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Source apportionment of PM₁₀ in a North-Western Europe regional urban background site (Lens, France) using Positive Matrix Factorization and including primary biogenic emissions

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Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

In this work, the source of ambient particulate matter (PM₁₀) collected over a one year period at an urban background site in Lens (France) were determined and investigated using a Positive Matrix Factorization receptor model (US EPA PMF v3.0). In addition, a Potential Source Contribution Function (PSCF) was performed by means of the Hysplit v4.9 model to assess prevailing geographical origins of the identified sources. A selective iteration process was followed for the qualification of the more robust and meaningful PMF solution. Components measured and used in the PMF include inorganic and organic species: soluble ionic species, trace elements, elemental carbon (EC), sugars alcohols, sugar anhydride, and organic carbon (OC). The mean PM₁₀ concentration measured from March 2011 to March 2012 was about 21 µg m⁻³ with typically OM, nitrate and sulfate contributing to most of the mass and accounting respectively for 5.8, 4.5 and 2.3 µg m⁻³ on a yearly basis. Accordingly, PMF outputs showed that the main emission sources were (in a decreasing order of contribution): secondary inorganic aerosols (28 % of the total PM₁₀ mass), aged marine emissions (19 %), with probably predominant contribution of shipping activities, biomass burning (13 %), mineral dust (13 %), primary biogenic emissions (9 %), fresh sea salts (8 %), primary traffic emissions (6 %) and heavy oil combustion (4 %). Significant temporal variations were observed for most of the identified sources. In particular, biomass burning emissions were negligible in summer but responsible for about 25 % of total PM₁₀ and 50 % of total OC at wintertime. Conversely, primary biogenic emissions were found to be negligible in winter but to represent about 20 % of total PM₁₀ and 40 % of total OC in summer. The latter result calls for more investigations of primary biogenic aerosols using source apportionment studies, which quite usually disregards this type of sources. This study furthermore underlines the major influence of secondary processes during daily threshold exceedances. Finally, apparent discrepancies that could be generally observed between filter-based studies (such as the present one) and Aerosol Mass Spectrometer-based PMF analyses (organic fractions) are also discussed here.

25327

ACPD

13, 25325–25385, 2013

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

1 Introduction

During the last decades, the air pollution has become of an increasing concern, especially in urban areas, due to its adverse effect on human health (Pope and Dockery, 2006). Respiratory and cardiovascular illnesses related to PM exposure have been well established in the literature (e.g. Ramgolam et al., 2009; Pope et al., 2009). Therefore, identification of PM sources is necessary to develop air quality improvement strategies in order to be able to control and reduce ambient PM concentrations through targeted actions. To address this issue, many tools were used for the identification and quantification of PM sources (Paatero and Tapper, 1994; Schauer et al., 1996; Ulbrich et al., 2009). Chemical speciation of ambient PM coupled to receptor modeling is currently considered one of the most powerful tool with this respect (Srimuruganandam and Shiva Nagendra, 2012).

Receptor models have been applied in many studies and in many regions and have shown the ability to identify accurately the potential emissions sources at a receptor site (Watson et al., 2001; Chow et al., 1992). They are based on the hypothesis that mass conservation as well as mass analysis can be used to apportion sources of ambient PM in the atmosphere. However, the choice of a specific receptor model depends largely on the knowledge of sources (Viana et al., 2008). If the chemical sources profiles are available, chemical mass balance (CMB) model can be used (Schauer et al., 1996). Positive Matrix Factorization (PMF) model (Paatero and Tapper, 1994) is highly recommended when sources are not formally known, but it requires post treatment source identification. PMF was used in many studies dealing with air pollution in urban areas in the USA, Europe, and Asia (Shrivastava et al., 2007; Viana et al., 2008; Pandolfi et al., 2011). Many sources such as road transport, industrial emissions, sea-salt, and crustal dust were identified using this method (Viana et al., 2008). However, there have been few PMF studies where metals and inorganic aerosols are combined with organic tracers (Shrivastava et al., 2007; Wang et al., 2012). In addition, the contribution of biogenic sources (primary and secondary fractions) has not been clearly

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



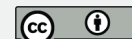
Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of PM₁₀ in Lens, France by means of the PMF modelA. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

investigated up to now (Jimenez et al., 2009). These sources can represent a very important fraction (> 40 %) of PM mass (Hallquist et al., 2009; El Haddad et al., 2011) and should not be neglected. Recently, some sugar alcohols (polyols) tracers such as arabitol, sorbitol and mannitol were used to apportion the primary contribution of this fraction (Graham et al., 2003; Yttri et al., 2007; Bauer et al., 2008). However, and to the best of our knowledge, the contribution of primary biogenic sources has not yet been investigated using PMF.

In this work, we report a source apportionment of ambient PM₁₀ concentrations in an urban background site located in the northern part of France, a region which is frequently submitted to PM₁₀ limit value exceedances (MEDDE, 2011). As explain below (Sect. 3), this site can also be considered as representative of baseline urban background conditions in North-Western Europe. The data collected during a one year period (2011–2012) included the characterization and the quantification of more than 36 species including metals and trace elements, major ions, sugars anhydride, and some sugar alcohols tracers. PMF receptor model, with a stringent quality control, was applied for the estimation of the emission sources, including primary biogenic aerosols.

2 Methodology

2.1 Sampling site

Daily PM₁₀ filter samples were collected by the local air quality monitoring network (ATMO Nord Pas de Calais) at an urban background site in Lens, north of France (Fig. 1). The city of Lens (35 000 inhabitants, 50°25' N latitude and 2°21' E longitude) is actually part of a larger conurbation of more than 500 000 inhabitants and is surrounded by other large populated areas in the north of France (conurbation of Lille, Bethune and Valenciennes). Whereas, the sampling site is located relatively far away from major roads (approximately 1 km) and stationary emission sources, it is expected to be influenced by many anthropogenic activities. Road transport emissions should

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

be considered, with several highly trafficated highways (A1, A21, A26) passing through this north to south transit region. In addition, many petrochemical, metallurgic, and non-metallurgic industrial companies are located in the coastal zone or dispersed in the region. Being a populated area (325 inh. km^{-2}), it is also influenced by domestic emissions, including residential wood burning (ATMO Nord Pas de Calais, 2009, www.atmo-npdc.fr). Finally, it is located 80 km from the Strait of Dover, and can therefore be influenced by oceanic sea spray episodes as well as maritime transport. The climate in Lens is oceanic with an average temperature of 4°C in winter and 18°C in summer during the period of the study.

2.2 Measurements

Chemical analyses were performed on PM₁₀ daily samples collected every third day from the 9 March 2011 to the 6 March 2012. These samples consist of 150 mm diameter quartz fiber filters (Pall-Gelman 2500 QAT-UP) obtained using a high volume sampler (DA80, Digital) equipped with a PM₁₀ cut-off inlet and operating at a flow rate of $30 \text{ m}^3 \text{ h}^{-1}$. The filters were preheated at 500°C for 12 h before exposure in order to remove any trace of organic contaminants. Results presented here have been obtained on a total number of 117 samples (and 8 blank filters). PM₁₀ mass concentrations were measured at the same site using a beta gauge (MP101M, Environnement S.A.) equipped with a PM₁₀ inlet and smart heater system (so-called RST) allowing for removing water at high ambient relative humidity.

Chemical analyses were performed for various elements and components using a range of instrumental techniques on sub-sampled fractions of the filters. The elemental carbon (EC) and organic carbon (OC) were analyzed using the thermo optical transmission method on a Sunset Lab analyzer (Birch and Cary, 1996). A punch of 1.5 cm^2 was directly analyzed following the EUSAAR-2 protocol (Cavalli et al., 2010) and automatic split time was used to differentiate EC and OC. Sugar anhydrides (levoglucosan, mannosan, and galactosan) and sugar alcohols (arabitol, sorbitol, mannitol) were analyzed by HPLC-PAD using a set of Methrom columns (MetroSep A Supp 15

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

and Metrosep Carb1). For this analysis, extraction of filter samples was performed using ultrapure water under mechanical agitation for a period of 30 min, the extract being filtrated with 0.22 µm Nuclepore filters before injection (Piot et al., 2012).

Soluble anions and cations were analyzed by ionic chromatography (IC, Dionex DX-600). Briefly, samples were soaked for one hour in 10 mL of Milli-Q water, and then filtered using 2 µm-porosity Acrodisc filters before analysis. AS/AG 17 and CS/CG 12A columns were used respectively for anions and cations analyses. Only NO₃⁻, SO₄²⁻, Cl⁻ and NH₄⁺ concentrations subsequently obtained were used for PMF analysis. As for other species, this methodology is similar to the ones commonly used for ambient air filter samples (e.g. Jaffrezo et al., 2005).

Concentrations of seven major (Al, Na, Mg, K, Ca, Fe and Ti) and seventeen trace elements (As, Ba, Cd, Ce, Co, Cs, Cu, La, Mn, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn) were analyzed by ICP-AES (IRIS Intrepid, Thermo-Scientific) and ICP-MS (ELAN 6100 DRC, Perkin Elmer), respectively (Alleman et al., 2010). Prior to analyses, each sub-samples were acid digested (HNO₃; HF; H₂O₂) with a microwave oven (Milestone ETHOS). Repeated measurements were performed on acid blanks, quality control standard solutions and standard reference material (SRM 1648a, urban particles). Finally, As, Cd, Ce, Co, Cs, La, and Mn were not included in the PMF model due to relatively high uncertainties and detection limits on quartz filters for these trace elements.

A total number of 8 field blank samples were also analyzed with the same techniques in order to determine the detection limits (DL) of the methods. The average blank values were subtracted from the samples before calculation of atmospheric concentrations.

2.3 Source apportionment

Source contributions of ambient PM₁₀ samples were estimated using PMF (Paatero and Tapper, 1994) by means of the US EPA PMF v3.0 software. This multivariate tool uses measured concentrations and their uncertainties to solve the mass balance equation $\mathbf{X} = \mathbf{G} \times \mathbf{F} + \mathbf{E}$. \mathbf{X} is the data matrix, \mathbf{F} is a matrix which vectors represent the profiles of p sources, \mathbf{G} is a matrix which columns represent the contributions of the p sources

and the matrix **E** is the residual matrix. Further description of the model can be found in Paatero and Tapper (1994). During this study, a total of 117 samples were used for the PMF source apportionment. The variety of number of factors, species and parameters tested which led to the best PMF solutions are detailed in Sect. 3.2.

5 2.4 Potential contribution source function

In order to be able to evidence prevailing geographical origins of major PM₁₀ sources identified through PMF analysis, air masses origins were computed using potential contribution source function. This data treatment procedure, so-called PSCF, represents the probability that an air parcel may be responsible for high concentrations measured at the receptor site. It allows geographical identification of potential emission areas, by associating any concentration time series (i.e. chemical species or temporal contributions of PMF factors) with back-trajectories (Ashbaugh et al., 1985). The methodology is extensively described elsewhere (Polissar et al., 1999). Briefly, at each *ij*th grid cells, probabilities are calculated as:

$$15 \text{ PSCF}_{ij} = \frac{m_{ij}}{n_{ij}}$$

where n_{ij} represents the total number of back-trajectories passing through each *ij*th cell, and m_{ij} the number of back-trajectories passing through the same cell that are associated with measured concentrations over an arbitrary threshold. This threshold is usually empirically set. Whereas low criterion values cause noisy and hazy analysis, a threshold too high would not be statistically representative. In this study, the 75th centile of each PMF factors' temporal contribution was chosen, as it gave the best geographical representativeness. It means that only the higher 25 % of the samples are used for defining the main source regions for a given source factor. A weighing function can then be implemented in order to avoid artifacts usually linked to high concentrations with low n_{ij} values. Weighing coefficients are commonly empirically determined

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

(Hwang and Hopke, 2007; Jeong et al., 2011), but another method, based on a “back-trajectory density” ($\log(n+1)$), allows less manual work (Bressi, 2012). Following Bressi (2012) and Fig. 2, the weighing function is determined as:

$$W = \begin{cases} 1.00 & \text{for } n_{ij} \geq 0.85 \cdot \max(\log(n+1)) \\ 0.725 & \text{for } 0.6 \cdot \max(\log(n+1)) > n_{ij} \geq 0.85 \cdot \max(\log(n+1)) \\ 0.35 & \text{for } 0.35 \cdot \max(\log(n+1)) > n_{ij} \geq 0.6 \cdot \max(\log(n+1)) \\ 0.1 & \text{for } 0.35 \cdot \max(\log(n+1)) > n_{ij} \end{cases}$$

where $\max(\log(n+1)) = 2.69$, or $\max(n_{ij}) = 497$ in this study. For graphical purpose, resulting PSCF values were filtered by a Gaussian smoothing algorithm and eventually normalized. All calculations were performed under Python.

In addition, one hypothesis in the PSCF is that particles are transported without dispersion and deposition; however, wet deposition is roughly estimated, assuming that even low precipitation (≥ 0.1 mm) would clean up the air parcel, by setting to 0 all grid cells in between the rainy one and the end point (Bressi, 2012). Finally, PSCF was applied here to temporal variations of the PMF factors. 72 h back-trajectories, along with meteorological parameters (i.e. precipitation) were calculated using the PC-based version of Hysplit (v.4.9 ; Draxler and Hess, 1997), every 12 h from 9 March 2011 LT to 6 March 2012 LT. The longitude, latitude and altitude of the end point were set at 2.81° , 50.43° , and 500 m, respectively. As concentration data are available only every 3rd day, 3 back-trajectories are associated with each data point, centered at the mid sampling time (i.e. $t_{\text{average}} \pm 12$ h). The spatial frame of the grid cells is set from (30° ; 66°) to (25° ; 45°), with a grid resolution of $0.5^\circ \times 0.5^\circ$.

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

3 Results and discussions

3.1 PM mass concentration and chemical composition

An average PM₁₀ concentration of $21 \pm 13 \mu\text{g m}^{-3}$ is obtained from collocated automatic measurements with dates corresponding to filters sampled for the study. This is quite similar to the overall mean PM₁₀ concentration from 9 March 2011 to 6 March 2012 ($22 \pm 15 \mu\text{g m}^{-3}$), and in the range of annual mean concentrations observed at Lens urban background sites from 2008 to 2012 ($19\text{--}24 \mu\text{g m}^{-3}$). The percentage of the highest daily concentrations ($\geq 40 \mu\text{g m}^{-3}$) is also equivalent for daily samples investigated here and for overall daily concentrations during the period of study (13% in both cases). It should however be noted that the percentage of exceedances of the $50 \mu\text{g m}^{-3}$ daily threshold is significantly lower for the subset investigated here (4%) than that corresponding to the whole period (8%), which can be explained by the random distribution of these daily threshold exceedances and the relative shortness of the dataset. The city of Lens is located relatively far away from major stationary sources which could significantly influence other urban sites of northern France, so that concentrations recorded in other cities of the region are generally higher (up to 50%) than that of Lens (ATMO Nord Pas de Calais Report, 2009). Similarly, annual mean concentrations recorded in Lens are in the lower range of urban background concentrations observed in neighboring countries (see e.g. Maenhaut et al., 1996; Harrison et al., 2003; Yin et al., 2005 and Beuck et al., 2011, respectively for Belgium, UK, Ireland and Germany). Therefore, results presented below can certainly be considered as representative of regional urban background conditions in this part of Europe.

A clear seasonal pattern of PM₁₀ concentrations can be observed (Table 1), with maximum concentrations in spring ($29 \pm 13 \mu\text{g m}^{-3}$) and minimum concentrations in summer ($14 \pm 13 \mu\text{g m}^{-3}$).

OM is calculated with an OM/OC ratio of 1.75 for all the seasons, as reported by Turpin and Lim (2001). As presented in Table 1 and Fig. 3, it is the major contributor to

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

the PM₁₀ mass except in spring and accounts for 34 % (6.7 μg m⁻³), 21 % (6.1 μg m⁻³), 23 % (3.2 μg m⁻³), and 37 % (7.1 μg m⁻³) of the PM₁₀ in winter, spring, summer, and fall, respectively. During the measurement period, OM concentrations and contributions are significantly larger in winter (Fig. SI1, Supplement) as usually observed in Europe (Bergström et al., 2012; Gianini et al., 2012; Legrand and Puxbaum, 2007). This is generally partly explained by the significant influence of residential wood burning during this period of the year (Favez et al., 2009). In the present study, seasonal variations of potassium (K) and Levoglucosan which are commonly used as tracers of biomass combustion (Puxbaum et al., 2007; Dall'Osto et al., 2009; Zhang et al., 2012) showed a similar variability as OM, with increased concentrations during the winter and to a lesser extent in the fall season (Table 1 and Fig. 3).

The second major contributor to PM₁₀ total mass concentrations is nitrate and accounts for 22 % of the total PM₁₀ mass on average (yearly mean concentration of 4.5 μg m⁻³). A clear seasonal variation is also observed for this species, the highest and lowest contributions being observed during spring (8.5 μg m⁻³) and summer (2.0 μg m⁻³), respectively. More precisely, the highest concentrations are observed at the end of winter–beginning of spring. As already vastly reported, this can be related to the semi-volatile character of ammonium nitrate and frequent occurrences of photochemical episodes during a period of the year which also corresponds to rather humid conditions, low temperatures and increased agricultural activities in this part of Europe (Dall'Osto et al., 2009). Accordingly, ammonium concentrations are also higher in spring (Table 1 and Fig. 3). It is to note that the sharp decrease of nitrate and ammonium concentrations in summer could be partly linked to sampling artifacts because of the thermal instability of ammonium nitrate (Querol et al., 2001). The third major contributor is sulfate accounting for 11 % of the total PM₁₀ mass on average (yearly mean concentration of 2.3 μg m⁻³). It shows a relatively flat seasonal pattern (Table 1 and Fig. 3), due to increased SO₂ emissions and lower boundary layer height in winter, but more efficient photochemical processes in summer.

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Among the other measured components at Lens, EC, Cl⁻, and Na are also important contributors to total PM₁₀ mass concentration accounting for 4.6 % (1.0 μg m⁻³), 3.4 % (0.7 μg m⁻³) and 2.5 % (0.5 μg m⁻³) of the total mass on an annual basis, respectively. EC, a good tracer for combustion (Bond et al., 2004), follows a temporal variation with maximum concentrations in fall (1.3 μg m⁻³) and minimum concentrations in summer (0.5 μg m⁻³). Sea spray tracers (Cl⁻ and Na) both have increased concentrations in the winter season (Fig. S12, Supplement) whereas during the other seasons, no significant seasonal variation is observed. Crustal material such as Ca and the ∑Metals and trace elements which accounts for 3 % (0.6 μg m⁻³) of PM₁₀ mass, were slowly increased during the spring season (March through May 2011) accounting for 4 % (1.1 μg m⁻³) of the total PM₁₀ mass concentration.

The concentration of ∑Sugar alcohols, considered as tracers of primary biogenic emissions (Yttri et al., 2007) increased during the summer season (Table 1). Indeed, the fraction compared to the OM deduced from the OC measurement in (mg/gOM) showed that arabitol reached a value of 7.5 mg g⁻¹ of OM during summer, much higher than during the winter season (1.9 mg g⁻¹ of OM). An identical seasonal variation was observed for mannitol, with values of 1.1 mg g⁻¹ and 5.5 mg g⁻¹ of OM calculated for winter and summer, respectively (Fig. 3). The increase of the ∑Sugar alcohols concentrations during summer is generally related to the increase of biological activity due to higher thermal convective activity and photophoresis (Graham et al., 2003). For levoglucosan, a marker of biomass combustion, the calculated fraction increase in the winter season with a value of about 47 mg g⁻¹ of OM compared to a value of 3.6 mg g⁻¹ of OM reported in summer (Fig. 3). The winter maximum is obviously related to the use of wood combustion for household heating.

3.2 PMF model optimization

In order to provide the best solution in term of stability, performance, accuracy, and geochemical likeliness, several numbers of factors (from 6 to 14) and combinations of

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

included species (from 17 to 28) following an objective step by step methodology were tested. It happens that a solution with 9 factors provided the most adjusted and robust results. This selection is a multi-steps process where, at first, all the available chemical species are included. Further adjustments of the list of included species and number of factors are based on an iterative process. One of the criteria used to evaluate the stability of the results at each step is the capability of the model to reproduce experimental concentrations, particularly for species considered as tracers, while the likelihood of the solution factors are evaluated against geochemical and seasonal considerations about the potential sources observed at the site. The quality of the bootstraps and examinations of the residuals are also considered.

In the final PMF solution (hereafter called “the base run”), the chemical species kept are OC*, EC, SO₄²⁻, NO₃⁻, Cl⁻, NH₄⁺, Ca, Mg, Na, K, Polyols*, levoglucosan, Ba, Cu, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn, Al, Fe, Ti, and PM₁₀. For Na, Ca, K and Mg, we used the total concentrations obtained by ICP-AES instead of the ionic soluble concentrations. Due to the fact that some organic tracers such as levoglucosan, sorbitol, mannitol, and arabitol are injected in the PMF, the OC is replaced by OC* (OC* = OC – [levoglucosan/1.25 + sorbitol/2.5 + mannitol/2.5 + arabitol/2.5]). These ratios were determined based on the carbon molar mass of each compound. The “Polyols*” represents the sum of arabitol, sorbitol and mannitol concentrations. Missing values ($n = 13$) for PM₁₀ were replaced by a reconstructed mass concentration for PM₁₀ (PM₁₀ = EC + 1.75*OC + 3*Na + 10*Ca + NO₃ + nssSO₄²⁻ + NH₄⁺ based on such chemical mass closure calculated for other filters ($R^2 = 0.97$; equation, $y = x - 0.56$). Sea-salt sulfate (ssSO₄²⁻) was calculated by multiplying the mass concentration of Na by a factor of 0.252 following the methodology described by Seinfeld and Pandis (2006). The non-sea salt sulfate (nssSO₄²⁻) could then be calculated by subtracting the ssSO₄²⁻ mass concentration from the total mass concentration of sulfate. All species measured below detection limit (DL) were replaced by 1/2 DL (Norris et al., 2008). All these species were defined from weak to strong in the PMF based on their signal-to-noise ratio. Mo, Fe, and Ti having a signal-to-noise ratio less than 2 were defined as “weak”. Therefore,

the lowest signal-to-noise ratio of 1.27 used in this study was for Mo. PM₁₀ is defined as “total variable” and is automatically categorized as “weak”.

Because the treatment of uncertainties has a significant effect on the outputs of PMF results and because no standardized methodology is provided for PMF resolution (Hopke et al., 2013), several tests for the uncertainties assessments were performed. At a first step, the individual estimation of uncertainties of the data set is performed following the methodology described by Gianini et al. (2012) adapted from Anttila et al. (1995). It uses the Detection Limit (DL, twice of the standard deviation of the field blanks) and the coefficient of variation (CV, Standard deviation of repeated analysis divided by the mean value of the repeated analysis). Following this procedure, trace metal elements characterized by very low concentrations would lead to unrealistic low uncertainties, a situation that should be avoided in the PMF input data sheets as recommended by Hopke et al. (2013). For this purpose, the uncertainty calculation of these trace elements was performed using the expanded relative uncertainties for each species instead of CV and the total uncertainties were calculated by multiplying these relative uncertainties by the concentration of each species. These relative uncertainties included variability from contamination, sampling volume, repeatability and accuracy (through the digestion recovery rate). Finally, we used an expanded relative uncertainty of 10 % (Lim et al., 2003) for OC, 15 % (Schmid et al., 2001; Cavalli et al., 2010) for EC and 15 % for monosaccharide sugars such as levoglucosan, arabitol, sorbitol and mannitol (Piot et al., 2012; Iinuma et al., 2009).

Besides the use of the methodology described above, two additional tests concerning uncertainties were performed using the methodology described in the PMF guide book 3.0 (Norris et al., 2008). The results (Table 2) showed that when trace metals having low concentrations and low uncertainties were calculated with both the Gianini (Gianini et al., 2012) and the Polissar (Polissar et al., 1998) methodologies the outputs are not stable (scaled residuals are not within the range of -3 and +3 of the standard deviation and bootstrap results cannot be mapped for all the factors). However, when using the relative uncertainty fraction instead of the CV for these trace elements

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

and in both methodologies, the results appear to be more consistent and stable. The simulation used with the Gianini methodology and the relative uncertainty fraction for trace elements was selected since using the Polissar methodologies would require the Method Detection Limit (MDL) obtained from blank laboratory filters. During this study, only field blank filters were available.

In order to test the stability of the base run, the same parameters used for this 9-factors solution were also employed by varying the number of factors from 8 to 14. Briefly, the results showed that with an 8-factors solution, the “oil combustion” and the “aged marine aerosols” sources were combined in the same factor (Fig. SI4, Supplement) whereas a 10-factors solution showed some species dispatched between two factors with one of the factor profile which cannot be clearly ascribed to a specific source. In addition, the bootstrap results for the 10-factors solution were not satisfactory, with a factor ascribed in only 33% of the runs (Table SI1, Supplement). Furthermore, above 11 factors, a growing number of runs are not converging. Finally, introducing additional factor is lowering loosely the values of Q robust and Q true.

Additional runs were performed in the trial and error selection process, including a simulation using the sugars alcohols separated and not combined as a unique specie, a simulation using total OC instead of OC*, and a simulation excluding the sugar alcohols. The results obtained showed that an 8-factors solution with the exclusion of the sugars alcohols tracers (polyols*) allows the identification of the same sources (Fig. SI6, Supplement) as in the base run except for the biogenic emissions source, suggesting that these markers have no discriminant effect on the sources other than the primary biogenic emissions. The final choices of gathering polyols in a single component (polyols*) and using OC* instead of OC are discussed below.

Model performance for the base run showed determination coefficients (R^2) between the modeled and experimental concentrations for PM₁₀, OC, and EC of 0.97, 0.98, and 0.96 (Table 3), respectively. Most of the other chemical species are also well reconstructed, except for some trace elements like Zn, V, Sb, Ni, Mo, and Ba. It is most probably due to the fact that PMF is not always able to model accurately species having

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

low concentrations and high uncertainties. These results are within the range of those presented in many PMF studies, with for example, values of 0.71 reported for a study in Spain (Cusack et al., 2013) and of 0.96 for a study in Germany (Beuck et al., 2011) for PM mass reconstruction. Scaled residuals between -3 and $+3$ are obtained for all of the major components, and the value of Q robust is strictly identical to the value of Q true, all of these showing that no specific event is affecting the results and that the base run can be regarded as stable. In addition, 100 bootstrap runs were systematically performed for every tested configuration (Table S11, Supplement); the results, with the lowest correlation between bootstrap solutions and “true solution” being 0.6, showed that all 9 factors are well mapped in the base run.

Comparison of the results stability for the other simulations demonstrated that when total OC is used instead of OC*, the stability of the solution is very comparable in terms of bootstrap results to that of OC*. For this purpose, OC* will be used instead of total OC in order to avoid double counting of some of the carbon mass. For the simulation with separated sugars alcohols markers, the results of the stability in term of bootstraps were slightly less satisfactory with Q robust and Q true of 2155.6 and 2156.3, respectively, while the amount of residuals samples and species exceeding the range of -3 to $+3$ were higher compared to the base run. In addition, bootstrap results were also worse, with values as low as 63 % of the runs for some factors. Therefore, the 9-factors solution with sugars alcohols combined was chosen and these results are presented and discussed in the following sections.

3.3 Contributions of the factors to total PM₁₀ levels

The identified sources are fresh sea salt, primary biogenic emissions, mineral dust, biomass burning, oil combustion and traffic emissions, complemented by two secondary aerosols fractions characterized respectively by high sulfate and nitrate contents as well as a factor that could mixed aged sea salt and maritime transport emissions (aged marine). These sources were identified based on their overall chemical profiles and loading with specific species (highlighted in Table 4) defined as tracers.

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Most of these source profiles and tracer species are well documented in the literature and will be discussed in details in the next sections. However, it should be noted that since the PMF model results are based on internal correlations among species or markers which have similar time series, it is somewhat difficult to distinguish between two sources that do not vary independently (Paatero et al., 2002). Moreover, and since measurement are on 24 h basis every third day, some variability (like the diurnal variation of some sources, or the weekends, when measurement occurred for Saturday or Sunday only) are not captured and processes could not be identified. Thus, this sampling methodology has its limitations in terms of sources identification. It is therefore likely that some primary and secondary sources are not separated.

The contribution of these sources to total PM₁₀ mass concentration is presented in Fig. 4 on a yearly basis and averaged for each season, with winter (December–February), summer (June–August), spring (March–May) and fall (September–November) including 30, 28, 30, and 28 samples, respectively. On a yearly basis, the major PM₁₀ contributors at Lens appear to be secondary aerosols and the aged marine fraction. They account, respectively for 28 % (5.6 μg m⁻³) and 19 % (3.8 μg m⁻³) of the total PM₁₀ mass. The second series of sources are the mineral dusts (13 %, 2.6 μg m⁻³), the biomass burning (13 %, 2.6 μg m⁻³), the primary biogenic emissions (9 %; 1.8 μg m⁻³) and sea salts (8 %; 1.6 μg m⁻³). Primary traffic and heavy oil combustion emissions only account for 6 % (1.2 μg m⁻³) and 4 % (0.8 μg m⁻³) of the total PM₁₀ mass on a yearly average, respectively.

However, there are large changes in the importance of these sources according to the seasons. The contribution of primary biogenic emissions increases and accounts for 20 % of the total PM₁₀ mass concentration, becoming an important source contributor during the summer season (Fig. 4). Among other sources, the contribution of the secondary nitrate factor decreases during summertime due to the thermal instability of ammonium nitrate whereas the contribution of the sulfate secondary factor increases. In addition, the contribution of the “aged marine” factor also increases in summer with a comparable pattern to that of sulfate (cf Sect. 3.1).

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

In winter, the biomass burning source is the major contributor accounting for 25 % of the total PM₁₀ mass concentration, related to the use of wood combustion for domestic heating (BASIC, 2012). The contribution of the fresh sea salt factor also increases in winter (15 %) whereas the contribution of mineral dust and primary biogenic emissions are largely decreasing (Fig. 4) due the limited biological activities and to the larger occurrence of rainy days responsible for the efficient scavenging and trapping on the ground of the mineral dust. Indeed, during the winter season of 2011/2012, the cumulated precipitation in the region was above the normal levels (Meteo France, 2012).

In fall, biomass burning is still an important contributor whereas the contribution of the nitrate-rich factor decreases significantly with a comparable pattern to that of summer. During the spring season, major contributors are the nitrate-rich and the mineral dust factors accounting together for more than 48 % of the total PM₁₀ mass concentration (Fig. 4a). Additional information on the monthly variability of the factors as well as the weekdays to weekend's temporal variations is provided in Fig. 4b. Moreover, chemical profiles, temporal trends and PSCF outputs obtained for each individual factor are presented in Figs. 5–7.

3.3.1 Secondary aerosol factors

The two secondary aerosol factors which are the major contributors to total PM₁₀ mass concentration are characterized mostly by the presence of ammonium, sulfate, and nitrate (Table 4). They are split into a nitrate-rich and a sulfate-rich factors (Fig. 6). In addition, many other components associated with anthropogenic emissions (like Mo, Ni, Pb, V, and Zn) were observed in the source profiles of these factors, potentially highlighting links with industrial sources on a regional scale. These markers were identified as markers from industrial smelters in the literature (Amato et al., 2009; Moreno et al., 2011; Richard et al., 2011). However, their contributions in the source profiles in Lens do not exceed 20 % of the respective mass of each element.

Sulfate, nitrate, and ammonium are commonly identified as markers of the secondary aerosols in many source apportionments studies performed over Europe. According to

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

the review conducted by Viana et al. (2008) for 28 source apportionment studies in Europe, 32 % of the results identified these markers (sulfate, nitrate, and ammonium) as source signature of a secondary aerosol component, related to regional background and long-range transport. Their contribution vary from a low 12 % of the total PM₁₀ mass in Cork (Ireland) to 46 % of the total PM₁₀ in Milan (Italy) (Viana et al., 2008 and references therein). The sum of the contribution of these two secondary aerosol factors (28 % on a yearly average in Lens) is similar to the ones previously reported for various urban sites in Europe (Beuck et al., 2011; Belis et al., 2013).

Temporal variations of these factors (Fig. 6) indicate common evolutions during the fall and winter seasons, arising from the large scale distribution of the sources and processes leading to these factors, while higher spring contributions and lower summer contributions of ammonium nitrate is notably related to meteorological conditions and the semi-volatile character of this compound as discussed above. Mid-distance transport from the N–NE sectors is well highlighted by PSCF analysis (Fig. 7) for these two secondary factors, notably because NH₃, NO_x and SO₂ emissions are intense in the Benelux area (Pay et al., 2012).

3.3.2 Mineral dust factor

The mineral dust factor (Table 4 and Fig. 5) is identified by the presence of Ca (50 % of the Ca mass), Al (58 % of the Al mass), Fe (31 % of the Fe mass), and Ti (72 % of the Ti mass). The high shares of Ca, Al, Fe, Ti and also of Sr are clear markers of such a source (Querol et al., 2002; Lucarelli et al., 2004; Almeida et al., 2005; Dall'Osto et al., 2013). The important share of Rb (35 % of the total Rb mass) is also an indication of mineral particles injected into the atmosphere from eroded soils and rocks (Moreno et al., 2010). It is noteworthy that this factor also includes a significant share of OC (15 % on a yearly average of OC by mass), an indication of mixing of dust and organic matter during aging or by entrainment of organic materials from the soils (Kuhn, 2007).

The reported yearly average concentration of the mineral dust factor for Lens (2.6 μg m⁻³) is in excellent agreement with the reported value of 2 μg m⁻³ for the re-

gional mineral contribution to total PM₁₀ mass concentrations in most Northern and Central Europe (Querol et al., 2004). In terms of relative contribution, the share of 13 % of PM₁₀ reported at Lens is in agreement with the reported values in Italy (16 %, Mossetti et al., 2005), Ireland (7–16 %, Yin et al., 2005) and Denmark (10 %, Andersen et al., 2007).

Time series of this source (Fig. 5) showed important contributions during the spring season in agreement with the seasonal evolution of the measured concentrations for Ca and the Σ metals. However, PSCF analysis shows a very similar trend for this dust factor and for primary traffic emissions, suggesting a major influence of road transport for particles re-suspension. This latter hypothesis seems to be further confirmed by the significant correlation coefficient ($R = 0.76$) obtained for the daily concentrations of the mineral dust and traffic factors.

3.3.3 Primary biogenic emission factor

Primary biogenic emissions at Lens account for 9 % of the total PM₁₀ mass concentration on a yearly average (Fig. 4). During the last decades, the awareness of the important contribution of this source increased (Bauer et al., 2008), with studies proposing values for its contribution between 5 and 50 % of total PM (Jaenicke, 2005). To the best of our knowledge, no PMF study conducted in Europe has previously allowed to apportion this source.

According to Table 4, this factor is characterized by the large amount of polyols and accounts for 66 % of their mass on average. These markers were identified in the literature as tracers of primary biogenic emissions originating from primary biological aerosol particles (Caseiro et al., 2007; Elbert et al., 2007; Jia et al., 2010; Yttri et al., 2011). Specifically, sorbitol was associated with plants debris (Yttri et al., 2007) whereas arabitol and mannitol were identified as markers for fungal spores (Bauer et al., 2008).

Temporal variation of this factor as presented in Fig. 5 shows a clear seasonal variation with maximum concentrations observed during the summer season from June

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

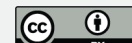
Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

through September. The concentrations of these sugars tracers increase by a factor of 3 between winter and summer. Maximum concentrations of arabitol and mannitol were also previously observed during the summer season (Pashynska et al., 2002; Yttri et al., 2007; Bauer et al., 2008), in relation to higher biological activity because of intense solar radiation and higher surface temperatures which result in an increase of fungal spores, fern spores, as well as pollen grains and plant fragments (Graham et al., 2003). The identified seasonal variation for this source is also typical for Secondary Organic Aerosols (SOA) which increases in the summer season. Therefore, some fraction of (SOA) could be included in the primary biogenic emissions source since separation between daytime and nighttime among the sources is not possible. However, it should be noted that photo-oxidation processes are not too important in the region in the north of France such as Lens. Since SOA markers were not quantified and included in this PMF study, the separation between primary organic aerosols (POA) and SOA is also not possible. Weekday to weekend variation showed higher contribution of the primary biogenic source during the weekend (12% of total PM₁₀ mass; 1.40 μg m⁻³) in comparison to a contribution of 8% (1.18 μg m⁻³) during the week. The reason for these difference are currently unknown. In the PSCF analysis, hotspot pointing to the south (S–SO) and observed for the primary biogenic emission factor could be a calculation artifact, suggesting local emissions. Indeed, the weighing function cannot be fully optimized for each factor since long-distance hotspots associated with low n_{ij} values are easily down weighted by the weighing function, whereas cells relatively closed to the sampling site are still considered as statistically representative. However, such a result could also be consistent with the presence of large agricultural activities in the French Picardie region, south to the sampling site as well as in the southern part of the Nord Pas de Calais region.

3.3.4 Biomass burning factor

The biomass burning factor is non-ambiguously characterized by the presence of high shares of levoglucosan (84% of the total levoglucosan mass on average), potassium

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

(20 % of K mass), Rb (22 % of Rb mass), along with an OC/EC ratio of 4.4, typically observed in wood burning emissions (Fine et al., 2001). Levoglucosan is a degradation product of cellulose burning and is used as a tracer of biomass burning emissions (Simoneit et al., 1999; Puxbaum et al., 2007). K is used in many source apportionment studies conducted in Europe as an indicator of biomass burning (Andersen et al., 2007; Rodríguez et al., 2004). Rb is a good indicator as well of biomass burning emissions (Godoy et al., 2005). It should be noted that the contribution of Biomass burning might be under-estimated here for the summer season (where open burning could take place), notably due to a possible reactivity of levoglucosan during this period of year (Hennigan et al., 2010).

The contribution of 13 % of the PM₁₀ for this factor observed in Lens is consistent with the average reported contribution of 12 % to total PM₁₀ mass concentration for Europe (Belis et al., 2013). Biomass burning impact at Lens classically follows a well-marked seasonal variation with higher concentrations observed in the winter season when this share reaches 25 % of the PM₁₀ mass. Such high contributions were already observed in Europe: for example, biomass burning contributes in winter to 40 % of the OM mass (Favez et al., 2010), in Grenoble (France), 20 % of the total PM_{2.5}, in Paris, France (Favez et al., 2009) and 20 % of the total PM₁₀ mass in Salzburg, Austria (Caseiro et al., 2009). Indeed, large contributions (20–67 %) of this source during the cold season were reported in many source apportionment studies in Europe (Puxbaum et al., 2007; Herich et al., 2013). As mentioned above, the observed PSCF analysis hotspot pointing to the South (Fig. 7) could be a calculation artifact (Sect. 2.3.3). Nevertheless, biomass burning air masses originated from distant local emissions such as the south and transported to the measurement area in Lens would not be surprising, especially from the densely populated Ile-de-France region, where wood burning is found to be a major source during winter time (Favez et al., 2009). Large emissions from the Nord Pas de Calais region, and especially its southern rural part, are also expected.

3.3.5 Traffic emissions

The primary traffic emissions factor identified in Lens is a minor source, accounting on an annual average for only 6 % of the total PM₁₀ mass concentration. It is characterized by high fractions of EC (45 % of the EC mass), Cu (52 % of the Cu mass), Sb (44 % of the Sb mass) and an OC/EC ratio of 0.25. The high fraction of EC together with an OC/EC ratio lower than 0.7 are good indicators from exhaust traffic emissions (Amato et al., 2011; El-Haddad et al., 2009). Important fractions of Ba (37 % of the Ba mass), Mo (36 % of the Mo mass), Pb (36 % of the Pb mass), and Zn (38 % of the Zn mass) are also reported. Cu, Ba, Zn and Fe are indicators of the brakes abrasion (Johansson et al., 2009) while the presence of Zn could be related to the mechanical abrasion of tires (Amato et al., 2011). In addition, the Cu/Sb ratio of 8.1 obtained from the source profile of this factor remain within the range of 5.35 ± 2.9 reported in a road tunnel study in Switzerland to account for emissions factors for the metals originating from traffic (Sternbeck et al., 2002). The important share of EC observed in the source profile of this factor and the remarkable correlation ($R = 0.83$) between the source profile of this factor and EC are indications that direct traffic emissions are the controlling sources. However, the important shares of Fe, Sb, Cu, and Zn are also indications of a non-negligible contribution of non-exhaust primary traffic emissions originating from brakes and tires abrasion (Dall'Osto et al., 2013; Gianini et al., 2012). Weekdays-to-weekends variation (Fig. 4b) is logical with an increase in the use of vehicles for transportation during working days and accounting for an increase in road transport emissions (Baldasano et al., 2008; Waked et al., 2012).

The contribution of traffic emissions seems to be much lower in Lens than in other urban background sites like in Milan (27 % of PM₁₀; Marcazzan et al., 2003) and Madrid, Spain (34 %; Querol et al., 2004) but comparable to the reported contribution of 4 % in Dublin, Ireland (Yin et al., 2005) and 8 % in north west Germany (Beuck et al., 2011). The differences observed for the traffic contributions among these regions could be related to many reasons. First, it should be recalled that the sampling site is located

**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

away from major roads. However, when considering gaseous traffic emissions and their secondary processing into particulate matter, the road transport sector (on a regional scale) may finally account for a much more significant contribution to PM₁₀. For instance, the major contribution of the nitrate-rich factor has to be partly related to NO_x emissions originated from this sector especially during the spring season when nitrates and dust markers were at their highest levels. Furthermore, vehicle emissions of volatile organics may also account for a significant fraction of secondary organic aerosols (Gentner et al., 2012; Bahreini et al., 2012). Traffic is usually considered as a local source. As PSCF is not well adapted to local emissions, a haze and uniform origin around the sampling site should be obtained from PSCF analysis. The shift of the PSCF hotspot to the N-NE sectors (Fig. 7) however suggests a more regional traffic origin, where transport from surrounding areas could occur.

3.3.6 Heavy oil combustion factor

The high fractions of Ni (52 % of Ni mass) and V (57 % of V mass) in the source profile of this factor (Table 4), along with the significant share of sulfate (> 10 % of SO₄²⁻ mass in this factor) indicates its heavy oil combustion origin, that might be more particularly related to power generation, shipping or industrial emissions (Querol et al., 2009). The share of this source at Lens is low and accounts for 4 % of the total PM₁₀ mass. Conversely, other source apportionment studies in Europe (Barcelona in Spain and Florence in Italy) revealed a larger share for the oil combustion in the range of 8–37 % (Viana et al., 2008). For sites closer to the one investigated here, a PMF study recently conducted for various places in the Netherlands also indicates a very low contribution of residual oil combustion species (about 1 %) to PM₁₀ on an average (Mooibroek et al., 2011).

In the literature, the presence of sulfate with V and Ni is proposed as markers for fuel oil/pet-coke combustion, petrochemical sources, or industrial emissions (Querol et al., 2002; Pandolfi et al., 2011; Alleman et al., 2010). In some studies, these markers are also associated with secondary aerosols sources (Viana et al., 2008 and references

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

therein). Looking at the negligible contributions of sea salt markers (Cl^- , Na and Mg) in this source profile, we can assume that this fuel oil combustion source is most probably related to land activities (power generation and industrial emissions) rather than shipping emissions. The major influence of industrial activities is moreover supported in our case by the value of the ratio of Ni/V (0.71) which is in line with the ratios of 0.96 and 0.75 previously reported for stainless steel and ceramic industries (Querol et al., 2007; Moreno et al., 2010), whereas shipping and power plant emissions generally display much lower ratios in the range of 0.2 to 0.4 (Viana et al., 2009). Indeed, the regional petro-chemistry industry mainly established on the seashore display a Ni/V ratio of 0.4 according to Alleman et al. (2010). Due to prevailing industrial activities in the region, the stainless steel and ceramic production sectors are thus expected to account for a large part of this heavy oil combustion factor. The continental origin of this factor is finally supported by the PSCF analysis (Fig. 7).

3.3.7 Aged marine

This factor is mostly characterized by important shares of Na (48 % of Na mass), Mg (53 % of Mg mass) and Sr (27 % of Sr mass) along with a negligible share of Cl^- suggesting that it mainly consist of aged marine aerosols (Dall'Osto et al., 2013; Beuck et al., 2011). The important contribution of 19 % of the total PM₁₀ mass for this source during this study is in agreement with the value of 18 % reported in Barcelona (Dall'Osto et al., 2013) and higher than the value of 10 % reported in north west of Germany (Beuck et al., 2011). This last site is distant by more than 250 km from the sea, consistent with a lower influence of sea-salt markers.

This source follows a strong seasonal variation with the highest contribution (23 %) in the summer season. This seasonal variation is also observed for the sulfate-rich factor with a contribution increasing from 14 % to 19 % during summer. The comparable seasonal variation observed among the sulfate-rich and the aged marine aerosol factors along with an important share of sulfate (22 % of the sulfate mass) suggests that these marine aerosols acquire an anthropogenic signature as a result of the combustion of

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

fuel in shipping emissions and/or petrochemical activities located on coastal areas. Indeed, the contribution of marine aerosols markers in this source profile is 35 % whereas anthropogenic emissions account for the remaining 65 %. As a matter of fact, the PSCF analysis strongly points out to potential associations with the Strait of Dover, presenting the highest ships density in the world, for this source (Fig. 7) and thus suggests an important share of shipping emissions as well as seashore petro-chemistry.

3.3.8 Fresh sea salt factor

This sea-salt factor typically accounts for 8 % on a yearly average of the total PM₁₀ mass concentration. It is characterized by the presence of Na, Cl⁻, and Mg with 47, 84, and 25 % of the mass of each species accounted for in this factor, respectively. Minor contributions (less than 10 % of their total mass) of Ca, nitrate, Al, Ti, and K (Table 4) are observed, together with some share of Sr (12 % of Sr mass). These markers (Cl⁻, Na, and Mg) are clearly related to sea salt, sea spray and marine aerosols sources in the literature (Pio et al., 1996) with Mg mostly reported as a tracer in Central and Northern European sites (Belis et al., 2013). The presence of Sr in the source profile of the sea salt factor is not unusual due to the fact that this compound is present in the sea salt composition along with Ca (Viana et al., 2008). Both are present together in calcareous algae and foraminifera found in large numbers throughout the photic zone of the ocean.

The contribution of this factor in Lens (8 % of the PM₁₀) is lower than the contribution of 35 % reported at an urban site in Ireland (Yin et al., 2005) and to the contribution of 15 % reported by Dall'Osto et al. (2013) at an urban site in Barcelona (Spain). It is in the same range as results from more continental sites, like the reported value of 9 % in North-West Germany (Beuck et al., 2011). Remarkably, the ratios of Ca/Na and Mg/Na of 0.04 and 0.09, respectively, obtained from the source profile of this factor are in total agreement with the ratios of 0.04 and 0.12 expected for sea salt and reported in the literature (Millero, 1973; Seinfeld and Pandis, 2006) suggesting that the source profile obtained from this factor is rather free from mixing with other salt sources. In the

same way, the contributions of OC and levoglucosan are very low, indicating a large decoupling of these sources despite their common maximum in winter. The marine origin of this factor is confirmed by PSCF analysis outputs (Fig. 7).

3.3.9 Focus on the carbonaceous fractions (EC and OC)

As commonly observed in urban environments (Zhang et al., 2007), organic aerosols globally dominates ambient particulate matter by mass in this study. A better knowledge of its main emission sources and transformation processes in the atmosphere is still needed for the elaboration of efficient PM reduction action plans (Fiore et al., 2012). Conversely, elemental carbon presents relatively low concentrations, but is gaining more and more attention due to its climatic relevance as well as its potential use as an indicator of PM health effect (US EPA, 2012). The present section focuses on these two carbonaceous fractions.

The different factor contributions of EC obtained from the PMF analysis are presented in Fig. 8. The road transport sector represents the major source of EC with 37 % of its mass on an annual basis. EC also have important contributions from biomass burning and oil combustion sources, for 22 % and 21 % of its total mass, respectively. Such important contributions from these sources directly associated with combustions are well expected. The most unexpected feature is the contribution of 16 % on a yearly average of the primary biogenic factor to the EC mass, being as large as one third of the total EC load in summer. Such a behavior should be investigated further, to understand if it is related to an actual atmospheric mixing process and/or to limitations in the PMF de-convolution method, and/or to EC-OC analysis artifacts. However, it should be recalled that total EC concentrations are rather low and that a limited occurrence of one or several of the latter hypotheses would easily lead to the result observed here.

The different contributions of OC sources retrieved from the PMF analysis are presented in Fig. 9. On an annual basis, major contributors of OC are biomass burning (24 % of OC mass) and primary biogenic emissions (17 % of OC mass). These results underline the large contribution of modern carbon (as opposed to fossil fuel combustion

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

– derived carbon) within organic aerosols, which is in good agreement with previous studies (Yttri et al., 2007, 2011; Favez et al., 2010; Caseiro et al., 2009; Puxbaum et al., 2007). The sulfate-rich secondary factor appears to be the third contributors to total OC, illustrating similarities within the processes leading to the formation of ammonium sulfate and secondary organic aerosols (SOA) as previously observed (Lanz et al., 2007; Ulbrich et al., 2009). Besides these three major OC contributors, it is noteworthy to mention that other sources (nitrate-rich secondary aerosols, aged marine, traffic, oil combustion, and dust) are still making up together 44 % of the total OC load. Therefore, the important contributions for primary biogenic emissions and secondary aerosols such as the sulfate-rich and the nitrate-rich sources highlight the important influence of many primary and secondary emissions sources such as fungal spores and anthropogenic/biogenic signature for SOA. While the biogenic origin of SOA could be hidden within the primary biogenic fraction, its origin is mostly linked to sulfate-rich and nitrate-rich organics which are basically originating from anthropogenic sources. Certainly, seasonal variation of the primary biogenic emissions with maximum contributions during summer is expected due to intense solar radiation which favors photochemical reactions that leads to the formation of SOA. This seasonal variation was also observed for the sulfate-rich source with maximum contribution during the summer season. However, and for the nitrate-rich source, the identical contribution observed during summer and winter could be related to measurements artifact for nitrate in summer, where a part of particulate nitrate is volatilized due to its thermal instability. Emissions from fungal spores could also account for one third of this fraction. Indeed, using an arabitol/OC ratio for fungal spores of 0.092 (Bauer et al., 2008), the contribution of fungal spore to total OC would be 6 %. The processes involved could be numerous, including direct emissions, mixing during transport, and co-processing during ageing or thermal condensation in the particulate phase. Such a diversity of OC sources is usually derived from receptor models applied to filter-based off-line datasets and contrasts with results obtained from similar models applied to Aerosol Mass Spectrometer (AMS) datasets (Ulbrich et al., 2009; Lanz et al., 2010). Moreover, PMF analyses conducted on filter-

based datasets do usually not lead to the identification of a single factor mainly made of SOA, while PMF analyses applied to organic mass spectra indicate a predominance of Oxygenated Organic Aerosols (OOA) commonly considered as SOA (Zhang et al., 2007; Ulbrich et al., 2009; Lanz et al., 2010).

5 These discrepancies are firstly due to the fact that AMS studies focuses on submicron aerosols whereas filter-based receptor models generally investigate larger size fractions. In the present study, the investigation of PM₁₀ allows evidencing the significant influences of primary biogenic emissions and mineral dust particles (that could both reasonably be mainly attributed to coarse particles) on the total burden of organic matter within the main regulated PM size fraction in Europe.

10 Further, filter-based receptor modeling as performed here rely on a large diversity of organic and inorganic compounds while AMS studies are based on a very detailed description of the organic fraction only. Specifically, the use of trace metal elements helps differentiating between various fossil fuel combustion sources (in our case road transport, maritime transport and terrestrial heavy oil combustion) which is not straightforward when applying PMF to organic mass spectra. This diversity of tracers in filter based methods might also help differentiating between SOA formed through a chain of successive (photo-chemical) oxidation processes and Oxygenated Primary Organic Aerosols (OPOA) that undergo gas-particle partitioning as a function of their volatility

20 (Robinson et al., 2007). Indeed, biomass burning emissions, which contain high amounts of such OPOA (Grieshop et al., 2009), are commonly found in larger proportions when derived from filter-based source apportionment studies compared to PMF analyses applied to AMS organic datasets (e.g. Favez et al., 2010). In the present study, elevated contributions of biomass burning are obtained during the residential heating period (respectively 28 % and 49 % for the fall and winter seasons). Conversely, low contributions of the secondary nitrate-rich factor to total OC are obtained during this period (4 % on a yearly average), whereas high nitrate concentrations have been measured on our filter samples and AMS studies generally report a good correlation between nitrate and Semi-

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

5 Volatile OOA (Lanz et al., 2007; Ulbrich et al., 2009). It might be hypothesized that this semi-volatile organic fraction derived from biomass burning emissions could be directly related to the wood burning factor when using datasets containing a high diversity of tracers while it is mixed with other SV-OA when using organic mass spectra only. This hypothesis seems to be confirmed by recent combinations of organic mass spectra with other compounds. For instance, SV-OOA has recently been strongly related to biomass burning during wintertime when combining AMS datasets to PTR-MS datasets (Crippa et al., 2013). It then appears that PMF analyses conducted on a large variety of tracers could allow a better quantification of the prevalent contribution of biomass burning emissions on organic matter during the cold season by differentiating OPOA originating from biomass burning from other SV-OOA related to the secondary nitrate-rich factor, despite both of them partly following the same kind of gas-particle partitioning mechanisms. This is of prime interest when considering source apportionment studies in the frame of the elaboration of PM reduction action plans.

15 Nevertheless, it should also be kept in mind that, on a yearly basis, SOA may represent a major fraction of the total organic matter in Lens even if no unique SOA factor has been identified. The use of specific secondary organic tracers, such as n-heptacosanoic acid and n-octacosanoic acid could help better enlightening the abundance of this fraction in ambient air when applying PMF analysis on off-line datasets (Wang et al., 2012).

3.3.10 Major sources for the highest PM₁₀ daily concentrations

This section focuses on the major sources responsible for the most severe pollution events during the period of study. Due to the low number ($n = 5$) of $50 \mu\text{g m}^{-3}$ daily threshold exceedances, preventing sound statistics, and considering an uncertainty of 15% on the measurement of the PM₁₀ mass, a limit of $42.5 \mu\text{g m}^{-3}$ has been artificially set to allow isolating the highest PM₁₀ concentrations samples. In the following, the term “exceedances” refers to this limit and concerns 11 days: 15 March 2011, 21 March 2011, 27 March 2011, 17 April 2011, 23 April 2011, 26 April 2011, 3 August

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2011, 7 November 2011, 22 November 2011, 30 January 2012, 11 February 2012. The exceedance day of 3 August 2011 is considered as an exception and excluded from the average because it is the only one during summer. As typically observed in the region, exceedance days are in paramount part during the cold period. Comparison of the PM₁₀ mass balance during these exceedance days and the average mass balance calculated from November to April for the other days shows that the global chemical composition of PM₁₀ in Lens during exceedances is mainly characterized by a larger fraction of nitrate (representing 31 % of PM₁₀ mass vs. 25 % for the cold season average), organic matter (relatively stable around 27 % during and out of exceedances) and to a lesser extent by higher fractions of ammonium and sulfate. The changes of contributions of sources between exceedance days and the cold season average are further investigated with the results of the PMF analysis. During exceedance days, biomass burning emissions contribute to 18 % of PM₁₀ compared to 25 % for cold season average, and the secondary nitrate-rich and sulfate-rich factors together comprise 54 % of PM₁₀ mass (34 % for nitrate-rich and 20 % for sulfate-rich) compared to 29 % on annual average.

These results illustrate the influence of photo-chemical processes on occurrences of higher PM₁₀ concentrations. Indeed, nitrate, sulfate and ammonium could reasonably be considered as mainly originating from secondary mechanisms and the chemical composition of PM₁₀ during exceedance days is also characterized by higher OC/EC ratio (≈ 5 during exceedance days compared to ≈ 3 during non-exceedance days) which can be partly related to SOA formation.

Such photo-chemical processes are favored by high pressure systems, corresponding in this region of Europe to air masses arriving from the continental sector. This could be evidenced by the investigation of metal-enrichment factors. Indeed, the enrichment factors in aerosol particles, relative to the average composition of the upper continental crust (EF_{crust}) are frequently used to decipher between natural and anthropogenic origins of metals in the atmosphere (Gao et al., 2002; Lim et al., 2010; Alleman et al., 2010). The crustal EF in PM₁₀ is defined as, $EF_{\text{crust}} = (E_{\text{atm}}/R_{\text{atm}})/(E_{\text{crust}}/R_{\text{crust}})$ where

**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

E and R are the concentrations of the element of concern and the reference element respectively for aerosols (atm) and crustal material (crust). Here, Ti was used as the reference element based on the average chemical composition of the upper continental crust given by McLennan (McLennan, 2001). Metals with EF_{crust} values close to unity are predominantly associated to crustal sources, while element with high EF_{crust} (> 10) are essentially from anthropogenic origins. On average, V, Ni, Cu, Zn, Pb, Mo and Sb are clearly mainly from human activities with EF_{crust} above 10 and up to 5000. It is worthy to consider separately PM₁₀ samples collected during exceedance and non exceedance days. Indeed, as expected, most of the elements display higher EF_{crust} values with high PM₁₀ concentrations except for Na, Mg and in a lesser extent Sr. These tracers of marine sources with higher enrichment during low PM₁₀ content days indicate that exceedance is generally associated to continental air masses while oceanic air masses are much cleaner. However, it cannot be concluded from the present study whether highest daily concentrations have to be mainly related to long-range transport. The impact of trans-boundary emissions during daily threshold exceedances would require the use of advanced Chemical-Transport Model (CTM).

4 Conclusions

In this work, chemical composition, concentrations and potential emissions sources of PM₁₀ at a North-Western Europe regional urban background site was investigated based on filter samples collected from March 2011 through March 2012. As previously observed in this area, PM₁₀ levels are dominated by OM, nitrates and sulfates in all seasons.

The identification of PM₁₀ potential emissions sources performed by PMF highlighted the importance of biogenic emission sources which should not be neglected in source apportionment studies. Furthermore, tests of the different methodologies for the calculation of uncertainties in the PMF model show that the choice of the methodology is very important and could largely affect the results obtained. The calculation of uncertainties

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

should also be improved in the PMF studies since no specific methodology in the PMF is available up to now. After numerous tests performed, nine sources were identified (secondary nitrate, secondary sulfate, aged marine aerosols, fresh sea salts, mineral dust, primary biogenic emissions, biomass burning, oil combustion and traffic emissions). Secondary nitrate-rich- and sulfate-rich factors together were found as dominant sources with a contribution of 28 % followed by aged marine aerosols (19 %), biomass burning and mineral dust (13 %). Less important contributions were estimated for fresh sea-salt (8 %), and primary biogenic emissions (9 %) while primary traffic emissions (6 %) and heavy oil combustion (4 %) were defined as the minor source contributors. Secondary nitrate increased during the spring season due to its semi-volatile character as well as more intensive agricultural activities and road transport emissions during this period of the year. Mineral dust (notably characterized by Ca, Fe, Al and Ti) exhibited a similar temporal variation, probably due to meteorological conditions favoring re-suspension from soils and roads. Among other identified sources, biomass burning emissions increased in winter due to a large use of wood burning for household heating while primary biogenic emissions increased in summer due to a higher biological activity. The sea salt source characterized by the presence of Cl, Na and Mg showed also a seasonal variation with maximum concentrations in the winter season. Similarly to the sulfate-rich source, aged marine aerosols, illustrated by important amounts of Na, Mg and Sr, showed a significant seasonal variation with the highest contribution recorded during summer due to enhanced photo-chemistry.

To our knowledge and according to the review of Belis et al. (2013), the present study is the first PM₁₀ source apportionment study combining inorganic and organic species in France. Furthermore, the significant contributions of biogenic emissions have been highlighted also for the first time in such a PMF analysis. However, future works are necessary in order to identify more potential sources and to be more accurate in the categorization of a factor to a specific source, including the aged marine aerosol source. These improvements should focus on the addition of organic tracers compounds such as Polycyclic Aromatic Hydrocarbons (PAH), hopanes and steranes,

which are markers for many anthropogenic sources and would help in the elaboration of additional factors. In addition, the quantification and the identification of biogenic Secondary Organic Aerosols (SOA) such as oxidations products of isoprene, α -pinene and β -caryophyllene would also help in the apportionment of the secondary biogenic emissions sources. In addition, the need for measurements on a 12 h or 4 h basis along with sampling for the whole week-end period (Saturday and Sunday for each week) would be also helpful for source and processes identification. The problem of rotational ambiguity preventing the PMF to sometimes separate two sources that do not vary independently could be reduced after these improvements are made.

Supplementary material related to this article is available online at <http://www.atmos-chem-phys-discuss.net/13/25325/2013/acpd-13-25325-2013-supplement.pdf>.

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25358

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



References

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Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

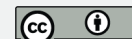
Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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- 30

**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**

A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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Source apportionment of PM₁₀ in Lens, France by means of the PMF modelA. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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Source apportionment of PM₁₀ in Lens, France by means of the PMF modelA. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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ACPD

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**Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model**

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

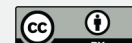
Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

25371



Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 1. Average measured concentrations of PM₁₀ and its major components in spring, summer, fall and winter 2011–2012.

Species	Spring 2011 ($\mu\text{g m}^{-3}$)	Summer 2011 ($\mu\text{g m}^{-3}$)	Fall 2011 ($\mu\text{g m}^{-3}$)	Winter 2012 ($\mu\text{g m}^{-3}$)
PM ₁₀	29.2	13.7	19.3	19.7
OM	6.07	3.19	7.11	6.68
EC	1.07	0.53	1.27	0.91
NO ₃ ⁻	8.44	2.00	3.02	4.36
SO ₄ ²⁻	2.85	1.99	2.22	2.15
NH ₄ ⁺	3.05	0.92	1.39	1.78
Cl ⁻	0.58	0.35	0.59	1.28
K	0.14	0.08	0.14	0.17
Na	0.61	0.52	0.53	0.89
Mg	0.11	0.08	0.08	0.10
Ca	0.51	0.21	0.23	0.19
Levogluconan	0.11	0.01	0.24	0.31
∑ Sugars alcohols ^a	0.03	0.06	0.05	0.02
∑ Metals and trace elements ^b	0.61	0.24	0.36	0.26

^a Arabitol, Sorbitol, Mannitol and Glucose.

^b Ba, Cu, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn, Al, Fe and Ti.

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Table 2. Parameters used for the tests of uncertainty calculation methods and the associated stability results obtained in the base run.

Parameters	Test 1	Test 2	Test 3	Test 4
Species included	EC, OC ⁺ , Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Levoglucosan, Polyols, Ba, Cu, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn, Al, Ca, Fe, K, Na, Mg, Ti et PM ₁₀	EC, OC ⁺ , Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Levoglucosan, Polyols, Ba, Cu, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn, Al, Ca, Fe, K, Na, Mg, Ti et PM ₁₀	EC, OC ⁺ , Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Levoglucosan, Polyols, Ba, Cu, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn, Al, Ca, Fe, K, Na, Mg, Ti et PM ₁₀	EC, OC ⁺ , Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Levoglucosan, Polyols, Ba, Cu, Mo, Ni, Pb, Rb, Sb, Sr, V, Zn, Al, Ca, Fe, K, Na, Mg, Ti et PM ₁₀
Uncertainty calculation methodology	Gianini methodology for all the species	Polissar methodology for all the species	Gianini methodology for all the species except for trace and metal elements where the relative uncertainty was used instead of CV	Polissar methodology for all the species except for trace and metal elements where the relative uncertainty was used instead of CV
Number of Factors	9	9	9	9
Scaled residuals	For many species, scaled residuals were not within the range of -3 and + 3 the standard deviation	For many species, scaled residuals were not within the range of -3 and + 3 the standard deviation	Set between -3 and + 3 the standard deviation	Set between -3 and + 3 the standard deviation
Bootstrap Results	For some factors, just 58% of the runs are correlated between base run and the bootstrap runs	For some factors, just 66% of the runs are correlated between base run and the bootstrap runs	Good correlation between factors: more than 92% of the runs are correlated between the base run and the bootstrap runs	Good correlation between factors: more than 92% of the runs are correlated between the base run and the bootstrap runs

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 3. Values of the coefficient of correlation (R^2) between measured and modeled concentrations in the base run.

Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na	NH ₄ ⁺	K	Mg	Ca	EC	OC*
0.999	0.999	0.996	0.986	0.997	0.831	0.973	0.885	0.947	0.976
Polyols*	Levo	Ba	Cu	Mo	Ni	Pb	Rb	Sb	Sr
0.967	0.988	0.436	0.717	0.433	0.785	0.810	0.916	0.624	0.756
V	Zn	Al	Fe	Ti	PM ₁₀				
0.952	0.609	0.908	0.928	0.859	0.968				

Table 4. Average yearly contributions of the identified sources in % of the measured PM₁₀ mass.

	Traffic	Aged marine	Biogenic emissions	Biomass burning	Mineral dust	Nitrate rich	Sulfate rich	Oil combustion	Fresh sea-salt
Cl ⁻	0.0	0.0	0.0	8.9	1.7	2.0	3.8	0.0	83.7
NO ₃ ⁻	4.3	13.2	0.0	10.0	0.7	70.2	0.0	1.6	0.0
SO ₄ ²⁻	0.0	21.6	2.9	0.0	0.0	10.4	54.2	10.9	0.0
Na	0.0	48.3	3.7	0.0	0.0	0.0	0.9	0.0	47.1
NH ₄ ⁺	0.0	0.0	0.9	9.4	0.0	55.1	27.4	4.5	2.7
K	5.4	23.6	6.6	20.4	17.5	0.9	19.7	0.0	5.9
Mg	2.1	53.2	3.8	0.9	13.2	0.3	0.0	1.5	25.2
Ca	19.2	9.9	4.4	0.0	50.3	7.9	0.8	3.9	3.6
EC	44.6	2.1	9.1	26.2	0.0	0.0	0.0	15.1	2.9
OC*	8.3	0.0	11.3	33.5	15.1	4.1	17.0	7.5	3.2
Polyols*	0.0	7.8	66.2	10.5	0.0	11.3	0.9	3.3	0.0
Levo	4.9	2.3	0.9	83.5	0.0	0.0	0.1	6.1	2.2
Ba	37.3	5.2	9.7	6.4	17.7	11.7	0.0	10.4	1.6
Cu	52.4	5.4	8.3	14.0	1.7	5.5	4.4	7.4	0.9
Mo	36.3	4.9	1.2	10.0	17.8	21.3	7.4	1.2	0.0
Ni	9.6	7.3	0.0	0.1	12.5	11.6	2.9	51.8	4.3
Pb	36.0	2.9	0.0	21.7	4.2	10.4	22.6	1.1	1.2
Rb	12.2	2.6	0.4	21.6	35.1	4.0	21.8	0.0	2.5
Sb	43.8	4.2	6.5	15.3	5.4	11.2	6.0	7.0	0.7
Sr	9.2	26.8	6.1	4.0	34.1	3.9	0.0	4.3	11.6
V	0.0	8.2	2.0	1.2	12.2	9.0	4.7	57.4	5.3
Zn	38.1	0.0	2.4	17.7	9.2	9.1	16.2	4.4	3.0
Al	13.9	0.0	2.0	3.1	58.5	8.2	0.0	9.0	5.3
Fe	33.5	5.8	0.0	15.5	31.3	11.7	2.3	0.0	0.0
Ti	0.0	2.5	2.9	3.1	72.6	0.0	8.2	6.9	3.9
PM ₁₀	7.1	13.2	6.1	14.8	13.5	20.4	15.7	3.5	5.7

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.



Fig. 1. Localization of the sampling site and surrounding region.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source
apportionment of
PM₁₀ in Lens, France
by means of the PMF
model

A. Waked et al.

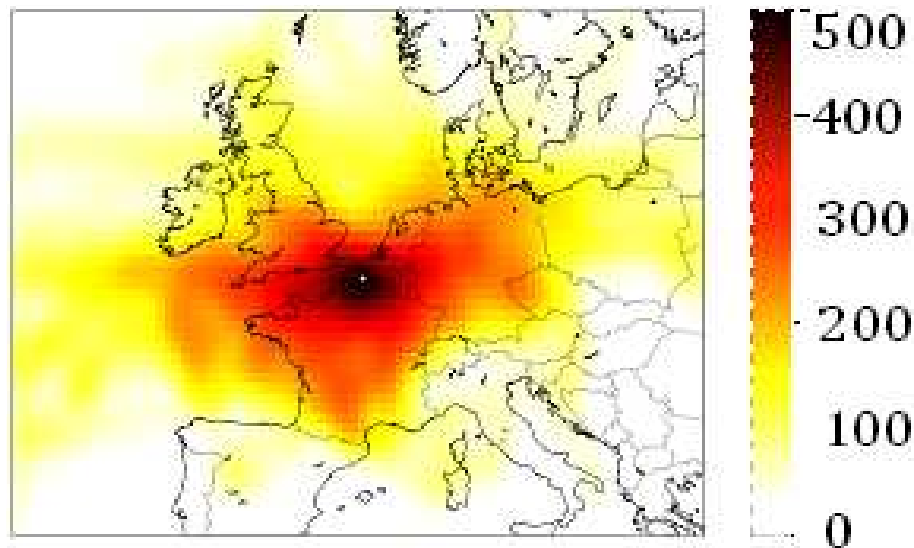


Fig. 2. Calculated back-trajectory density used to determine the weighing function coefficients, scaled by the number of back-trajectories passing through each cell.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

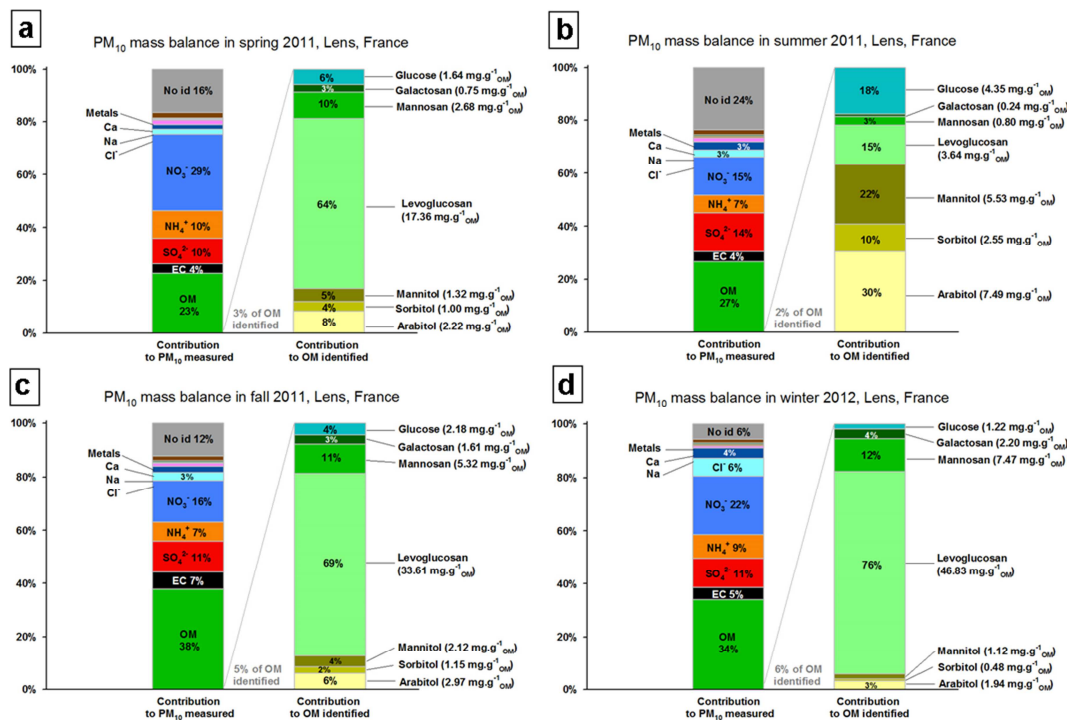


Fig. 3. PM₁₀ mass balance at Lens in spring (a), summer (b), fall (c) and winter (d) 2011–2012.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

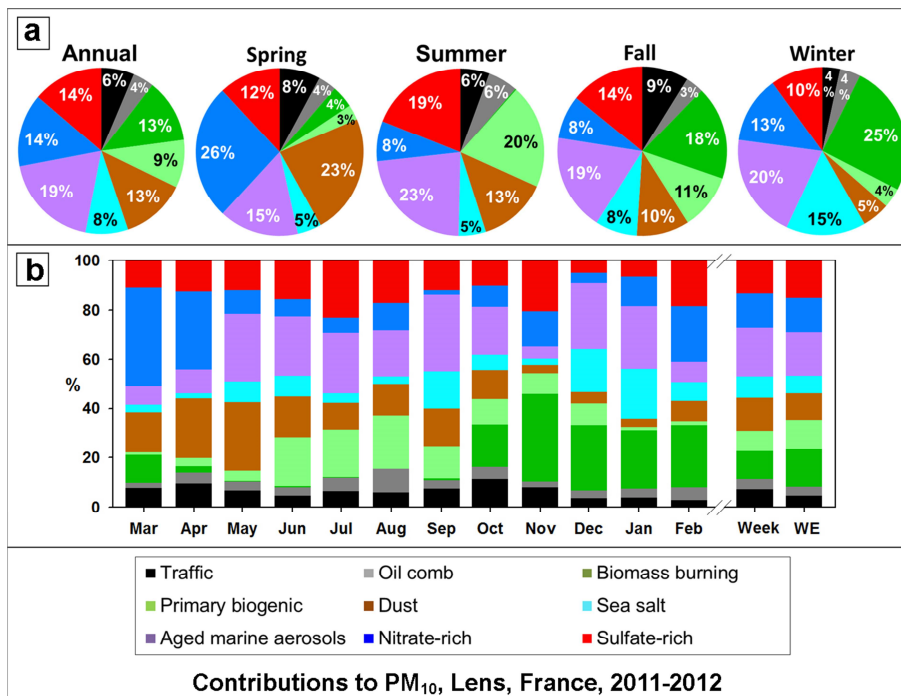


Fig. 4. Contributions of the identified sources in % to total PM₁₀ mass concentration for the full year and according to the seasons (a) and on a monthly basis along with weekdays to weekend's variations (b).

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

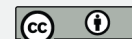
◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

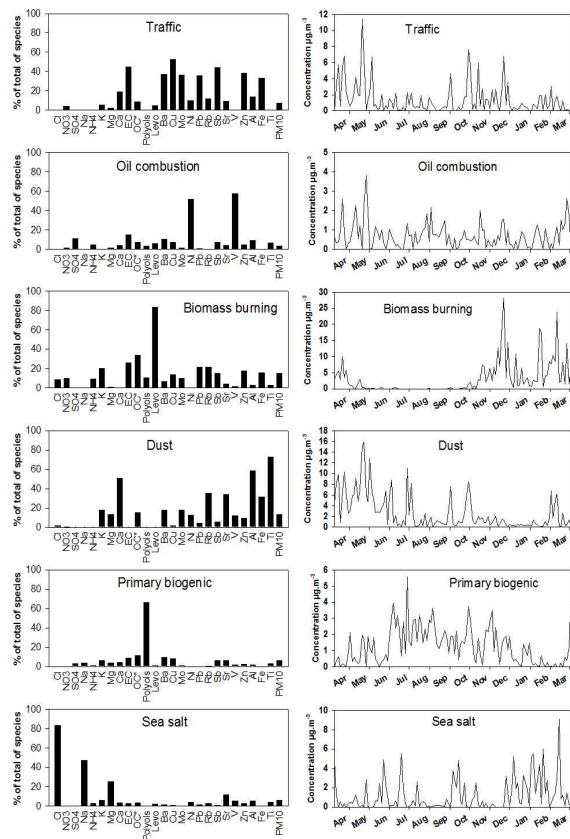


Fig. 5. Source profiles (left) and temporal evolutions (right) of contributions of primary factors.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

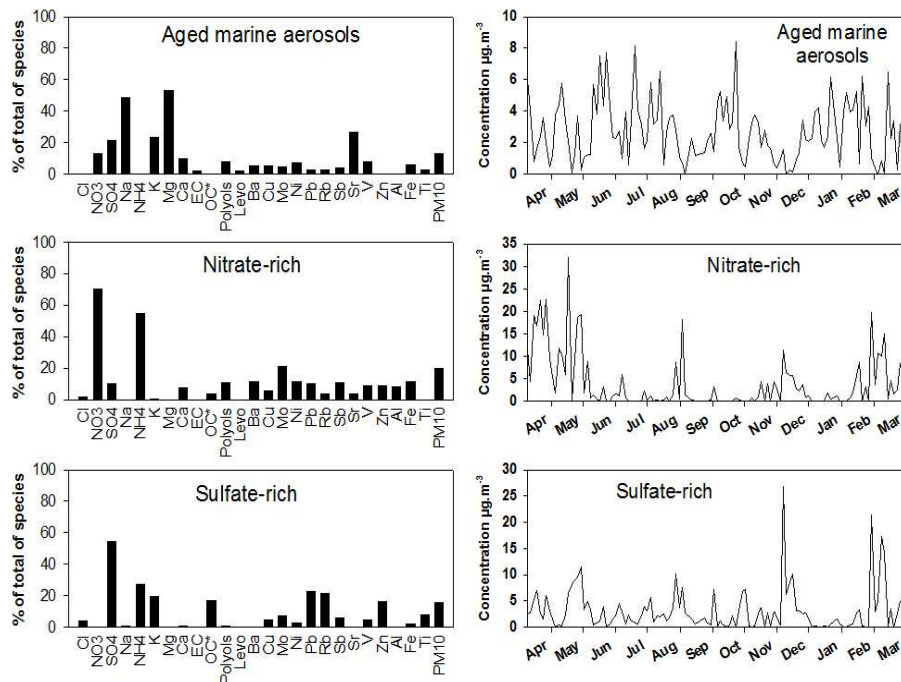


Fig. 6. Source profiles (left) and temporal evolutions (right) of contributions of secondary factors.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

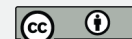
◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

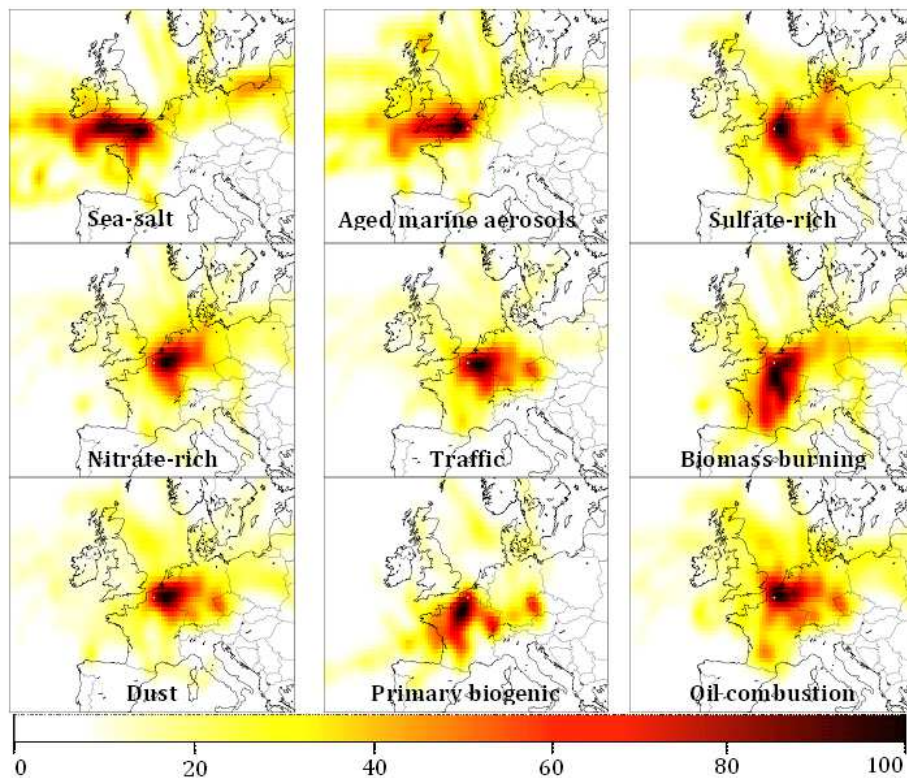


Fig. 7. Normalized PSCF values (in %) for all the identified sources in Lens.

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

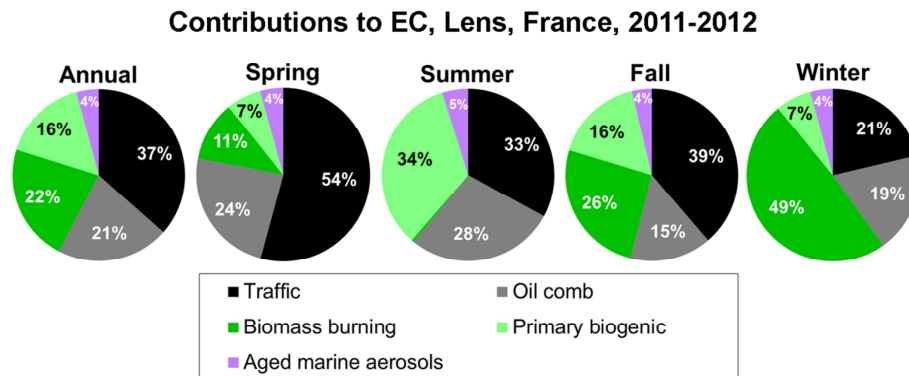


Fig. 8. Contributions of the identified sources in % to total EC mass concentration for the year 2011–2012, spring, summer, fall and winter.

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	

Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

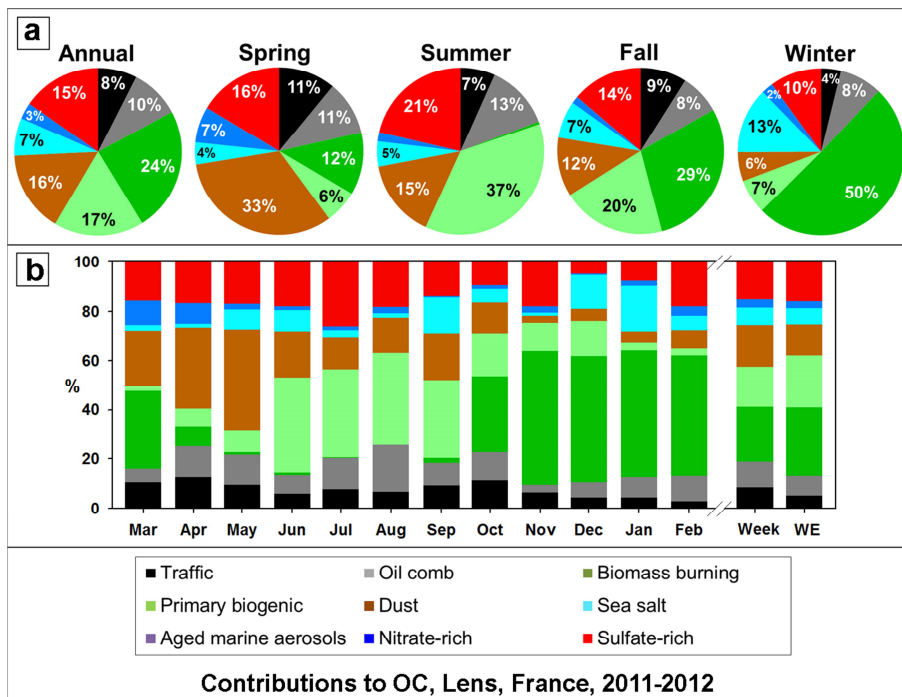


Fig. 9. Contributions of the identified sources in % to total OC mass concentration for the year 2011–2012, spring, summer, fall and winter.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

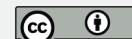
◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Source apportionment of PM₁₀ in Lens, France by means of the PMF model

A. Waked et al.

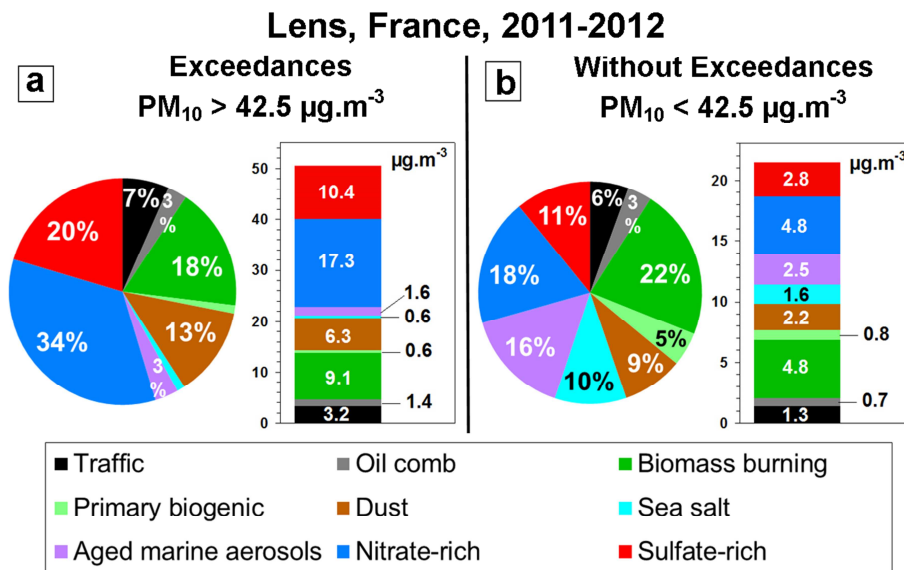


Fig. 10. Contributions of sources during exceedances (a) and for annual average (march 2011 to march 2012) without such exceedances (b) in Lens in 2011–2012.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	