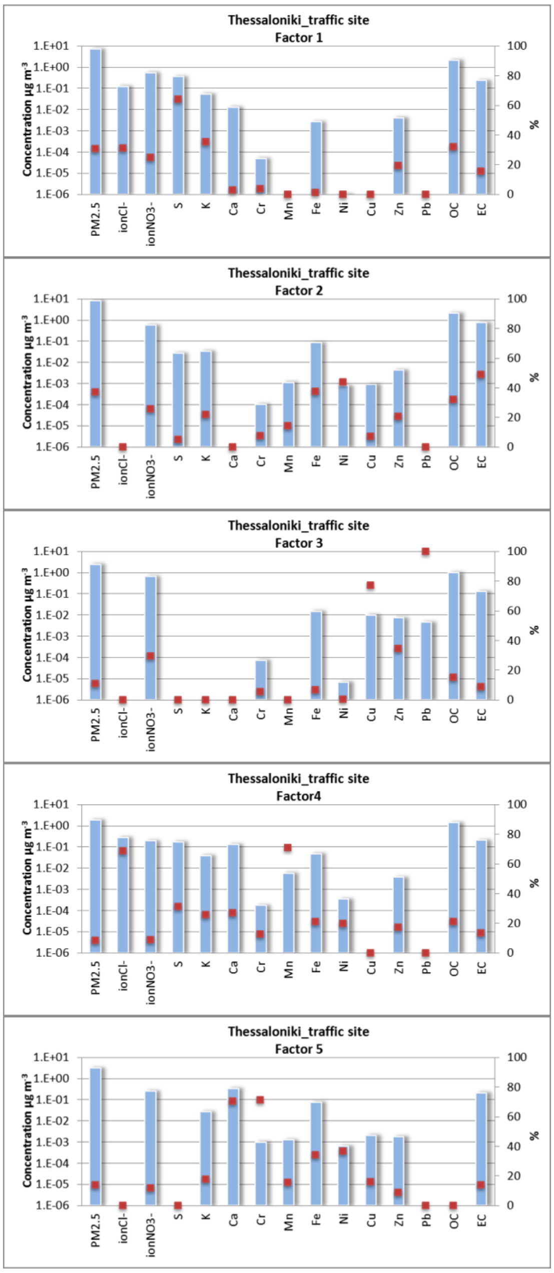


Figure 3.18. % contribution of the four factors/sources to the measured PM2.5 concentration for the rural site in Ljubljana.

3.3.4 PMF results for Thessaloniki

3.3.4.a Thessaloniki traffic site

PMF resulted in **five sources/groups of sources** for the traffic site in Thessaloniki. Figures 3.19a-e present the factor profiles in μg m^{-3} and % contribution. Figure 3.20 presents the % contribution of the five factors/sources to the measured PM2.5 concentration.



Figures 3.19a-e. Factor profiles in μg m^{-3} and % contribution for the traffic site in Thessaloniki.

Factor 1 is strongly associated with **S, K and OC** which are characteristic **combustion** tracers. This factor accounts for the quite high percentage of **30.7%** of the measured PM2.5, which is expected during winter time (domestic heating).

Factor 2 corresponds to **traffic emissions** as it is characterized by EC, OC, Fe, Ni, Zn and nitrate. The ratio of OC/EC is lower than 0.7, which accordingly to previous studies (El Haddad et al., 2009; Amato et al., 2011; Waked et al., 2014) reveals traffic exhausts emissions. Further chemical analysis could possibly indicate that this factor includes shipping emissions from the port of the city. This factor accounts for the quite high percentage of **36.8%** of the measured PM2.5 at the traffic site of Thessaloniki.

Factor 3 is strongly associated with **Cu, Zn, Pb** which are tracers of **industrial emissions o**r/and **non-exhaust traffic emissions** (e.g. brake and tire wear, combustion of lubricating oil). This factor contributes the **10.7 %** of the measured PM2.5 at the traffic site of Thessaloniki.

Factor 4 is characterized by high percentages (>50%) of **chloride** which is one of the **sea salt** tracers. This factor accounts for the **8.1%** of the measured PM2.5 at the traffic site of Thessaloniki.

F**actor 5** reveals a natural-origin source of **mineral dust** as it is traced by significant percentages of **Ca, K, Fe**. The contribution of this source to the measured PM2.5 levels at the traffic site is **13.7%**.

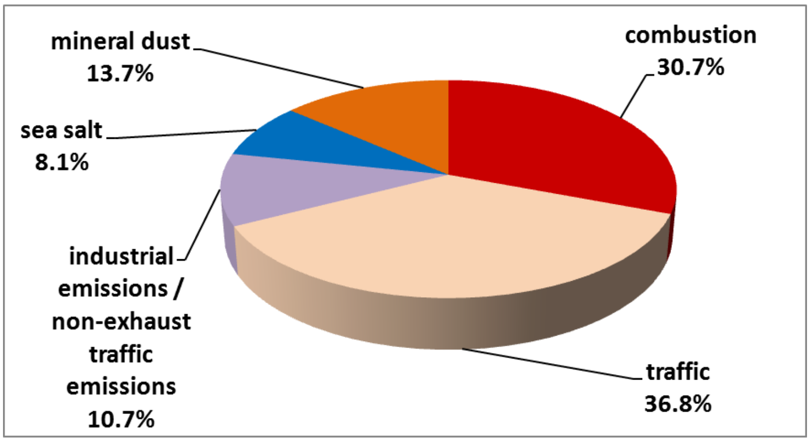
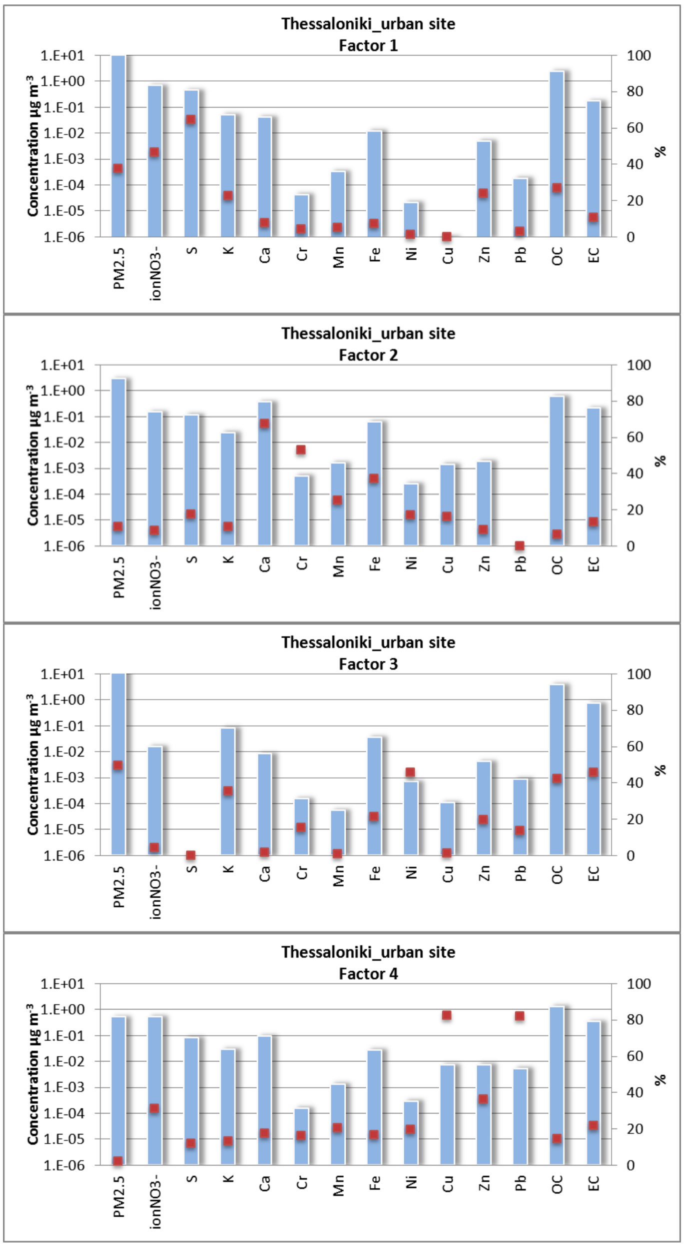


Figure 3.20. % contribution of the five factors/sources to the measured PM2.5 concentration for the traffic site of Thessaloniki.

3.3.4.b Thessaloniki background site

PMF resulted in four sources/groups of sources for the urban background site in Thessaloniki. Figures 3.21a-d present the factor profiles in μg m^{-3} and % contribution. Figure 3.22 presents the % contribution of the four factors/sources to the measured PM2.5 concentration.



Figures 3.21a-d. Factor profiles in μg m^{-3} and % contribution for the urban background site in Thessaloniki.

Factor 1 is characterized by high percentages (>50%) of **nitrate** and **S** as well as an amount of **organic carbon** (27%) which reveal a **secondary aerosol** source. This factor accounts for the **37.7%** of the measured PM2.5 at the urban background site of Thessaloniki.

F**actor 2** reveals a natural-origin source of **mineral dust** as it is traced by significant percentages of **Ca** and **Fe**. The contribution of this source to the measured PM2.5 levels at is **10.8%**.

Factor 3 is strongly characterized by significant percentages of **K, EC** and **OC** as well as contribution of **Ni**. This factor corresponds to **combustion-related** source or group of sources (including **domestic heating, traffic** and **agricultural activities**) while accounting for **49.6%** of the measured PM2.5.

Factor 4 is associated with **Cu, Zn, Pb** which are tracers of **industrial emissions o**r/and **non-exhaust traffic emissions** (e.g. brake and tire wear, combustion of lubricating oil). This factor's contribution is underestimated (**1.9 %)** probably because traffic-source has been presented also in Factor 3.

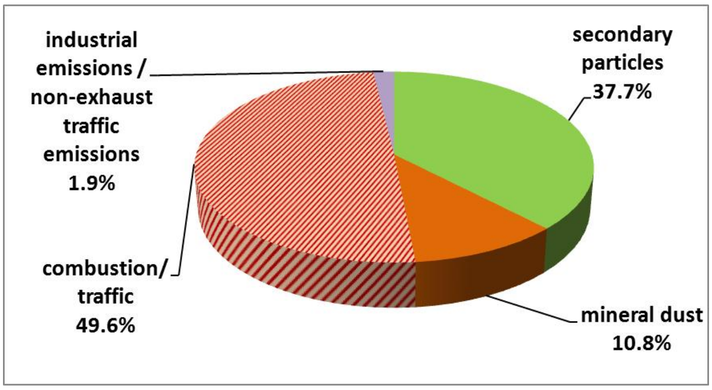
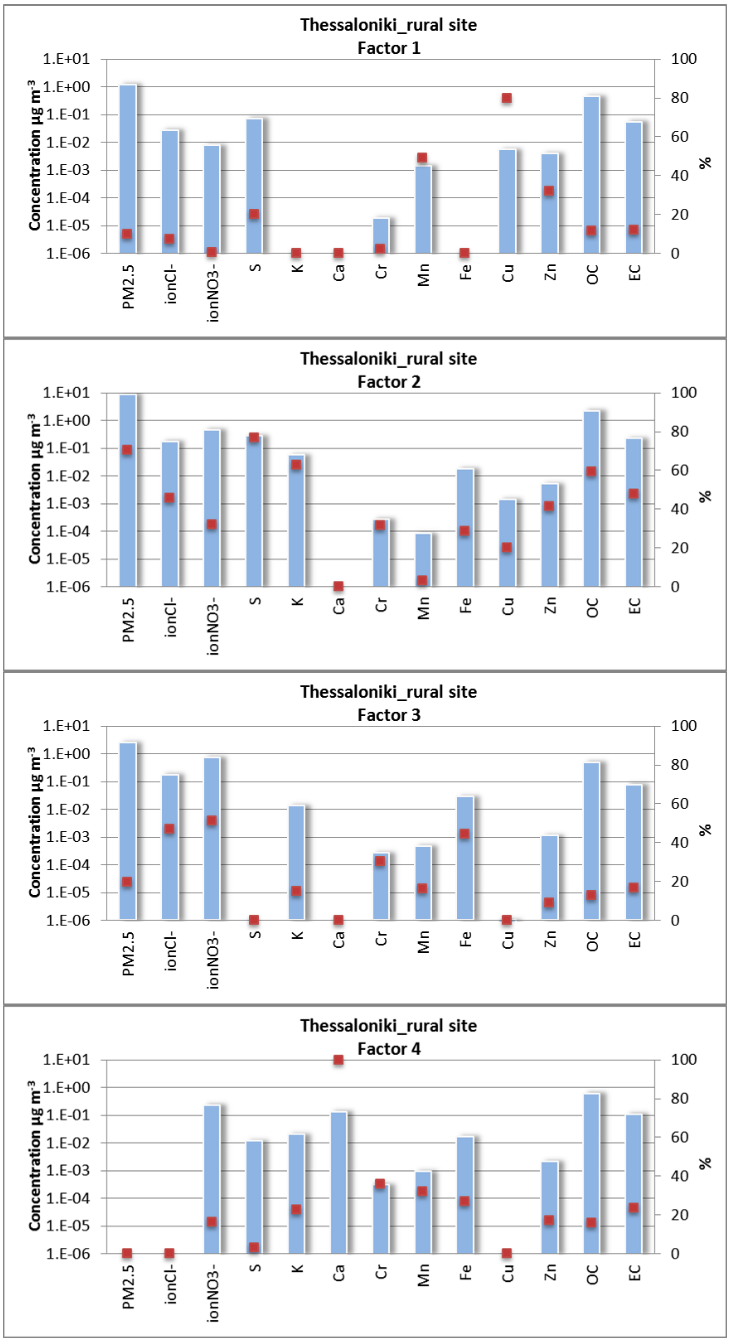


Figure 3.22. % contribution of the four factors/sources to the measured PM2.5 concentration for the urban background site of Thessaloniki.

3.3.4.c Thessaloniki rural site

PMF resulted in four sources/groups of sources for the rural site in Thessaloniki. Figures 3.23a-d present the factor profiles in μg m^{-3} and % contribution. Figure 3.24 presents the % contribution of the four factors/sources to the measured PM2.5 concentration.



Figures 3.23a-d. Factor profiles in μg m^{-3} and % contribution for the rural site in Thessaloniki.

Factor 1 is associated with **Cu, Zn, Mn** which are tracers of **industrial emissions o**r/and **non-exhaust traffic emissions** (e.g. brake and tire wear, combustion of lubricating oil). This factor's contribution to PM2.5 concentration is **11 %.**

Factor 2 is strongly characterized by significant percentages of **K, EC** and **OC** as well as contribution of **S**. This factor corresponds to **combustion-related** source or group of sources (including **domestic heating, traffic** and **agricultural activities**) while accounting for **52.9%** of the measured PM2.5.

Factor 3 is characterized by high percentages (>50%) of **chloride** and **nitrate** which reveal a **secondary aerosol** source probably mixed with a **sea salt** source. In presence of specific tracers (e.g. ammonium, sodium), this factor would be split into two sources. This factor accounts for the **28.8%** of the measured PM2.5 at the rural site of Thessaloniki.

F**actor 4** reveals a natural-origin source of **mineral dust** as it is traced by significant percentages of **Ca, K** and **Fe**. The contribution of this source to the measured PM2.5 levels at is **7.4%**.

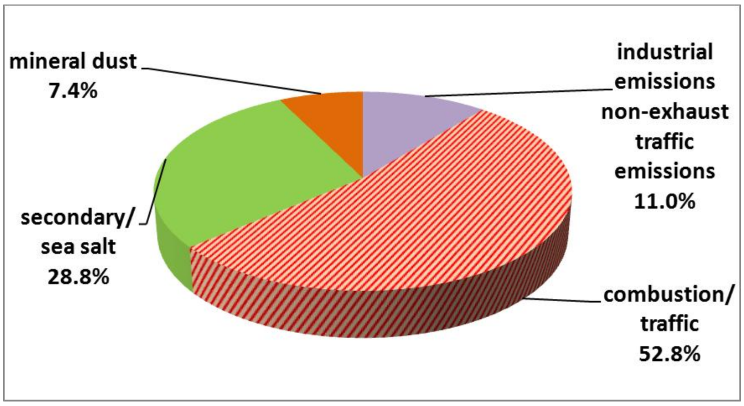


Figure 3.24. % contribution of the four factors/sources to the measured PM2.5 concentration for the rural site of Thessaloniki.

3.3.5 Overview of PMF results

A well-known weakness of PMF is that it is not always a direct tool to distinguish individual sources, as the user has to distinguish between factors explaining emission sources, in which tracers sharing emission sources are grouped, and factors explaining formation/transformation processes (e.g. secondary nitrates and sulfates). In cases of concurrent emissions from different sources, the analysis can be rather complicated as the obtained factors represent combined sources or atmospheric processes rather than single emission sources.

An overview of the percentages of each source contribution to the measured PM2.5, as derived from PMF is presented in Table 3.2. Mixed sources (e.g. traffic non-exhaust and/or industrial emissions) are noticed with an asterisk. In most cases, secondary particles and sea salt sources were not distinguished. In presence of specific tracers (e.g. ammonium, sodium), this mixed source would be split into two sources. Similarly in a number of cases, the combustion-related source seems to include also traffic source, presenting quite high percentages.

Table 3.2. PMF % contribution of each source.

	source:	**traffic non-exhaust**	**traffic exhaust**	**secondary particles**	**sea salt**	**mineral/crustal/dust**	**road dust**	**industrial /fuel oil**	**biomass burning/ combustion**
city:	**sampling site:**								
Athens	traffic	11.4%*		24.5%		4.8%	28.8%	*	30.5%
	urban background	8.1% *		60.2%			19.5%	*	11.9%
	rural	8.1%*		28.0%		19.0%	34.1%	*	10.9%
Brno	traffic	46.7%*		*/**		13.9%			39.4%**
	urban background	25.7%		*		15.6%			58.7%*
	rural	55.8%		*		9.9%			34.3%*
Ljubljana	traffic	12.6%	*	27.5%			38.1%	13.2%	8.6%
	urban background	7.7%*	*	34.8%			49.9%	*	7.6%
	rural	10.2%*	*	2.3%			41.9%	*	45.6%*
Thessaloniki	traffic	*	36.8%		8.1%		13.7%	10.7%*	30.7%
	urban background	1.9%**	*	37.7%		10.8%		**	49.6%*
	rural	*	*	28.8%		7.4%		11.0%	52.9%*

4 Principal Component Analysis (PCA)

4.1 PCA application

The Principal Component Analysis (PCA) on the PM2.5 chemical analysis data was performed using the IBM SPSS 24.0 statistical software package (IBM, Armonk, NY, USA). The PCA transforms the set of intercorrelated variables into a set of independent uncorrelated variables, called principal components (PCs), which are linear combinations of the original variables. The first PC is the linear combination of PM species concentrations with maximal variance and explains or accounts for the maximum amount of the variability of the original variables. The second PC is the linear combination (uncorrelated with the first PC) that represents the next largest variability not already accounted for by the first PC. The third, fourth, etc., PCs are defined similarly. The eigenvectors of the correlation matrix must be linearly combined to form the source vectors. Typically, this is conducted by applying the “VARIMAX” rotation method, retaining PCs whose eigenvalues are larger than 1. The relationship between the PC (hereafter called “factor”) and the compound is indicated by the factor loadings and is related to the source emissions composition. The factor loadings are the coefficients of Pearson's correlation between the hypothetical sources and the compound in question. A loading close to (+1) indicates that the pollutant is highly and positively correlated with the source vector. On the contrary, a loading close to (-1) indicates that the pollutant is highly and negatively correlated with the source category. Absolute coefficients less than 0.60 have been omitted from the tables to provide a clearer picture of the groups of variables characterizing the single factors or sources. By critically evaluating the factor loadings, through a combined literature survey, an estimate of the main source responsible for each factor can be made.

In the present PCA analysis, the PMF input data bases were used. The same data pre-treatment was followed.

4.2 PCA results

Tables 4.1-4.4 present the results of the PCA (Varimax rotation) for PM2.5 chemical components for the three sites (traffic, urban background, rural) of **Athens, Brno, Ljubljana,** and **Thessaloniki**. The coefficients between factors and chemical species concentrations are reported. The % cumulative rotation sums of squared loadings varied between 65.3% and 94.5% and the number of components between 2 and 4.

Table 4.1. PCA results for Athens (traffic, urban background, rural site).

Athens_traffic site				
	Components (Factors):			
Compounds:	**1**	**2**	**3**	**4**
OC	**0.940**			
SPAH	**0.921**			
ionNO3-	**0.801**			
K	**0.796**	0.360		
ionSO42-	**0.670**			
Zn	**0.622**	0.362	0.499	
ionCl-	0.416			-0.363
Ti		**0.946**		
Ca		**0.943**		
Fe		**0.929**		
Mn		**0.857**	0.365	
EC	0.581	**0.655**		
Cu			**0.878**	
Pb			**0.873**	
Ni				**0.906**
Cr		0.499		**0.692**
source:	biomass burning/combustion/secondary	road dust	non-exhaust traffic	industrial/fuel oil combustion
Rotation Sums of Squared Loadings				
% of variance	*27.8%*	*27.6%*	*12.9%*	*11.3%*
Cumulative %	*27.8%*	*55.5%*	*68.4%*	***79.6%***

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown

Athens_urban background site				
	Components (Factors):			
Compounds:	**1**	**2**	**3**	**4**
Fe	**0.924**			
Ca	**0.900**			
K	**0.577**			0.423
OC		**0.926**		
EC		**0.805**		
ionSO42-		**0.697**		
Pb			**0.928**	
Cu			**0.876**	
SPAH	-0.437		-0.442	
ionCl-				**0.836**
ionNO3-				**0.647**
source:	mineral dust	traffic combustion	non-exhaust traffic	secondary particles/ sea salt
Rotation Sums of Squared Loadings				
% of variance	*21.2%*	*19.6%*	*17.3%*	*12.7%*
Cumulative %	*21.2%*	*40.7%*	*58.0%*	***70.7%***

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown

Athens_rural site				
	Components (Factors):			
Compounds:	**1**	**2**	**3**	**4**
Mn	**0.933**			
Fe	**0.929**			
Mg	**0.921**			
Ti	**0.900**			
Cr	**0.849**	0.328		
Ca	**0.786**	0.462		
V	**0.608**	0.333		0.384
OC	0.470	**0.812**		
EC	0.434	**0.806**		
SPAH		**0.771**		
K		**0.753**		
ionNO3-		0.571		0.535
Cu			**0.913**	
Zn		0.309	**0.871**	
Pb			**0.808**	
Ni	0.422		**0.566**	
ionCl-				**0.873**
ionSO42-		0.437		**0.649**
source:	mineral dust	biomass burning /combustion	non-exhaust traffic	secondary particles /sea salt
Rotation Sums of Squared Loadings				
% of variance	*32.9%*	*20.0%*	*16.1%*	*10.5%*
Cumulative %	*32.9%*	*52.9%*	*69.0%*	***79.5%***

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown

Table 4.2. PCA results for Brno (traffic, urban background, rural site).

Brno_traffic site					Brno_urban background site			Brno_rural site		
	Components (Factors):					Components (Factors):			Components (Factors):	
Compounds:	**1**	**2**	**3**	**4**	**Compounds:**	**1**	**2**	**Compounds:**	**1**	**2**
SPAH	**0.931**				**ionSO42-**	**0.935**		**OC**	**0.966**	
Pb	**0.914**				**SPAHs**	**0.888**	0.382	**K**	**0.958**	
K	**0.882**				**OC**	**0.888**	0.382	**EC**	**0.915**	
OC	**0.882**		0.352		**Pb**	**0.861**		**ionNO3-**	**0.887**	
Zn	**0.866**		0.305	0.302	**K**	**0.851**	0.372	**Zn**	**0.887**	
Mn	**0.724**	**0.604**			**Zn**	**0.839**	0.498	**Fe**	**0.858**	
Ca		**0.987**			**ionNO3-**	**0.724**	0.313	**ionSO42-**	**0.847**	-0.322
Ti		**0.969**			**Mn**	**0.680**	**0.586**	**Cu**	0.581	
Mg		**0.936**			**ionCl-**	**0.672**	0.476	**Ca**		**0.909**
Fe	0.402	**0.851**			**Cu**	**0.582**	0.521	**Ti**	0.489	**0.702**
Cr	0.375	**0.809**			**Ca**		**0.890**	**SPAH**	0.423	0.477
ionNO3-			**0.975**		**Fe**	0.497	**0.840**	**Mn**	0.451	0.538
ionSO42-	0.381		**0.899**		**EC**	0.365	**0.750**			
ionCl-	0.378		**0.895**		**Cr**	0.363	**0.730**			
EC				**0.945**						
Cu	0.420	0.362		**0.727**						
source:	combustion /traffic I	road dust	secondary particles /sea salt	traffic II	**source:**	combustion/ traffic/ secondary	road dust	**source:**	combustion/ traffic/ secondary	mineral dust
Rotation Sums of Squared Loadings					**Rotation Sums of Squared Loadings**			**Rotation Sums of Squared Loadings**		
% of variance	*34.2%*	*30.1%*	*18.7%*	*11.5%*	**% of variance**	*49.4%*	*30.5%*	**% of variance**	*55.6%*	*18.2%*
Cumulative %	*34.2%*	*64.3%*	*83.0%*	***94.5%***	**Cumulative %**	*49.4%*	***79.9%***	**Cumulative %**	*55.6%*	***73.8%***
Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown					Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown			Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown		

Table 4.3. PCA results for Ljubljana (traffic, urban background, rural site).

Ljubljana_traffic site			
	Components (Factors):		
Compounds:	**1**	**2**	**3**
EC	**0.941**		
OC	**0.935**		
K	**0.880**		
Ca	**0.797**		
SPAHs	**0.773**		
Pb	**0.755**		
Cu	**0.695**	0.447	
Cr		**0.990**	
Fe		**0.941**	
Mn		**0.925**	
Zn	0.406	**0.852**	
ionSO42			**0.832**
ionNO3		0.549	**0.697**
Ni		0.469	-0.630
source:	traffic / combustion	traffic (non-exhaust) /industrial/ fuel oil	secondary particles
Rotation Sums of Squared Loadings			
% of variance	*36.7%*	*31.4%*	*12.7%*
Cumulative %	*36.7%*	*68.1%*	***80.8%***

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown

Ljubljana_urban background site				
	Components (Factors):			
Compounds:	**1**	**2**	**3**	**4**
Pb	**0.91**			
Zn	**0.89**			
Cu	**0.89**			
ionCl	**0.78**			
OC	**0.78**		0.40	
SPAHs	**0.75**	0.37		
EC	**0.72**		0.53	
Mn		**0.94**		
Cr		**0.94**		
Fe	0.34	**0.80**	0.44	
Ti			**0.83**	
Ca	0.48		**0.71**	
K	0.57		**0.67**	
ionNO4				**0.79**
ionSO43			0.32	**0.77**
source:	traffic / combustion	traffic (non-exhaust) /industrial/ fuel oil	mineral dust	secondary particles
Rotation Sums of Squared Loadings				
% of variance	*36.4%*	*18.9%*	*17.1%*	*10.2%*
Cumulative %	*36.4%*	*55.3%*	*72.4%*	***82.6%***

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown

Ljubljana_rural site			
	Components (Factors):		
Compounds:	**1**	**2**	**3**
K	**0.909**		
OC	**0.896**		
EC	**0.854**	0.368	
Ca	**0.813**		
Fe	**0.757**	0.409	0.319
Cu		**0.923**	
Zn	**0.562**	**0.754**	
Pb		**0.692**	0.494
Cr		**0.553**	-0.423
ionNO3			**0.882**
ionSO42			**0.856**
Mn	0.304	0.460	**0.658**
source:	traffic/ road dust	traffic (non-exhaust) /industrial/ fuel oil	secondary particles
Rotation Sums of Squared Loadings			
% of variance	*34.5%*	*23.4%*	*22.0%*
Cumulative %	*34.5%*	*57.9%*	***79.9%***

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown

Table 4.4. PCA results for Thessaloniki (traffic, urban background, rural site).

Thessaloniki_traffic site				Thessaloniki_urban background site				Thessaloniki_rural site			
	Components (Factors):				Components (Factors):				Components (Factors):		
Compounds:	**1**	**2**	**3**	**Compounds:**	**1**	**2**	**3**	**Compounds:**	**1**	**2**	**3**
Fe	**0.947**			**OC**	**0.819**	0.423		**K**	**0.910**		
Ni	**0.929**			**Pb**	**0.807**			**EC**	**0.797**	0.321	
Mn	**0.904**			**K**	**0.794**	0.505		**OC**	**0.787**	0.402	
Cr	**0.892**			**EC**	**0.758**	0.533		**Ca**	**0.766**		
K	**0.875**			**Cu**	**0.751**	0.367		**Pb**		**0.892**	
Zn	**0.851**	0.406		**Zn**	**0.725**	0.557		**Cu**		**0.788**	
Ca	**0.840**			**NO3**	**0.674**		0.374	**Zn**	**0.633**	**0.654**	
EC	**0.839**			**Ca**		**0.963**		**SO4**		0.478	
OC	**0.765**			**Fe**		**0.944**		**Fe**	0.369		**0.837**
NO3	**0.715**			**Cr**		**0.920**		**NO3**			**0.743**
Cl		**0.892**		**Mn**	0.387	**0.774**		**Cr**	0.492		**0.719**
Pb		**0.777**		**Ni**	**0.599**	**0.600**		**Mn**		0.427	0.528
Cu	**0.659**	**0.703**		**SO4**	0.421		**0.773**	**Cl**			**0.686**
SO4			**0.953**	**Cl**	0.349		-0.717				
source:	road dust / traffic / combustion	traffic	secondary particles	**source:**	traffic / combusti on	road dust	secondary/sea salt	**source:**	road dust / traffic / combustion	traffic	secondary nitrates/sea salt
Rotation Sums of Squared Loadings				**Rotation Sums of Squared Loadings**				**Rotation Sums of Squared Loadings**			
% of variance	*56.1%*	*16.4%*	*8.1%*	**% of variance**	*35.6%*	*34.5%*	*9.9%*	**% of variance**	*27.9%*	*20.0%*	*17.4%*
Cumulative %	*56.1%*	*72.5%*	***80.6%***	**Cumulative %**	*35.6%*	*70.1%*	***80.0%***	**Cumulative %**	*27.9%*	*47.9%*	***65.3%***
Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown				Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown				Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in six iterations. Only factor loadings ≥0.6 listed. Only factors with eigenvalue ≥1 are shown			

5 Conclusions

The scope of the present report is to present the results from the source apportionment methods application on PM2.5 data collected in four European cities (Athens, Brno, Ljubljana and Thessaloniki) in the frame of ICARUS project. PM2.5 chemical composition data were inserted in PMF (Positive Matrix Factorization) and PCA (Principal Component Analysis) models with the scope of identifying the main groups of sources and estimating their contribution to PM2.5 concentrations.

Depending on the case, the PMF model resulted in a number of three, four or five identified PM2.5 sources/group of sources for each site/city. In most cases, secondary particles and sea salt sources were not distinguished. In presence of specific tracers (e.g. ammonium, sodium), this mixed source would be split into two sources. Similarly in a number of cases, the combustion-related source seems to include also traffic source, presenting quite high percentages. Traffic is presented in three different source-categories: exhaust-traffic, non-exhaust traffic and road dust. Significant variability is observed on the percentages of each source among the sites of the same city. Although significantly varying among the sites of each city, biomass burning/combustion source is aggravated during wintertime due to the domestic heating. Finally, when identified as a separate source, the natural source of crustal dust presented percentages between 5 and 20%. In the other cases, this source was identified as road dust (combined with non-exhaust traffic emissions).

The PCA model resulted in a lower number of components/factors (2-4). Most of them comprise combination of two or three sources (e.g. combustion-related group of sources). The % percentage cumulative rotation sums of squared loadings in PCA varied between 65.3% and 94.5%, which means that these percentages of PM2.5 variation were explained by the model.

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