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Source apportionment of PM_{2.5} and their associated metallic elements by positive matrix factorization at a traffic site in Constantine, Algeria

Lamri Naidja^{1,2,3} · Hocine Ali-Khodja¹ · Salah Khardi⁴ · Fairouz Bencharif-Madani¹ · Ahmed Terrouche¹ · Kanza Lokorai¹ · Mokhtar Bouziane¹ · Aurélie Charron⁵

Abstract

Chemical characterization of PM_{2.5} (major and trace elements) was carried out for a source apportionment study of PM_{2.5} at a traffic site in Constantine, Algeria, from March 2017 to March 2018. For this purpose, several tools were used of which PMF, CPF, PSCF, Spearman correlation matrix and HYSPLIT back trajectories. The mean annual concentration of PM_{2.5} at the sampling site was $54.07 \pm 28.81 \mu\text{g}/\text{m}^3$. This value is much higher than the annual threshold recommended by the WHO of $10 \mu\text{g}/\text{m}^3$. Based on the PMF modelling, five sources were identified: sea salts (15.1%), industrial activities (18.9%), non-exhaust emissions (wear of brakes, tires and road surfaces) (24.2%), exhaust emissions (mixed diesel and gasoline engine exhaust) (15.8%) and mineral dust (25.9%) as the main sources of metallic aerosols at the sampling site in Constantine. Our results revealed that anthropogenic activities (traffic and industry) contributed 59% to all emissions, while natural sources (mineral dust and sea salt) accounted for 41%. Traffic-related sources are the major contributor to anthropogenic emissions (40%) with non-exhaust emissions being the dominant source. As anthropogenic emissions are the leading sources, their control is of utmost importance for improving air quality. The concentrations of PM_{2.5} tend to increase appreciably with temperature and wind speed for a relative humidity less than 60%. The contribution of vehicular emissions is affected by relative humidity and temperature while industrial emissions are affected mainly by wind intensity and direction. Relative humidity, temperature and wind speed are determining parameters of mineral dust. There are seasonal variations in the contributions of the PMF-derived sources. Summer undergoes significant contributions of three factors: sea salts, industrial emissions and mineral dust. Spring is affected mainly by industrial emissions and non-exhaust emissions. Winter experiences a drastic contribution of the exhaust emission factor. Autumn is the least affected season by mineral dust and sea salts. It is rather affected by industrial and traffic emissions.

Keywords Metallic aerosols · PM_{2.5} · PMF · CPF · PSCF · Source apportionment · Traffic site

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Introduction

Particulate matter (PM) is a complex mixture of small particles and liquid droplets whose physical and chemical characteristics vary by region and time. Common chemical constituents of PM include sulphates, nitrates, ammonium, other inorganic ions, metals, elemental carbon and organic compounds.

The health effects were related to the distribution of PM sizes, the latter were divided into two parts: PM₁₀ (coarse particles) and PM_{2.5} (fine particles). The potential impact of smaller particles on human health has been revealed by several authors (Biersteker 1976; Bevan and Manger 1985; Quackenboss et al. 1989). Boubel et al. (2013) showed that PM_{2.5} could not be retained in the upper part of the

respiratory system and can accumulate in the lungs and enter the airways, due to Brownian motion, unlike PM_{10} which could be retained. Besides the size, the effect of aerosols on health is also a function of their chemical composition. Most heavy metals are dangerous because they tend to bio-accumulate in the human body. Metallic elements cannot be degraded, and can be transported by air, and enter water and human food supply. However, at higher (although relatively low) concentrations they can become toxic (Järup 2003). Many studies have linked certain metals to several health problems such as cardiovascular disease, heart disease, aggravated asthma and lung cancer (Kampa and Castanas 2008; Sudheer and Rengarajan 2012).

The problem of air pollution is clearly ignored by many African countries, largely because it is not seen as a priority over economic development. Therefore, these countries are clearly in no rush to solve air quality problems. In many African countries, air quality data is scarce and virtually absent, especially in sub-Saharan countries. The lack of data is therefore one of the main constraints to understanding the harmful effects on human health of fine particles.

Ambient air quality improvement strategies involve reducing emissions from primary sources. It is therefore essential to be able to identify and distribute the contributions of these sources and then implement effective regulations and policies that reduce the levels of particulate pollution. Receptor models are often presented as a reliable tools which allow the identification of the contribution of emission sources and which require speciation data for the aerosol fraction sampled at a receptor site (Hoke 2010).

In Algeria, few studies have been carried out to identify PM emission sources. Terrouche et al. (2016) used the varimax rotated factor analysis, a multivariate technique. Their study targeted only eight metallic elements. Bencharif-Madani et al. (2019) and Talbi et al. (2018) both used the PCA technique which did not differentiate separate sources. Moreover, these methods do not provide a quantitative distribution of mass contributions.

In addition, the absence of a non-negativity requirement implies that PCA can give factors with negative loadings and could promote physically unreasonable results (Paatero and Tapper 1994). To solve this problem, specific methods such as positive matrix factorization (PMF) have been specifically developed (Paatero and Hopke 2003). In order to distinguish between nearby and distant sources, the *CPF* and *PSCF* tools have been used.

Materials and methods

Description of the sampling site

Constantine is a city in northeastern Algeria, located about 390 km from Algiers, the country's capital (Fig. 1) and 63 km from the Algerian coast. This metropolis is the third most populous city in the country. The agglomeration of Constantine had 943,112 inhabitants in 2015, of which only 54% live in the municipality of Constantine (448,000 inhabitants). This city is characterized by a hot and dry semi-arid climate in summer with an average maximum temperature of 36 °C and a humidity of around 25% while



Fig. 1 Location of the sampling site and city of Constantine, Algeria

the winter is cold and wet. Precipitation totals 560 mm per year, with monthly precipitation varying from 0 to 80 mm (Bencharif-Madani et al. 2019).

To attain the objectives of our work, an urban traffic site was chosen in Zouaghi, Constantine (36° 31' North, 6° 62' East, 649 m above sea level). The site is located 5.65 km southwest of the city of Constantine (Fig. 1). It has around 26,000 inhabitants.

The national vehicle fleet is composed approximately of 50% vehicles running on gasoline and the remaining 50% on gas-oil. However, gas-oil represents 70% of total fuel consumption.

The nearest main road (national road N79) is about 5 m west of the study site and has two lanes of traffic in each direction (Fig. S1). This road connects the city of Constantine with the new town, Ali Mendjli, as well as other cities such as Batna, Oum El Bouaghi and Biskra. The East–West motorway is 1.7 km south of the site. The sampling site is also surrounded by three urban areas. These are located to the east, south and northwest of the site (Fig. S1). In addition, Mohamed Boudiaf Airport is located 2.86 km south of the sampling site.

Sampling campaign

PM_{2.5} was collected on quartz fibre filters (WHATMAN®, diameter 47 mm, QMA grade) at the frequency of one sample every 3 days at the entrance of the Faculty of Earth Sciences in Zouaghi. The sampling campaign ran from March 15, 2017 to March 15, 2018, using a low volume LVS (low volume sampler) brand “Tactical Air Sampler” (Fig. S2). Each sampling was carried out for a period of 24 h with a suction flow rate of 5 l of air/min.

The sampler is attached to a metal pipe at a height of 1.80 m and at a distance of approximately 5 m from the N79 national road. It operates from a battery that can power the sampler for at least 24 h continuously, making the sampling site independent of mains power.

The exposed filter is recovered within hours of the end of the sampling period to prevent damage to the filter or change in particle mass due to passive absorption or volatilization. The exposed filters are placed in petri dishes, placed with the exposed side of the filter facing up and kept in the desiccator for 48 h. They are weighed before and after each sampling using a microbalance with an accuracy of 1 µg (Shimadzu AUW 120D) to determine the mass of particles captured on the filters. All precautions are taken during the installation and recovery of the filters (wearing gloves, using the pliers) to avoid any contamination. A total of 120 samples were collected throughout the sampling period of 12 months.

Mineralization protocol

We have chosen the mineralization method proposed by Querol et al. (2001). Kemmouche et al. (2017) were able to demonstrate that the chosen protocol is the most effective since it promotes the complete recovery of all the elements by the essential use of hydrofluoric acid (HF) which remains the only reagent capable of releasing major elements and traces linked to silica with recovery rates greater than 80%. This protocol is best suited for source distribution studies, which require a large number of elements and where it is strongly recommended to consider total dissolution of samples using HF to ensure reliability of results.

First, the filters are cut in half. Half undergo the digestion protocol and the other half are stored in the fridge at 3 °C for further analysis. The half-filters are digested in a solution containing 1 ml of nitric acid (HNO₃) and 2 ml of HF in closed Teflon-PFA (PerFluorAlkoxy) vials in the oven at 90 °C overnight (at least 8 h of time). After cooling, 1 ml of perchloric acid (HClO₄) is added. Then, the containers are heated for 4 h on a hotplate at 240 °C until the acids have completely evaporated. 2.5 ml of HNO₃ are then added to the remaining dry residue and then diluted in a 25-ml flask, making up the volume with ultra-pure water (MilliQ) to obtain 5% HNO₃ solutions. Then the contents of each vial are transferred to test tubes and centrifuged for 2 min at 3000 rpm to remove residues from the mineral filters. The solutions obtained are stored in polypropylene (PP) bottles fitted with tight caps.

Filter analysis

The 120 PM_{2.5} samples, including the 5 blank filters, were analysed by two instruments, inductively coupled plasma emission spectroscopy (ICP-OES, Perkin Elmer Optima 4300 DV model) for sulphur (S) and plasma mass spectrometry with inductive coupling (ICP-MS, Perkin Elmer NexIon 300 X model) for the rest of the elements, at the CEREGE Sustainable Development Team laboratory (UM34 University of Aix Marseille), Technopole Environnement Arbois-Méditerranée. Twenty elements which include Na, Mg, P, K, Ca, Sr, Zr, Ba, Pb, Al, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Mo and S were measured. The detection limits and measurement uncertainty of metallic elements analysed by ICP-MS and ICP-OES are reported in Table S1.

Computation of the concentration of mineral dust

Mineral dust from the Sahara/Sahel region is mainly composed of oxides (such as SiO₂, Al₂O₃, FeO, Fe₂O₃, CaO) and carbonates (such as CaCO₃, MgCO₃) allows the determination of the mineral load brought by the Sahara (Denier van der Gon et al. 2010; European Commission 2011). Among

all other elements, oxygen is present as an oxide and is never directly measured in particle samples (Denier van der Gon et al. 2010).

There is a formula that calculates the concentration of mineral dust in their oxide forms directly, given in the equation below. According to Remoundaki et al. (2011), these oxides represent the mineral dust portion of PM_{2.5}:

$$\text{Mineral Dust oxide} = [\text{SiO}_2] + [\text{Al}_2\text{O}_3] + [\text{Fe}_2\text{O}_3] + [\text{K}_2\text{O}] \\ + [\text{CaO} + \text{CaCO}_3] + [\text{MgO}] + [\text{TiO}_2]$$

To estimate the calcium levels in its most abundant forms (CaO + CaCO₃), Ca was multiplied by a factor of 1.95 (Remoundaki et al. 2011).

The silicon concentration was not obtained by analysis because quartz filters are rich in silicon. In our study, silicon was estimated by multiplying the concentration of aluminium by a factor of 3 (Alastuey et al. 2005).

Data analysis by PMF

The PMF approach is a multivariate factor analysis technique that has been widely applied in numerous studies in recent years. The fundamental principle of PMF is to decompose the matrix of ambient data X comprising the measurements of concentration of n chemical species in m samples and their corresponding uncertainties, into two matrices, the matrix G which represents the contribution of each factor for each sample and the matrix F which represents the chemical composition of the profile of each factor. PMF calculates the site-specific source profiles with the temporal variations of these sources based on the correlations between the data as shown in equation:

$$x_{ij} = \sum_{k=1}^p g_{ik}f_{kj} + e_{ij} \quad (1)$$

where x_{ij} is the concentration of species j measured on sample i , g_{ik} is the relative contribution of any k factor to sample i , f_{kj} is the concentration of species j in the k factor profile, e_{ij} is the residue associated with the concentration of species j measured in sample i , and p is the number of independent sources.

The PMF model tries to reproduce x_{ij} by minimizing the sum of the squares of the scaled residuals (Q):

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{S_{ij}} \right)^2 \quad (2)$$

Thus, PMF identifies a set of p factors that best characterize PM_{2.5} on our site. The results are constrained so that no sample can have a negative contribution to the source.

The identification of the main sources of particulate pollution as well as the contribution of each source to the

levels of the PM_{2.5} is based on the protocol compiled by Belis et al. (2013), the recommendations of the EPA PMF v.5 guide (Norris et al. 2014) as well as the recommendations of Brown et al. (2015).

The number of factors (p) was determined using two criteria: the interpretability of the resulting PMF factor profiles and the factor matching success rate for the bootstrap (BS), displacement (DISP) trials and combination two (BS—DISP).

The following approach has been used consistently. The solution with the greatest number of physically significant factors is the one chosen. This choice is confirmed by visual inspection of any sharp drop in the Q_{true}/Q_{exp} graphs, relative to the number of adjusted factors. A detailed examination of the model results and goodness of fit was performed for the solutions of 2 to 9 factors.

Two input files must be specified, a concentration file and a file of uncertainties associated with these concentrations in order to be able to use PMF (Huang et al. 2001; Poirot et al. 2001).

As the PMF solution is highly dependent on the concentration and uncertainties, their estimation is crucial to obtain reliable results. Initially, all available species and samples (20 chemical elements and 120 samples) in the dataset are considered for the PM_{2.5} source apportionment, then data processing was performed to exclude species, samples or outliers.

To ensure a strong data signal, only samples with less than 10% data below zero or below the limit of detection (LD) are accepted; the uncertainty attributed to these values is given as follows:

$$\sigma_a^2 = LD_j^2 + (\alpha_{ij}X_{ij}) \quad (3)$$

where LD_j is the detection limit of element j , α_{ij} is the measurement uncertainty (%) of the concentration of element j in sample i and X_{ij} is the concentration of element X_j in sample i .

In addition, the analytical determination also includes the subtraction of the concentrations of the metallic elements of the blank filters which are filters different from those used for sampling. Blanks entail an additional source of uncertainty σ_{blk} which must be propagated together with σ_a to obtain the analytical determination uncertainty σ_A : $\sigma_A^2 = \sigma_a^2 + \sigma_{blk}^2$.

In order to include additional sources of uncertainty, the overall uncertainty was estimated by the following formula (Amato et al. 2009):

$$\sigma_{ij}^2 = \sqrt{\frac{\sigma_A^2}{V_i^2} + (\beta X_{ij})^2} \quad (4)$$

where V_i is the volume of air sampled for each sample i and β is a coefficient estimated at 0.15.

Species with a concentration below the detection limit have been replaced by $LD/2$ and the uncertainty S_{ij} of these values is calculated as follows:

$$\sigma_{ij} = 5/6LD \quad (5)$$

The data obtained is systematically filtered to detect unusual events and instrument performance, and is validated before modelling. The Grubbs test was also applied to the data set to detect maximum outliers. These values are removed from the data set and the test is repeated until no outlier is detected. An outlier may indicate an unusual event that disturbed the observed phenomenon to the point of making it incomparable to others. In such cases, the extreme value is either corrected where possible, or removed from the dataset. In our study, the outliers obtained during desert dust intrusions or other uncommon events were examined and retained and the uncertainties calculated for these values were increased by a factor of 3 (Kim et al. 2004, 2005).

The time series of species were also plotted and studied to identify any unusual outliers. Time series of uncertainties attributed to species concentrations were also plotted to detect any data anomalies, in which case the sample date is reconsidered to justify the extreme observation. If the anomaly is not justified, the actual value is taken as such in the modelling procedure. Each data set was also analysed to find the number of missing information, the number of validated data and the number of samples with a concentration below the LD value. Only species for which 50% of the data are above the mean detection limit were taken into account. Samples for which data are missing were removed from the PMF analysis. The final datasets contain 95 samples and 20 species.

Conditional probability calculation

The conditional probability function (CPF) was used to locate potential local sources based on local meteorological data (wind direction and speed) (Begum et al. 2004; Kim and Hopke 2004). CPF calculates the probability of a source that is in a particular wind direction sector, $\Delta\Theta$:

$$CPF = \frac{m_{\Delta\Theta}}{n_{\Delta\Theta}} \quad (6)$$

where $n_{\Delta\Theta}$ is the number of times the wind passed through the direction sector, and $m_{\Delta\Theta}$ is the number of times the source contribution peaked as the wind passed through the sector (Ashbaugh et al. 1985). It should be noted that the CPF is not suitable for distant sources because the air particles can be diverted from their path. A good knowledge of local sources is necessary.

To ensure statistical reliability, 24-h average source contribution data was applied to all 60-min wind direction

averages measured at the site for each date. The wind direction range has been divided into 24 sectors of 15° . All periods associated with wind speeds less than 1 m/s were removed from the dataset. CPF is useful for determining the direction of a source from a receiving site. However, it cannot determine the actual location of the source.

Similarly to CPF , $PSCF$ is a conditional probability function. In this case, regional potential sources are examined using backward trajectories. Potential source contribution function ($PSCF$) analysis has been widely used to identify regional sources that increase the level of pollutant concentration. $PSCF$ is calculated with the probability of concentration at each cell and is calculated using the following equation:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \quad (7)$$

where m_{ij} is the number of points whose concentration is greater than a threshold and n_{ij} is the number of endpoints of the return paths that pass through each cell of the grid (Karnae and John 2011). The back trajectories required for the analysis are generated using a model developed by the National Oceanic and Atmospheric Administration (NOAA), called a hybrid single-particle Lagrangian integrated trajectory model (HYSPLIT4). The level of pollutant concentration in each of the parameters is calculated. Using the geographic information system (GIS), a spatial grid of required resolution is generated and a 90th percentile threshold is established for the distributed mass of each source.

The emission source contributions from the PMF analysis were analysed using the conditional probability function (CPF) and the potential source contribution function ($PSCF$) to identify the geographic locations of likely sources. The simultaneous use of these two tools is an effective way to identify local and regional sources affecting the sampling site (Wimolwattanapun et al. 2011). The CPF was applied at the 75 percentile of contributions and the $PSCF$ at the 90 percentile (Pekney et al. 2006).

It should be noted that very few PMF studies have been conducted in Africa. The only source apportionment study conducted in Constantine is that of Bencharif-Madani et al. (2019), in which the authors used principal component analysis (PCA) to perform chemical speciation and identification of sources of PM_{10} .

Results and discussion

Characterization and temporal trend of $PM_{2.5}$ and metallic elements

The mean annual concentration of $PM_{2.5}$ at the Zouaghi sampling site was $54.07 \pm 28.81 \mu\text{g}/\text{m}^3$. It should be noted that the

relatively high standard deviation is due to a strong seasonal variation of $PM_{2.5}$ and the recording of high concentrations of $PM_{2.5}$ during unusual events. The maximum and minimum concentrations were 132.52 and $16.91 \mu\text{g}/\text{m}^3$, respectively.

Figure 2 illustrates the average monthly $PM_{2.5}$ concentrations observed between 3/15/2017 and 3/15/2018 at the sampling site. The highest averages were recorded during the dry period (June–July–August). The mean annual concentration of $PM_{2.5}$ obtained in this campaign is higher than those measured at the traffic sites of other Algerian cities, such as Algiers ($32.85 \mu\text{g}/\text{m}^3$) (Talbi et al. 2018), Algiers ($10.22 \mu\text{g}/\text{m}^3$) (Belarbi et al. 2020) and Tiaret ($33.57 \mu\text{g}/\text{m}^3$) (Khadidja et al. 2019), and comparable to other North African cities such as Kenitra ($51 \mu\text{g}/\text{m}^3$) (Zghaid et al. 2009), Kenitra ($50.73 \mu\text{g}/\text{m}^3$) (Tahri et al. 2013), Cairo, Egypt ($51 \mu\text{g}/\text{m}^3$) (Boman et al. 2013) and Giza, Egypt ($61.31 \mu\text{g}/\text{m}^3$) (Hassan and Khoder 2017). This value is much higher than the annual threshold recommended by the WHO of $10 \mu\text{g}/\text{m}^3$, the American annual limit (NAAQS) for $PM_{2.5}$ ($32 \mu\text{g}/\text{m}^3$) and exceeds by a factor of 2 the standard decreed by the European Commission ($25 \mu\text{g}/\text{m}^3$). However, this value is lower than the limit value set by Algerian regulations ($80 \mu\text{g}/\text{m}^3$). According to the study carried out by the Health Effects Institute and the Global Burden of Disease (GBD) project of the Institute for Health Metrics and Evaluation (IHME) in 2019, 90% of the world's population lives in an environment where annual average $PM_{2.5}$ concentrations exceed the WHO recommended threshold. Algeria is among the countries most exposed to fine particles in the world, with average annual concentrations between 35 and $45 \mu\text{g}/\text{m}^3$ in the north and exceeding $70 \mu\text{g}/\text{m}^3$ in the south. The areas most polluted in the world by fine particles are South and Southeast Asia,

North Africa, the Middle East and the countries of sub-Saharan Africa. The highest annual levels were recorded in Nepal ($100 \mu\text{g}/\text{m}^3$), Niger ($94 \mu\text{g}/\text{m}^3$), India ($91 \mu\text{g}/\text{m}^3$), Qatar ($91 \mu\text{g}/\text{m}^3$), Saudi Arabia ($88 \mu\text{g}/\text{m}^3$), Egypt ($87 \mu\text{g}/\text{m}^3$), Cameroon ($73 \mu\text{g}/\text{m}^3$) and Nigeria ($72 \mu\text{g}/\text{m}^3$) (Health Effects Institute 2019).

The WHO recommended daily limit ($25 \mu\text{g}/\text{m}^3$) was exceeded 83 times during the sampling period, representing 69.17% of all sampling days.

Elemental analysis of $PM_{2.5}$ revealed that Ca, Na and Al predominate, followed by the elements S, Fe, K, Ba and Mg (Table 1). The terrigenous species Al, Ca, Fe, K, Mg and Ti are important as they represent about 62% of the total mass of metallic elements and the mineral dust component represents 16.6% of the mass of $PM_{2.5}$. Similar results were also observed in Barcelona (Pérez et al. 2008; Querol et al. 2004a). The concentrations of trace metals are in decreasing order $\text{Pb} > \text{P} > \text{Mo} > \text{Zn} > \text{Zr} > \text{Cr} > \text{Sr} > \text{Mn} > \text{Ni} > \text{Cu} > \text{V}$ and are between 0.3 and $42 \text{ ng}/\text{m}^3$. The concentrations of Pb and Ni are not alarming. Their maximum values are $124 \text{ ng}/\text{m}^3$ and $14 \text{ ng}/\text{m}^3$, respectively.

The annual Pb concentration is well below the limit of $500 \text{ ng}/\text{m}^3$ recommended by the WHO and the limit set by the European Commission ($200 \text{ ng}/\text{m}^3$). This value is lower than those reported in $PM_{2.5}$ nearby traffic sites in Algiers ($371 \text{ ng}/\text{m}^3$, Talbi et al. 2018; $283 \text{ ng}/\text{m}^3$, Oucher et al. 2015), but it is close to that recorded in Barcelona ($37 \text{ ng}/\text{m}^3$) (Querol et al. 2008). In Algeria, the main source of Pb emissions is leaded gasoline commonly used by vehicles (Naidja et al. 2018; Bencharif-Madani et al. 2019). Pb is not a tracer element for the combustion of gasoline in other countries where its use is prohibited.

The measured annual average concentrations of Cr, Ni and Zn are $6 \text{ ng}/\text{m}^3$, $3 \text{ ng}/\text{m}^3$ and $16 \text{ ng}/\text{m}^3$, respectively. They are

Fig. 2 Mean monthly concentrations of $PM_{2.5}$

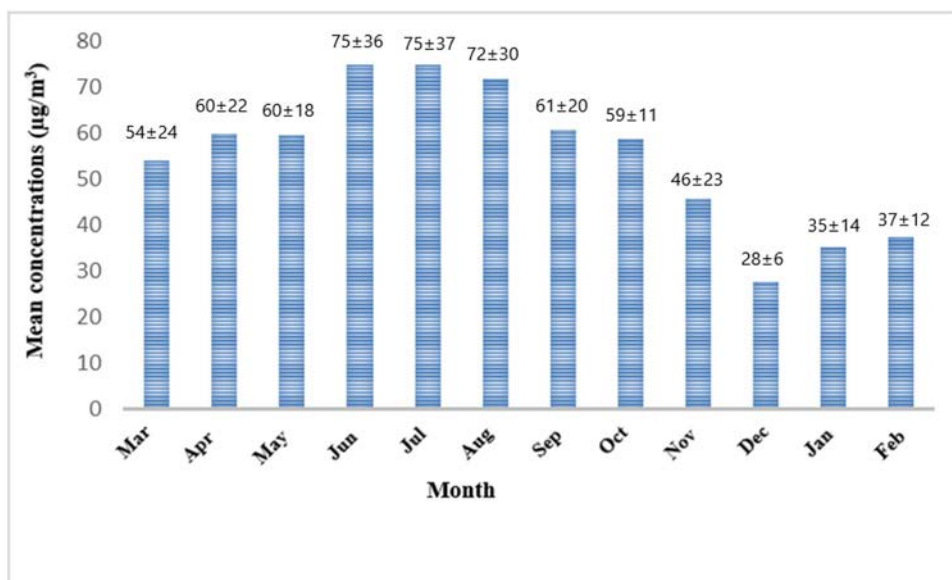


Table 1 Statistical data on the concentrations of PM_{2.5} (µg/m³) and their components (ng/m³)

| Species | Min | 25th | Mean | 75th | Max | Standard deviation |
|-------------------|------|------|------|------|------|--------------------|
| PM _{2.5} | 17 | 33 | 54 | 71 | 132 | 29 |
| Na | 10 | 352 | 769 | 987 | 2440 | 399 |
| Mg | 33 | 74 | 113 | 123 | 343 | 46 |
| P | 5 | 21 | 32 | 38 | 129 | 13 |
| K | 39 | 115 | 183 | 221 | 605 | 79 |
| Ca | 175 | 774 | 1249 | 1534 | 3771 | 496 |
| Sr | 2 | 4 | 6 | 7 | 14 | 2 |
| Zr | 7 | 10 | 14 | 17 | 27 | 4 |
| Ba | 11 | 65 | 120 | 139 | 442 | 54 |
| Pb | 0 | 30 | 46 | 58 | 123 | 19 |
| Al | 445 | 549 | 765 | 884 | 1767 | 220 |
| Ti | 7 | 13 | 20 | 22 | 62 | 8 |
| V | 0.14 | 0.3 | 0.6 | 0.7 | 2 | 0.3 |
| Cr | 0.01 | 4 | 6 | 7 | 28 | 2 |
| Mn | 2 | 4 | 5 | 6 | 49 | 2 |
| Fe | 57 | 122 | 158 | 218 | 5399 | 102 |
| Ni | 0.08 | 2 | 3 | 4 | 13 | 1 |
| Cu | 0.20 | 1 | 3 | 4 | 17 | 2 |
| Zn | 0 | 10 | 19 | 22 | 98 | 10 |
| Mo | 9 | 28 | 30 | 32 | 81 | 3 |
| S | 137 | 191 | 254 | 294 | 601 | 67 |

lower than those recorded in Algiers (46 ng/m³, 14 ng/m³, 78 ng/m³) (Talbi et al. 2018) and in Kenitra, Morocco (84 ng/m³, 166 ng/m³, 1413 ng/m³) (Zghaid et al. 2009). Spearman correlations between PM_{2.5}, mineral dust (MD), measured metals and meteorological parameters are presented in Fig. 3. This figure illustrates a strong correlation between mineral dust (MD) and terrigenous elements such as Al, Ca, Mg, Ti and K ($0.76 < r < 0.93$), except Fe ($r=0.36$). Fe is influenced by various anthropogenic sources (traffic and industry in particular). Kandler et al. (2007) analysed dust plumes from the Sahara and observed the predominance of the major elements Al, Si, Fe, Ti, Ca, S, Na and Mg to which are added some trace elements Sr, Zr, Ba, La, Th and Nd. In addition, a strong correlation between Fe and mineral dust was recorded in summer and in autumn ($r=0.91$ and 0.81 , respectively) (Figs. S3, S4) while in spring, this correlation is medium (0.51) (Fig. S5).

There is no significant correlation between Fe and mineral dust during winter, while they are fairly well correlated during the other seasons (Figs. S3-S6). This trend has been highlighted by Bencharif-Madani et al. (2019) who observed high concentrations of this element at the same site during spring, summer and autumn, a phenomenon which could be explained by the frequency of Saharan dust episodes in Algeria during these seasons. Fe has an excellent correlation with Mn ($r=0.97$). Mn is often an alloy with Fe to make steel. A probable source of emission of these two elements is the agricultural tractor company of Oued Hmimime, 10 km southeast from our sampling site.

Apart from Cr and Ni, which are fairly well correlated (59%), the trace elements Pb, Mo, Cu and Zn do not show a good correlation between them, with Ni and Cr and the major elements. This reflects the diversity of the sources of emission of these trace elements unlike the major elements which mainly come from the earth's crust. Figure 4 illustrates a similar time course for the major elements Na, Al, K and Mg indicating the existence of a common source. The major elements Ca, Fe, Ba and S do not follow the same pattern of temporal variation. This is due to the diversity of the sources of these major elements. Indeed, Fe, Ba and S are linked to traffic, industrial and natural emissions. V and Ti are, on the other hand, closely linked from the point of view of time evolution and also show a good correlation (76%). Several major elements (Na, Ca, Al, S, K, Mg, Ba) and traces (Sr, Ti, V, Cr) reach a peak during the period of July–August (Figs. 4, 5). As this season is known to be strongly impacted by desert dust (Lokorai et al. 2021) and temperature peaks favourable to the lifting of soil dust, it follows that these elements are essentially linked to sources of wind emissions.

Influence of meteorological conditions on PM_{2.5} concentrations

Conventional meteorological parameters such as temperature, wind speed, wind direction, relative humidity and precipitation level have a strong effect on PM concentrations (Tran and Mölders 2011; Kassomenos et al. 2014).

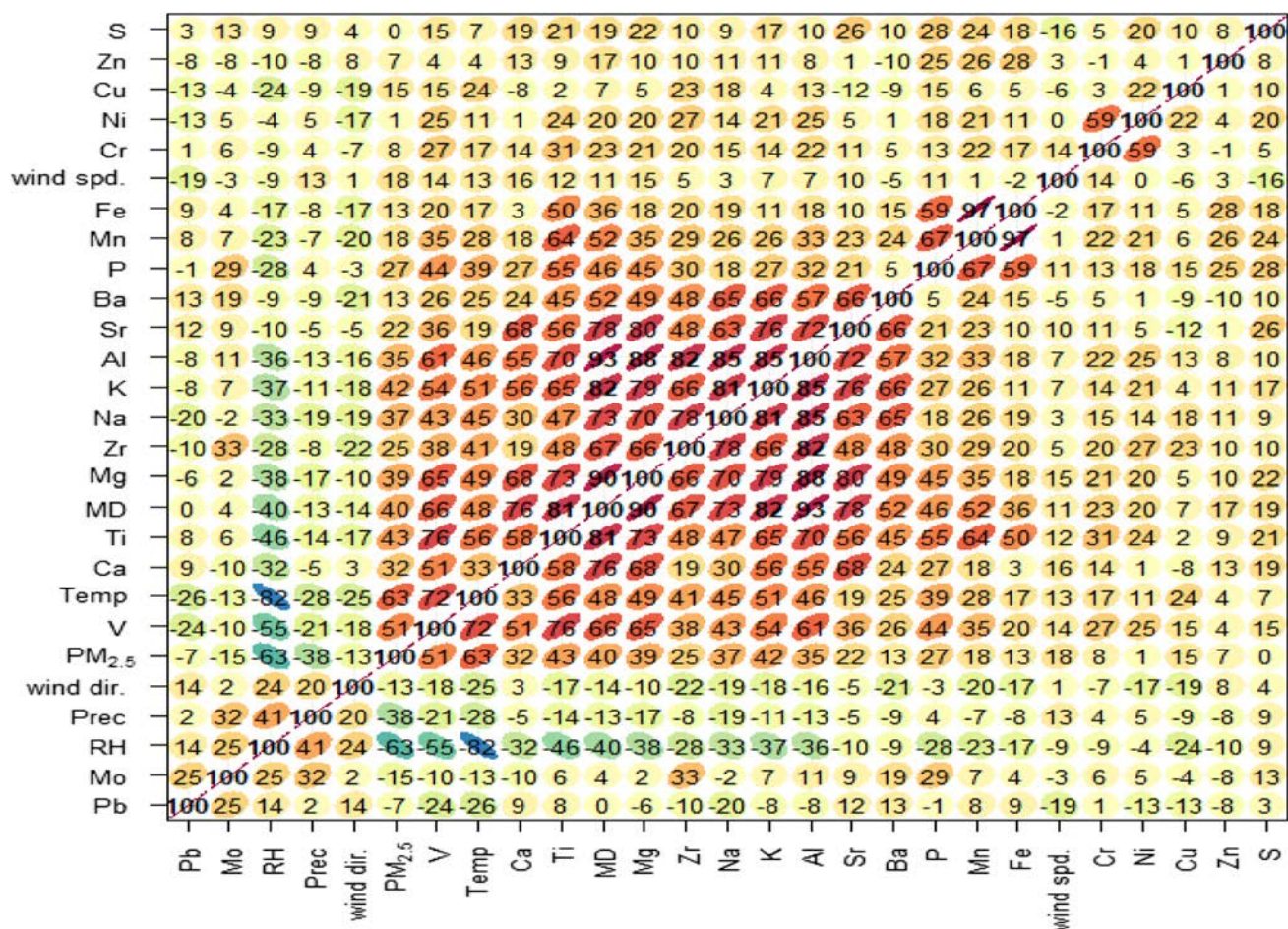


Fig. 3 Spearman correlation matrix showing the relationships between annual average concentrations of PM_{2.5} components and different meteorological parameters

The variations of these factors over the course of a year have an impact on the seasonality of the annual profiles. This trend was also observed in our study. The sampling period was characterized by a low level of precipitation as a total volume of 476.62 mm was recorded. The average daily value of relative humidity varies between 22.40 and 91.21% with an annual average of 60.18 ± 18.12 , the high values being recorded during winter. The wind rose specific to our site (Fig. S7) indicates that the prevailing winds are north-westerly (19.38%), north (18.95%) and west (15.61%), respectively. The mean annual wind speed was 3.96 m/s.

The figure of correlations between PM_{2.5} and its metal compounds and several meteorological variables indicates that relative humidity (RH) and precipitation (Prec) have a negative correlation with most pollutants during the entire study period (Fig. 3). This was predictable due to the leaching of the atmosphere by precipitation (Dawson et al. 2007; Feng and Wang 2012). In addition, humidity promotes the agglomeration of particles in the air and their wet or dry deposition (Anda et al. 2015; Kang et al. 2015) or

considerably reduces resuspension of dust by the wind (Hieu and Lee 2010). According to Chu (1997), a decrease in relative humidity (40–60%) led to higher PM concentrations. Positive correlations were found between RH and PM_{2.5} leading to an exacerbation of PM pollution until sufficient hygroscopic growth, aggregation and merging of small-sized particles led to PM deposition (Lou et al. 2017). The highest concentrations of PM_{2.5} were observed at elevated temperatures, thus facilitating the resuspension of soil dust by the wind.

Fig. S8 shows the combined effects of temperature, wind speed and relative humidity on PM_{2.5} concentrations. These results showed that, for an RH < 60%, the concentrations of PM_{2.5} tend to (i) increase appreciably with T and (ii) increase with wind speed (WS). For an RH > 60%, the concentrations of PM_{2.5} tend to (i) increase slightly with T but (ii) they are independent of the wind speed.

For temperatures below 15 °C, PM_{2.5} concentrations are independent of wind speed and relative humidity. Above

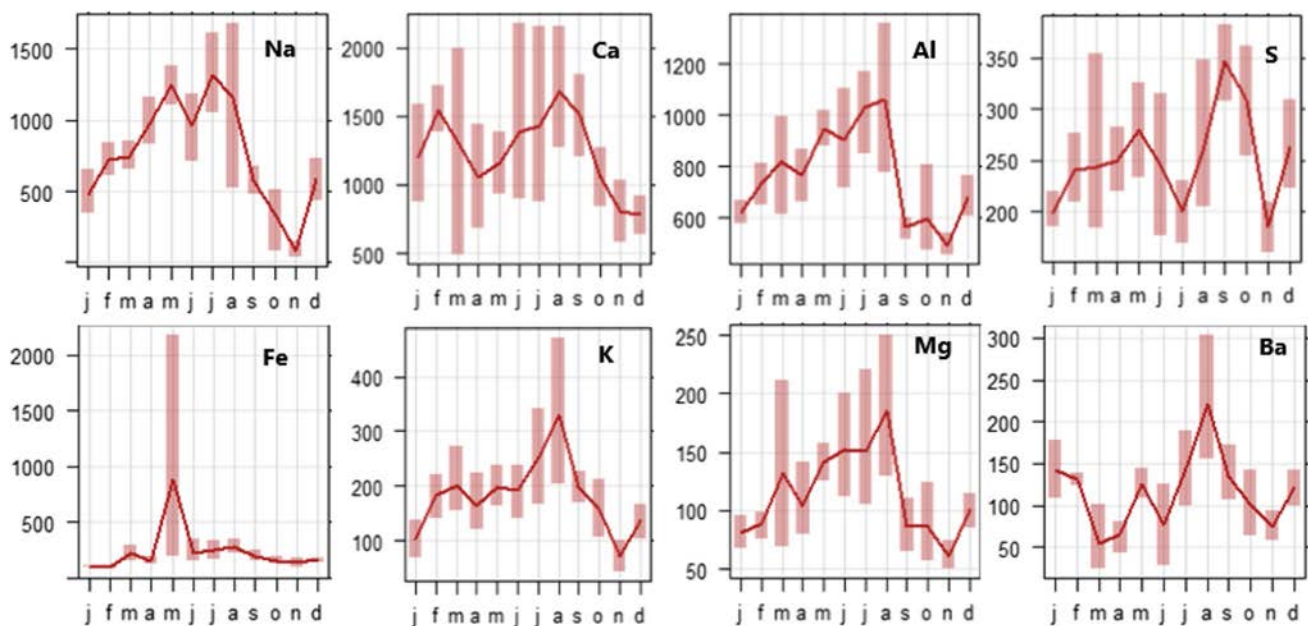


Fig. 4 Monthly average concentrations of major elements. The histogram represents the monthly minimum and maximum average values

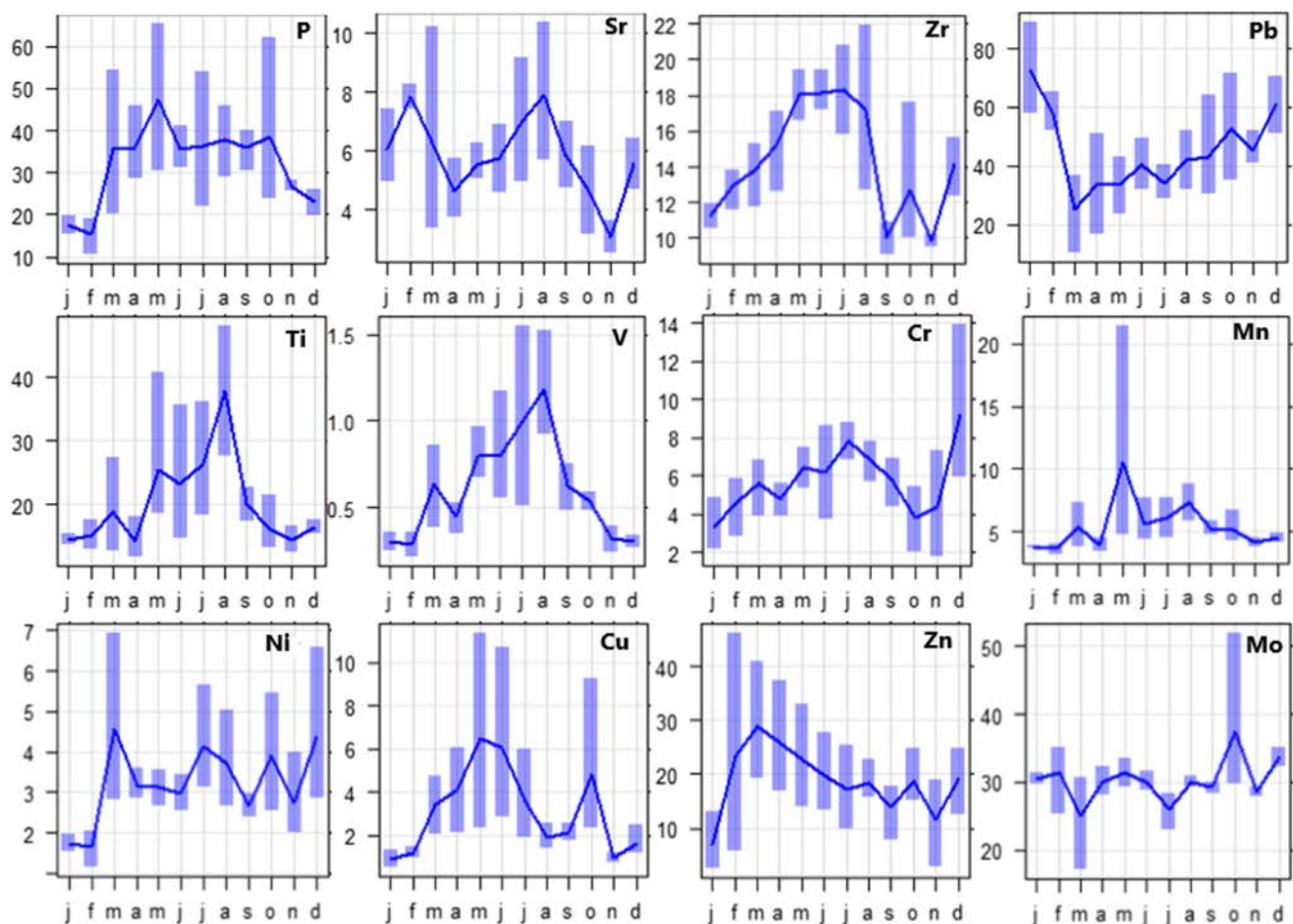


Fig. 5 Monthly average concentrations of trace elements. The histogram represents the monthly minimum and maximum average values

15 °C, $PM_{2.5}$ concentrations decrease with relative humidity but increase with wind speed.

In addition, the concentrations of $PM_{2.5}$ are not affected by the wind speed for a temperature < 15 °C and a relative humidity > 60%. In the latter case, relative humidity is linked to the agglomeration of particles, which weakens the influence of wind speed. This behaviour of $PM_{2.5}$ with respect to wind speed and relative humidity has been highlighted by Wang and Ogawa (2015) and Chen et al. (2018).

Identification of emission sources (profiles/contributions)

The high levels of particulate air pollution in Constantine are influenced by Saharan dust episodes (Ali-Khodja et al. 2008; Terrouche et al. 2016; Bencharif-Madani et al. 2019). The geographic proximity of North Africa to particularly active desert dust emission areas (Engelstaedter et al. 2006) such as the Libyan desert, Niger and Mali explains the high levels of particles in the air in this region (Safar and Labib 2010; Evan et al. 2015). In addition, the semi-arid climate, hot and dry in summer and cold and humid in winter where the frequency and intensity of precipitation is concentrated over a short period (on average 20 days per year), which characterizes the city of Constantine promotes the resuspension of dust and increases the concentrations of particles in the ambient air. It should be noted that activities related to agriculture are also an important source of dust and lead to soil erosion (Webb and Strong 2011; Ginoux et al. 2012; AVECILLA et al. 2015). Constantine is traditionally an agricultural town. Agricultural land represents 82% of the total area of the province, of which forests only occupy nearly 8%, or 17,858 hectares (ANDI 2013). These factors explain the significant presence of dust raised by the winds, especially in dry weather.

The choice of the optimal solution is based on the knowledge available a priori of the profile of the chemical composition of the sources and on the results of the error estimation methods. This subjectivity is part of the process of choosing the optimal solution (Ulbrich et al. 2009). The discussion above indicates that the five-factor solution is more stable and realistic than the six- and seven-factor solutions, due to the lower random errors and rotational ambiguity (Fig. S9). In addition, the scaled residuals (d) for all elements are in the range $-3 < d < +3$ and mostly obey a normal distribution, adding credibility to the chosen solution.

Five factors were shown to be physically significant (Fig. 6). The PMF model identified sea salts (15.1%), industrial activities (18.9%), non-exhaust emissions (24.2%), exhaust emissions (15.8%) and mineral dust (25.9%) as the main sources of metallic aerosols near road traffic in Constantine. Mineral dust and non-exhaust emissions were found to be the two main contributors to $PM_{2.5}$. The results also

show that 40% of the mass of $PM_{2.5}$ is attributable to vehicle emissions.

Sea salt

The species contributing to the first factor mainly include Na (86.7%), K (56%) and Mg (42.3%) (Fig. 7). This source is attributed to sea salts and contributes 15.1% to the concentration of $PM_{2.5}$. Indeed, the back trajectories of marine aerosols (85th percentiles) show that in many cases, the contribution of the Mediterranean Sea is undeniable (Fig. S10).

However, the contribution of the salt lakes in the region and the great chott of El Djerid which is crossed by south-eastern air masses cannot be overlooked (Troussset 1995). The importance of this source is explained by the proximity of the city of Constantine to the Mediterranean Sea (60 km). Na, K and Mg are commonly used as sea salts tracer elements (Saradhi et al. 2008; Gupta et al. 2012). Several authors have used the Mg/Na ratio (Buat-Menard et al. 1974; Savoie and Prospero 1980). Marine aerosols are generally characterized by a Mg/Na ratio close to 0.12 (Brewer et al. 1975). A ratio equal to 0.129 was recorded on the island of Lampedusa (Kishcha et al. 2011), while in Athens, Basel and Helsinki, this ratio was between 0.11 and 0.14 (Ilacqua et al. 2007). In our study, the Mg/Na ratio is 0.066. The enrichment of Na in this factor could be explained by the high Na content in soil dust as well as in aerosols from salt lakes and anthropogenic sources (Hoffman and Duce 1972). According to the latter authors, continental dust transported across the Pacific is the source of excess Na in sea salts. In Algeria, the “sabkha” salt lakes have high concentrations of Na (Demdoum et al. 2015; Lamini and Hacini 2018). Doumbia (2012) found a ratio equal to 0.33 and concluded that salt lakes are the main source of Na, Mg and Cl. Other studies

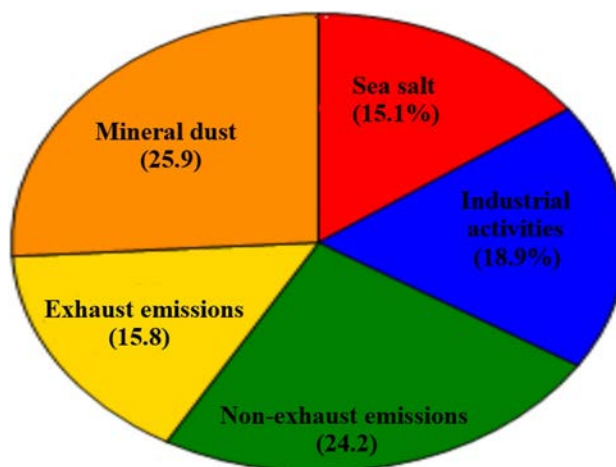


Fig. 6 Contributions of the five emission sources to $PM_{2.5}$

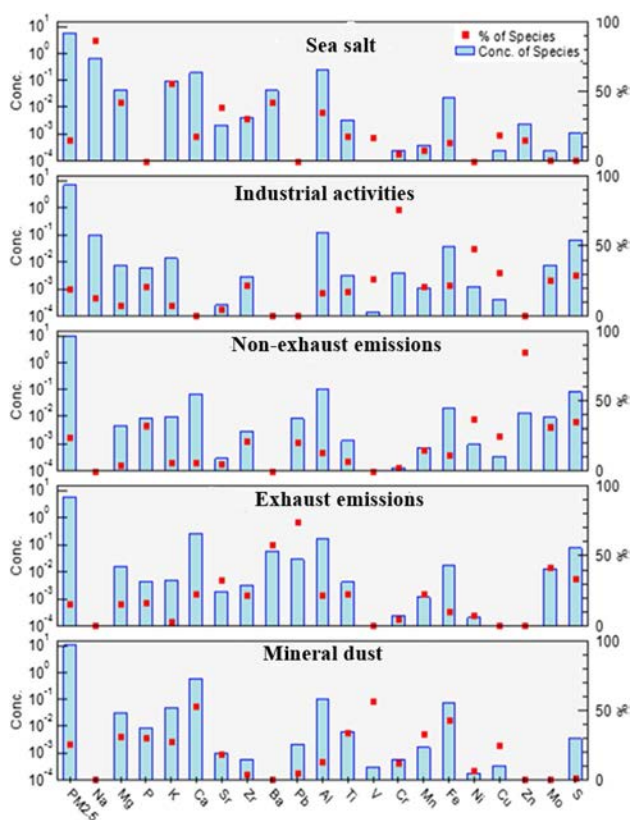


Fig. 7 PMF factor profiles for identified sources

have confirmed the association between high Na levels and salt lakes (Wen 1989; Cao et al. 2008).

The presence of elements such as Sr, Zr, Ca, Ba, Al and Ti in this factor could be linked to desert dust, which may be raised by air masses from the Mediterranean Sea. The Ca/Al ratio (0.77) calculated in this factor is in agreement with the reports of other studies (0.84 (Kong et al. 2011), 0.81 (Xu et al. 2004) and 0.76 (Shen et al. 2007)) which reflect the determining contribution of desert dust. Fig. S11 indicates that the Mediterranean Sea is the main source of this factor, the eastern contribution being more decisive than the northern contribution despite its remoteness from the sampling site. The contribution of the Sahara is not to be overlooked, either. The first factor can therefore, be considered as a mixture of sea salt and mineral dust.

The “sea salts” factor is more significant during the months of May, July and August as shown in Fig. S12. The scarcity of precipitation during the dry season facilitates wind erosion of soil and salt lakes.

Regarding the “sea salt” factor, its contribution tends to increase when $RH < 30\%$, $T > 20\text{ }^{\circ}\text{C}$ and $WS > 3\text{ m/s}$ (Fig. S13).

When RH decreases, sea salts tend to change from biphasic (saline) to crystals that can stay longer in the atmosphere and reach areas farther from the sea (Zezza and Macrì 1995;

Morcillo et al. 2000). The temperature increase coincides with the warm seasons (summer and spring) when the recorded precipitation is lower. As a result, leaching of the atmosphere is less frequent and leads to a higher concentration of sea salts in the air (Zezza and Macrì 1995; Gong et al. 1997; Feliu et al. 1999). Furthermore, the wind speed favours the production of marine aerosols and this effect is observed at wind speeds greater than 3 m/s (McKay et al. 1994; Gustafsson and Franzén 1996; Morcillo et al. 2000). Moreover, according to these authors, sea salt concentrations increase exponentially with wind speed above 3 m/s.

Industrial activities

The second factor is attributed to industrial emissions, which are characterized by high contributions of Cr (76.1%), Ni (48.3%), Cu (31.2%), S (29.3%), V (26.4%), Mo (25.9%), Fe (22.1%) and Mn (21.3%) (Fig. 7). Industrial emissions are generally heterogeneous and differ depending on the manufacturing process and the type of fuel used (Singh et al. 2017). In contrast, the choice of metal tracers specific to an industrial source is often difficult (Banerjee et al. 2015). S, Ni, Mo and V are linked to anthropogenic activities such as combustion and industrial processes (Marcazzan et al. 2003). Ni and V are natural constituents of petroleum and are found in all of its by-products (Turunen et al. 1995; Vale et al. 2004).

Ni and V are widely associated with fuel oil combustion and the refining industry (Celis et al. 2004; Pey et al. 2009). The V/Ni ratio in this factor is 0.11 and cannot be representative of diesel emissions with a characteristic factor > 4 (Moreno et al. 2010). A ratio of < 2 reflects the presence of Ni enriched by emissions from the surface treatment industry located west-southwest of our sampling site (Bencharif-Madani et al. 2019). The Cr and Ni pollution rose indicates a similar concentration profile indicating that these two elements have a common emission source (Fig. S14). Ten kilometres from our site and in this direction is a large mechanical industrial pole within the Ain Smara industrial area.

In general, Ni, Cr, Fe, Mn, Zn, Fe, S, Cu and Mo are used as tracers of industrial emissions (Song et al. 2006; Tauler et al. 2009; Sharma et al. 2014). Metals like Cr, Fe, Mn, Mo and Ni are associated with the metallurgical and steel industries (Lodhi et al. 2009; Mansha et al. 2012). The production of iron and steel is an important source of particles in the ambient air around industrial areas (Kumar et al. 2001; Mazzei et al. 2006). Studies have reported that the particles emitted by the steel smelter in the region of Cornigliano, Italy, represent 60% of the mass of PM_{10} (Prati et al. 2000) and 45% of the mass of PM_{10} in Port Talbot, UK (Taiwo et al. 2014). Cr, Ni and Cu are generally used in the

metallurgical industry and in foundries as a highly corrosion-resistant element in the steel plating process (Wadden et al. 1991; Kulshrestha et al. 2009). Fe, Cr, Ni and Cu are probably from construction sites (Kulshrestha et al. 2009) in Didouche Mourad and El Khroub city (Fig. S15). They can also be emitted by the cement and glass industries as well as brickworks in the industrial zone of Didouche Mourad (Tian et al. 2012; Hasheminassab et al. 2014).

Mo is used as an additive in lubricants (Zainal et al. 2018) and it may be released into the environment by the combustion of fossil fuels used in industry, explaining its significant presence in this factor (Salminen et al. 2005). S in the form of SO₂ is a pollutant emitted by power plants, petroleum refineries as well as industrial activities such as steel processing (Salminen et al. 2005; Shahbazi et al. 2016). Finally, Fe and Mn are tracers in the steel industry (Owoade et al. 2015).

Summer and spring have the highest concentrations of this factor. The impact of an industrial zone is closely linked to the frequency of the winds that pass through it. Indeed, the wind rose (Fig. S16) indicates that N, N-NE and SE winds are more frequent during summer compared to fall and winter. The industrial zones of Didouche Mourad, Skikda and El Khroub located in these respective directions are believed to be responsible for this marked increase in this factor in summer. In the spring, NN-E and SE winds are more frequent than in the fall and winter. The contribution of the Skikda industrial zones and El Khroub would be decisive. Moreover, Fig. S17 perfectly illustrates the nuanced increase in this factor between summer and spring.

The factor “industrial activities” does not seem to be affected by the meteorological parameters. No particular trend is observed with the variation of these parameters (Fig. S18).

Wind direction and emission intensity are the determining factors for particulate matter concentrations from industrial areas. Fig. S19 shows the sources of the emissions contributing to this factor. These sources are located in the industrial zones of El Khroub (south-west), Didouche Mourad (north), Palma (north-west) and construction sites in Ali Mendjeli (south-west), in Didouche Mourad (north-north-east), the El Hadaba plateau (north-west) and the quarry of the National Aggregates Company (south-east).

The contribution of industrial activities to PM_{2.5} in the study site (18.9%) is much less than that reported by Tahri et al. (2013) (52%) in Kenitra City, Morocco, who asserts that a significant number of small industrial facilities involved in metal works surround the sampling site located close to a bus station.

Non-exhaust emissions

The third factor includes elements resulting from the wear of brakes, tires and road surfaces as well as the resuspension

of soil particles. Querol et al. (2004b) showed that in European cities, non-exhaust sources account for half of all traffic emissions. Harrison et al. (2001) also estimate that particles resuspended by vehicles represent a fraction equivalent to that of exhaust emissions. Rehn et al. (2014) also found that the contribution of non-exhaust emissions was equivalent to that of exhaust emissions. In some northern European countries, non-exhaust particulate concentrations in PM₁₀ can represent up to 90% of airborne particles near the road (Forsberg et al. 2005; Omstedt et al. 2005). The contribution of this source at our sampling site is estimated at 24.2% of the total mass of PM_{2.5} (Fig. 6) and represents more than 60% of the total emissions related to road traffic. This factor is mainly characterized by high loads of Zn (84.6%), Ni (37%), S (34.5%), P (32.5%), Cu (22.5%), Mo (31.8%), Zr (21.2%) and Pb (20.8%) (Fig. 7). The main sources of non-exhaust emissions are brake and tire wear and pavement abrasion (Thorpe and Harrison 2008; Pant et al. 2017). Several studies have indicated that Fe, Cu, Pb and Zn are basic tracers and reliable markers of brake wear (Hildemann et al. 1991; Legret and Pagotto 1999; Westerlund and Johansson 2002). Hulskotte et al. (2014) reported that Cu, Zn, Fe and Sn represent 80 to 90% of the metals present in brake pads. Furthermore, Hjortenkrans et al. (2007) reported that brake dust contains significant amounts of Cu, Sb, Ba, Al, Si, S, Ti, Zn, Ni, Cr and Pb. Kennedy and Gadd (2003) reported that concentrations of Cu are 11 mg/kg (25% by mass). Other studies have revealed that Cu is the most abundant species and represents 11.8% (Westerlund and Johansson 2002), 14.2% (Legret and Pagotto 1999) and 2.4% of the total mass of brake linings (von Uexküll et al. 2005). A wide variety of components is commonly used in brake linings (Chan and Stachowiak 2004). This contributes to the difficulty of identifying the chemical composition of the resulting particles. The presence of Zr as zirconium oxide can be attributed to brake wear (Eriksson et al. 1999) or road wear (Huzita and Kasama 1983).

Zn is considered, according to several studies, to be the main indicator of tire wear in traffic areas or urban areas (Adamiec et al. 2016). Tires contain between 0.4 and 4.3% Zn (Smolders and Degryse 2002; Ozaki et al. 2004). These high contents result from the addition of ZnO and ZnS to the tire during vulcanization (Heideman et al. 2004, 2005). Zn, Cd, Co, Cr, Cu, Hg, Mo, Ni and Pb are associated with tire wear (Adachi and Tainosho 2004; Hjortenkrans et al. 2007). Amato et al. (2011) showed the predominance of Zn, Sb, Cu, S, Ni and Cr in the factor representative of tire wear. Similar results have been reported in source allocation studies using PMF (Bukowiecki et al. 2010; Doumbia 2012). On the other hand, the absence of other metals characteristic of tire wear and, consequently, of elementary ratios makes it difficult to use it as a tire tracer when it emanates from other anthropogenic or natural sources. This factor also contains

terigenous species such as Ca, Al, Ti, Fe and Mn as well as other trace elements such as Ni, Mo, S, Zr and P related to pavement wear (Lindgren 1996; Arditoglou and Samara 2005; Amato et al. 2011). Fe and Mn are generally associated with the mechanical wear of engine parts and various seals (Hildemann et al. 1991).

According to Fig. S20, the main sources of the “non-exhaust” factor are the entrance to the faculty, the parking, the traffic roundabout in front of the entrance to the faculty and the pedestrian crossing. Indeed, emissions increase significantly during braking and deceleration, especially at the traffic roundabout.

Fig. S21 illustrates the seasonal effect of the “non-exhaust emissions” factor on the temporal evolution of the contribution. The latter is minimum in winter during the months of January and February and maximum in spring during the months of March to June. In summer, a decrease in this contribution is observed, because it corresponds to a notable drop in traffic, which coincides with the holiday period. In winter, the effect of precipitation on particles generated by the wear of various components is evident.

The contribution of emissions to the “non-exhaust emissions” factor increases when $RH < 50\%$ and the temperature is below $15\text{ }^{\circ}\text{C}$ (Fig. S22). This is because low temperatures reduce the elasticity of the pavement surface, exacerbating friction on the harder inner layer. In addition, low temperatures tend to harden the tires thereby increasing the frictional force (Gustafsson et al. 2008). The effect of relative humidity is such that its decrease leads to favourable soil conditions for the resuspension of particles produced by various wear processes (Jeong et al. 2019). However, wind speed does not seem to have any effect (Fig. S22).

Exhaust emissions

The fourth factor is dominated by the presence of Pb, Ba, Mo, S, Sr and Mn, which contribute 73.9%, 57.6%, 41.5%, 33.9%, 32.4% and 22.9%, respectively, reflecting exhaust emissions. In India, vehicle emissions have been identified by Pb, Ni, Cu, Zn and Mn (Deshmukh et al. 2013; Sharma et al. 2016). Ba, Cd, Pb, Sb and Zn have been used as tracers of vehicular emissions in Pakistan (Lodhi et al. 2009; Mansha et al. 2012). In Sri Lanka, Seneviratne et al. (2011) identified K, Pb, Fe and S as tracers of vehicular emissions.

Pb, Ba and Mn are used as fuel additives to improve engine performance (Karar et al. 2006; Belis et al. 2013). The combustion of leaded gasoline remains the main source of lead emissions in the world (Pacyna and Pacyna 2001). It is an element that is often used as a marker of automotive emissions (Sabin et al. 2006; Zhu et al. 2018). Algeria is one of the few countries still using leaded gasoline (Naidja et al. 2018). Ba is considered to be a potential tracer of emissions from gasoline and LPG engines (Lin et al. 2005; Cheng et al.

2010). It is also used as a corrosion inhibitor in engine lubricating oils (Bem et al. 2003).

The profile of this factor also contains a substantial proportion of terrigenous elements such as Ca (23%), Ti (22.5%), Al (22%), P (16%) and Mg (15.4%) revealing the significant contribution of dust resuspended by road traffic. In our study, the Ca/Al ratio = 1.56 is close to those announced by Ho et al. (2003) (1.19) and by Kong et al. (2011) (1.29) which they attribute to dust from paved roads.

Fig. S23 shows the probability of the different directions contributing to the “exhaust emissions” factor at the local scale. The main sources are the road along the faculty as well as the roundabout in front of its entrance.

Winter experiences a drastic increase in concentrations of the “exhaust emissions” factor resolved by PMF (Fig. S24). Road traffic is usually intense during this season (Bencharif-Madani et al. 2019). This factor, associated with the reduction in the height of the atmospheric boundary layer and the stability of the lower atmospheric layers, increases remarkably during winter. In addition, the wind rose during winter is characterized by prevailing westerly winds where the main source of emissions contributing to the “exhaust emissions” factor is found, namely, the national road N79.

As for the “exhaust emissions” factor, its contribution increases when $RH > 60\%$ and $T < 10\text{ }^{\circ}\text{C}$ (Fig. S25). At low temperatures, meteorological conditions are more stable than in summer (Taghvaei et al. 2018). This results in a poor dispersion, which is at the origin of high concentrations of primary and secondary particles emitted directly into the atmosphere. The mixing of the hot vapours in the exhaust with the cold air considerably favours the nucleation phenomena for the atmospheric aerosols (Nilsson and Kulmala 1998; Charron and Harrison 2003). It is believed that nucleation increases the total mass of particles (Kulmala et al. 2001). Low temperatures associated with high relative humidity favour the formation of new particles (Easter and Peters 1994). In addition, high relative humidity promotes binary nucleation of sulphuric acid and water (Easter and Peters 1994; Hinds 1999; Seinfeld and Pandis 2016). Jambriška et al. (2008) suggest that the significant influence of RH on exhaust emissions may be related to particle agglomeration and changes in the hygroscopic properties of primary particles emitted by traffic which become hydrophilic under high-rate humidity conditions.

Mineral dust

The fifth factor represents 25.9% of the total mass of $\text{PM}_{2.5}$ and is dominated by V (56.7%), Ca (52.6%), Fe (43%), Ti (34.2%), Mn (32.9%), Mg (30.8%), P (30.3%) and K (27.7%) which are all terrigenous elements (Fig. 7). Therefore, this factor could be associated with the resuspension of soil dust. According to Lough et al. (2005), these dusts contain most

of the terrigenous elements Fe, Ca, Na, Mg, Al and K at high concentrations. Apart from K, these are the same terrigenous elements which are considered as tracers of soil dust: Al, Si, Ca, Mg, Fe and Na by Begum et al. (2006). A ratio of x/Al (where x is any element) could be considered a good criterion to distinguish the different sources of dust (Wang et al. 2006; Shen et al. 2016). The elementary ratios calculated in this study are more or less close to the ratios shown in Table 2.

The Ca/Al ratio of our study site is relatively higher compared to those of other sites. This is explained by the influence of anthropogenic emissions (Athanasopoulou et al. 2010). According to Fig. S26, we can assume that the construction sites near the sampling site (Ali Mendjeli, El Khroub), the quarries near El Khroub in the south-east and Ain Smara in the south-west and the area industrial plant of Didouche Mourad in the north (cement plant, brickyard, quarries) are the nearby sources that contribute the most to this factor.

On a regional scale, Fig. S27 reveals that the eastern region of Constantine contributes strongly to the “mineral dust” factor. This region is located in North Africa, between eastern Tunisia and eastern Algeria, and is considered a particularly intense and persistent source of dust (Prospero et al. 2002).

In fact, the prevailing synoptic winds are from the north-west (41.5%), east (39.4%) and west (19.1%) (Fig. S28).

The temporal evolution of mineral dust contributions perfectly illustrates the influence of the hot season on this factor. The period from June to September is the most favourable for the lifting of soil dust (Fig. S29).

The contribution to the “mineral dust” factor is favoured by $RH < 35\%$ and $T > 15\text{ }^\circ\text{C}$ and a wind speed $> 3\text{ m/s}$ (Fig. S30). The influence of relative humidity on dust levels has been highlighted by several authors due to its impact on the moisture content of the soil surface, and therefore on the cohesion of soil particles (Ravi et al. 2004; McKenna Neuman and Sanderson 2008). According to Csavina et al. (2014). Conversely, when RH is relatively low, soil particles are looser and more easily dispersed by the wind.

Moreover, wind speed is a main factor in the resuspension of soil dust (Yin et al. 2007). Temperature is also strongly correlated with dust concentrations (Hussein et al. 2006). The contribution of this factor increases in the hot season

due to the absence of leaching of the atmosphere which leads to the accumulation of dust in the air. Associated with strong winds, dry soil generates more dust in hot weather (Kassomenos et al. 2014; Li et al. 2015).

The influence of Saharan dust episodes, which are generally more frequent in summer, has been highlighted by several authors (Matassoni et al. 2009; Kopanakis et al. 2018; Querol et al. 2019).

Conclusion

The PMF model identified natural sources related to sea salt and mineral dust (41%) and vehicular emissions (exhaust and non-exhaust) (40%) as the main contributors to $PM_{2.5}$. Industrial emissions account for less than half of the contribution of each of these sources. The Mediterranean Sea is the main source of sea salt. The contribution of the Sahara is not to be overlooked, either. Emissions from natural sources are exacerbated during the hot season. The decrease of relative humidity and the increase of temperature favour sea salt particles and mineral dust resuspension. During winter, exhaust emissions are highest while non-exhaust emissions are lowest. These two components of vehicular emissions are affected in opposite ways by relative humidity and temperature. None of these parameters has an effect on industrial emissions which are mainly determined by wind intensity and direction. Moreover, the metallic tracers of different sources have been identified. These are Na for sea salt, Cr for industrial activities, Zn for non-exhaust emissions, Pb for exhaust emissions and Ca for mineral dust. It should be noted that Pb originates mainly from gasoline before its ban in August 2021. Ba and Mo could alternatively serve as metallic tracers for such exhaust emissions.

In order to be able to discriminate between influential sources, it would be wise to analyse a larger number of metallic elements and if possible other pollutants such as cations and anions. The analysis of organic tracers could be considered and integrated into future studies to better determine the influence of combustion sources.

A sufficiently large number of data can also allow separate studies according to the seasons. From a broader perspective, it appears very desirable to continue measurements

Table 2 Comparison between elementary ratios

| | K/Al | Ca/Al | Ti/Al | Mn/Al | Fe/Al | Pb/Al | References |
|------------------|------|-------|-------|-------|-------|-------|--------------------|
| Constantine | 0.48 | 5.92 | 0.06 | 0.02 | 0.75 | 0.021 | This study |
| Hong Kong | 0.40 | 1.93 | 0.05 | 0.03 | 1.54 | 0.030 | Ho et al. (2003) |
| North-west China | 0.73 | 3.73 | 0.12 | 0.04 | 1.35 | 0.008 | Shen et al. (2016) |
| North-east China | 0.41 | 2.65 | 0.09 | 0.03 | 0.95 | 0.004 | Shen et al. (2016) |
| North China | 0.47 | 3.07 | 0.10 | 0.03 | 1.14 | 0.006 | Shen et al. (2016) |

of the chemical composition of PM over the long term with, if necessary, a higher temporal resolution.

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Data availability All data generated or analysed during this study are included in this published article (and its supplementary information files).

Declarations

Ethics approval and consent to participate This study did not require ethics approval. Informed consent was obtained from all individual participants included in the study.

Consent for publication Consent to publish has been received from all participants.

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Source apportionment of PM_{2.5} and their associated metallic elements by positive matrix factorization at traffic site in Constantine, Algeria

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Table S1

Detection limits and relative standard deviations of major and trace elements analysed by ICP-OES and ICP-MS

| Element | Detection limit ($\mu\text{g/L}$) | Measurement uncertainty (%) |
|---------|-------------------------------------|-----------------------------|
| Na | 10 | 1.25 |
| Mg | 0.05 | 1.25 |
| P | 20 | 5.98 |
| K | 10 | 1.15 |
| Ca | 10 | 0.97 |
| Sr | 0.01 | 1.17 |
| Zr | 0.01 | 0.93 |
| Ba | 0.05 | 1.56 |
| Pb | 0.01 | 1.33 |
| Al | 0.02 | 11.15 |
| Ti | 0.05 | 8.02 |
| V | 0.01 | 6.60 |
| Cr | 0.01 | 3.96 |
| Mn | 0.02 | 6.24 |
| Fe | 0.1 | 3.58 |
| Ni | 0.01 | 1.79 |
| Cu | 0.05 | 1.65 |
| Zn | 0.1 | 3.02 |
| Mo | 0.05 | 1.66 |
| S | 150 | 4.52 |

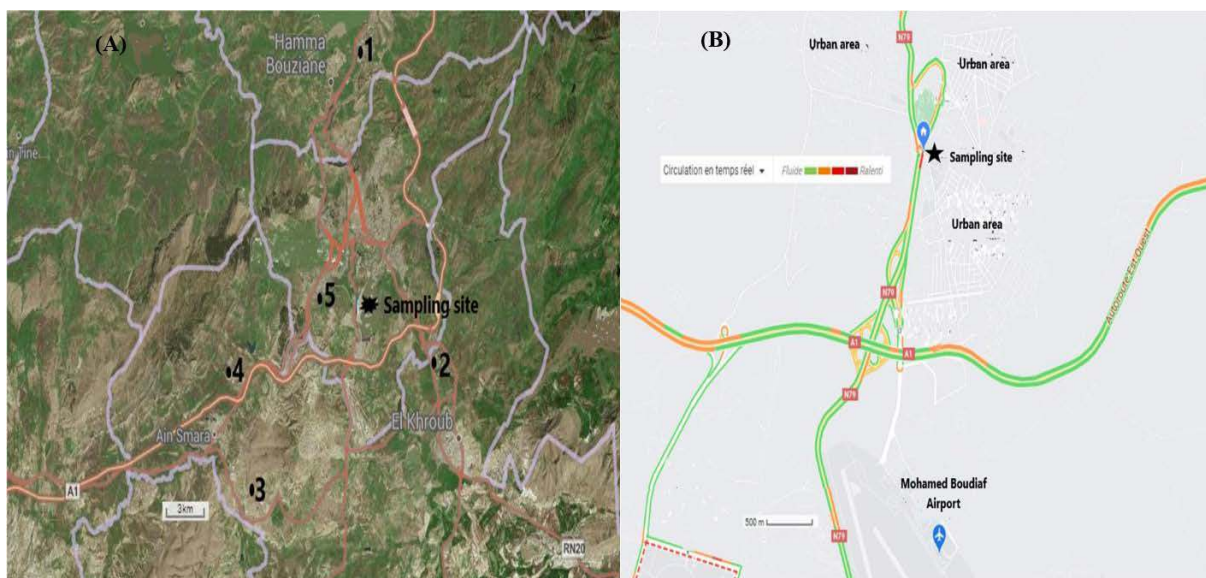


Fig. S1. Sampling site with (A) industrial emission sources and (B) nearby roads

- (1) Hammam Bouziane cement plant, (2) mechanical manufacturing plant (Oued Hmeimim), (3) stone production quarries, (4) mechanical complex (Ain Smara), (5) industrial activity zone (Palma).



Fig. S2. Sampler (LVS) used for sampling aerosols at the Zouaghi site

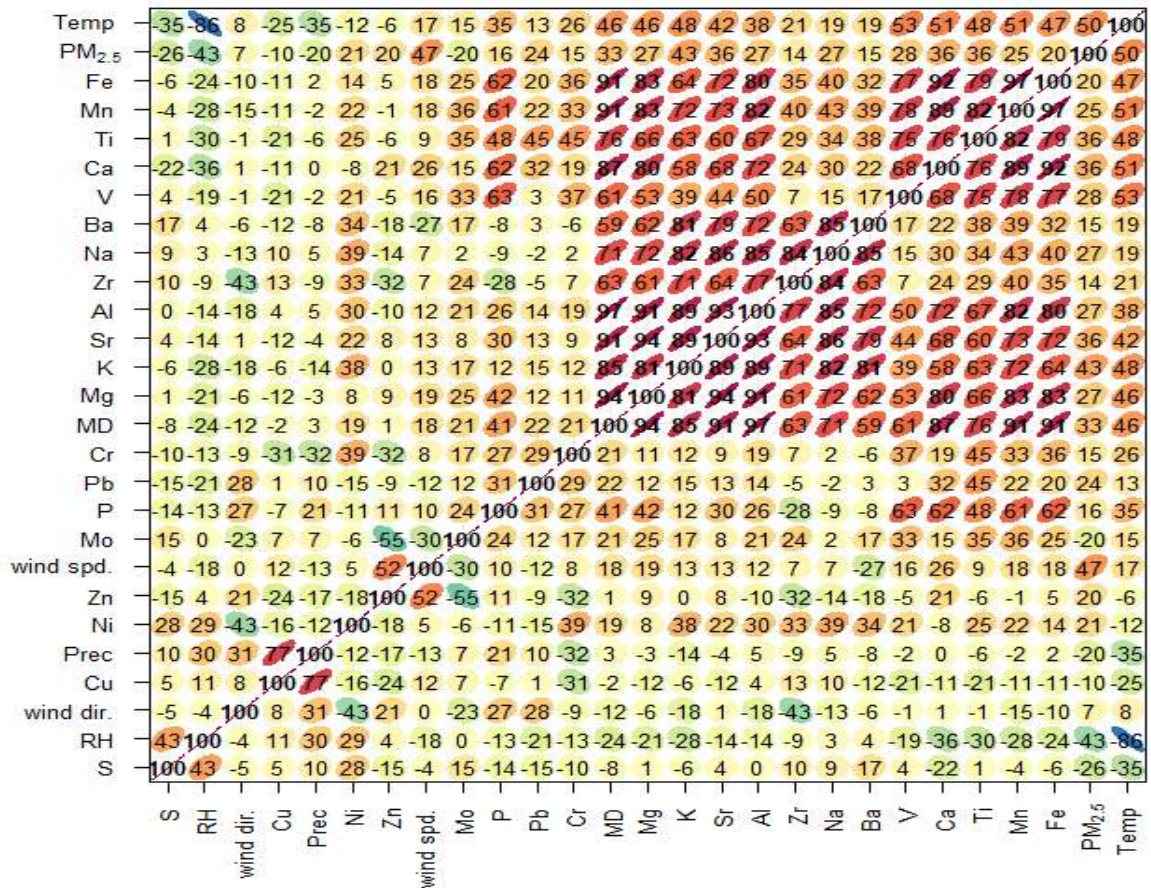


Fig. S3. Spearman correlation matrix showing the relationships between the average seasonal concentrations of PM_{2.5} components and the different meteorological parameters during the summer

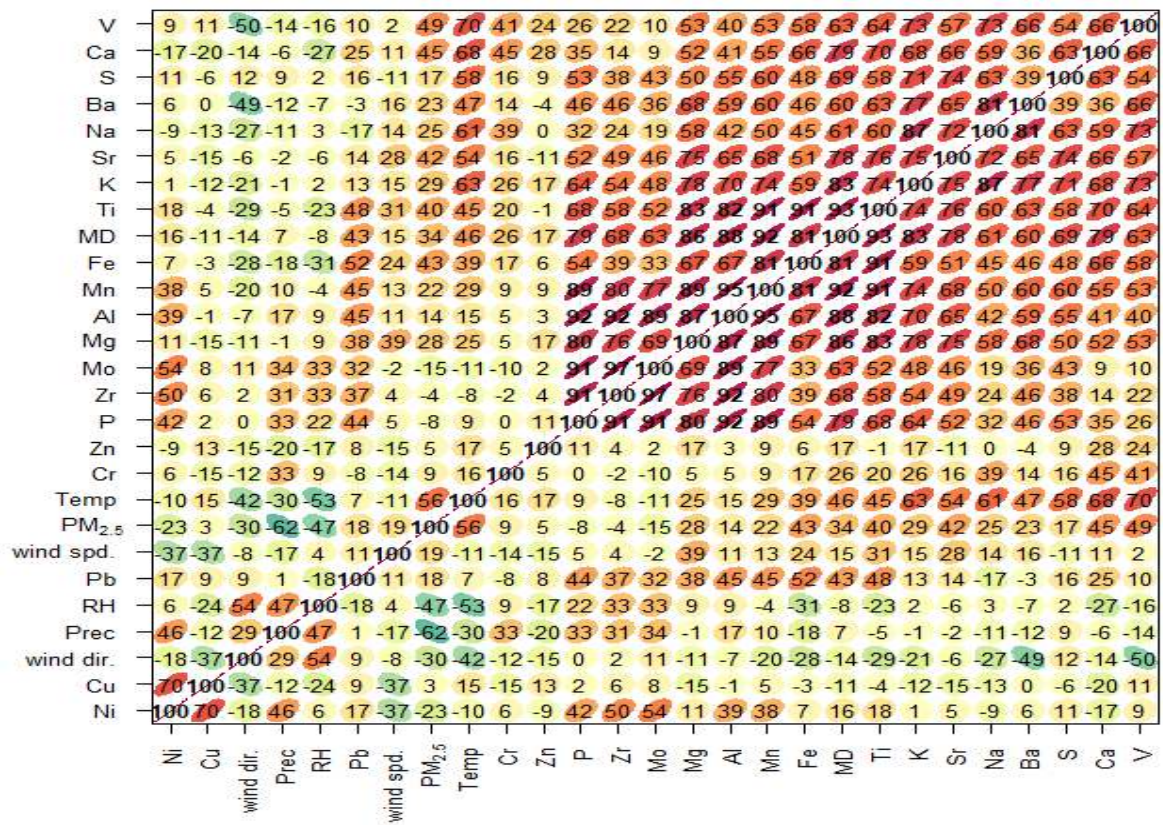


Fig. S4. Spearman correlation matrix showing the relationships between the average seasonal concentrations of PM_{2.5} components and the various meteorological parameters during the autumn

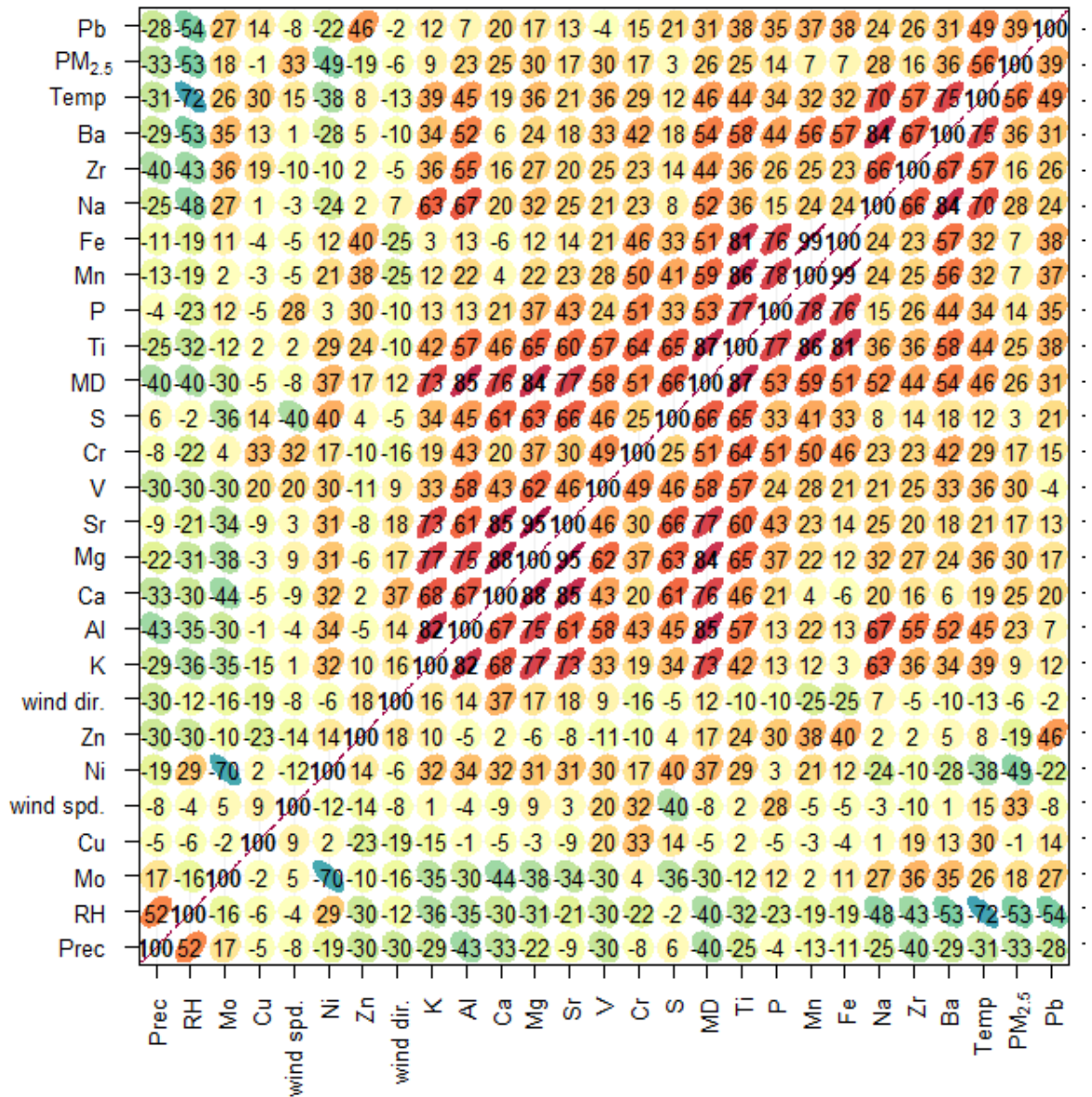


Fig. S5. Spearman correlation matrix showing the relationships between the average seasonal concentrations of PM_{2.5} components and the various meteorological parameters during the spring

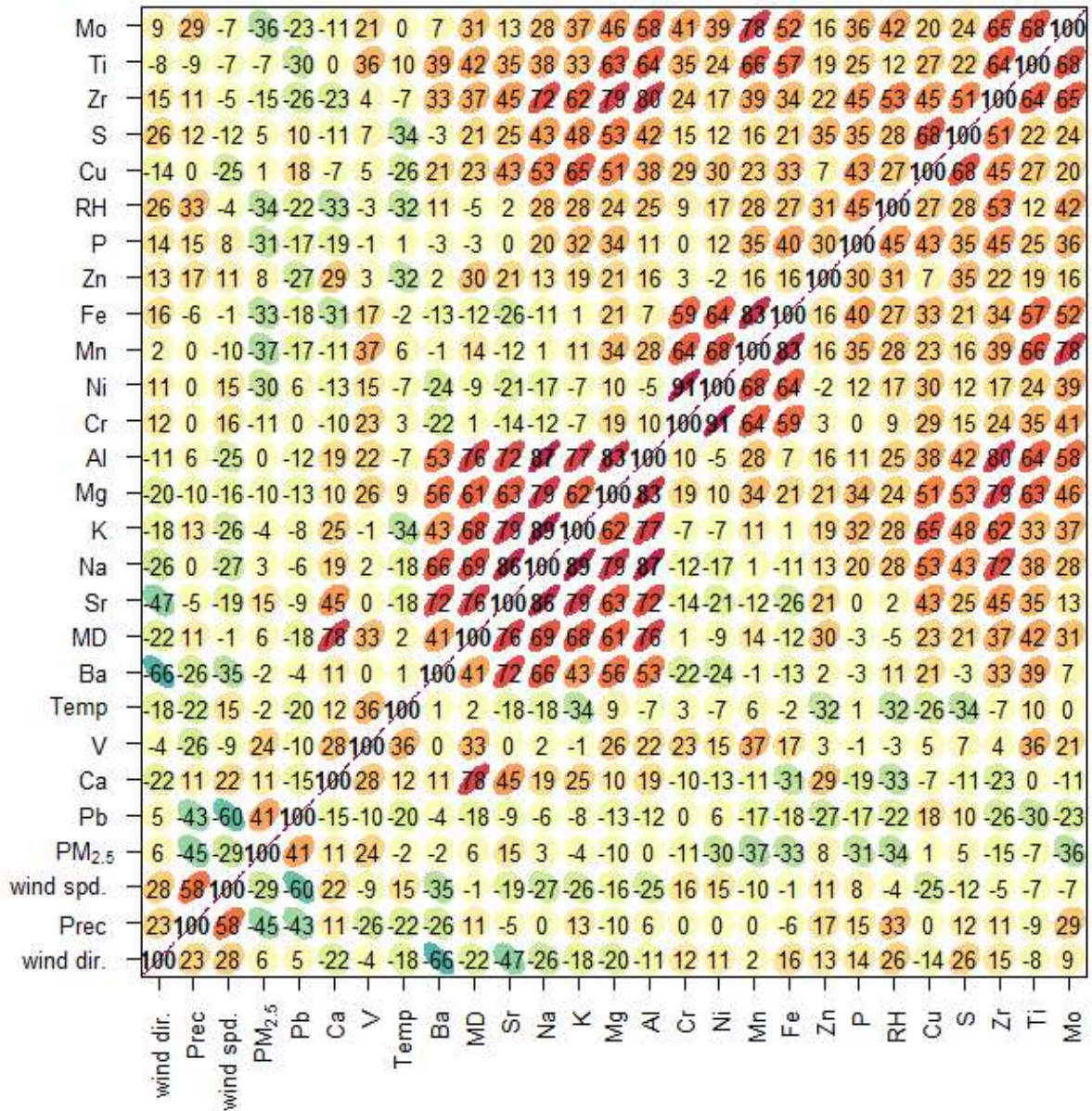


Fig. S6. Spearman correlation matrix showing the relationships between the average seasonal concentrations of PM_{2.5} components and the various meteorological parameters during winter

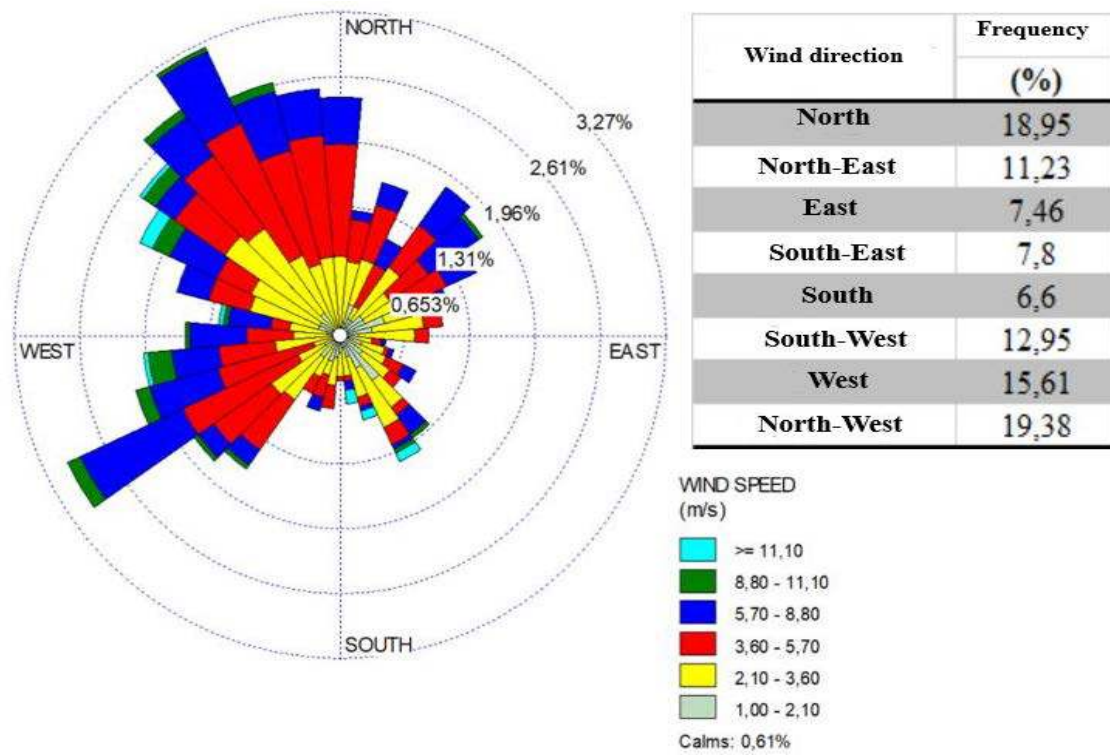


Fig. S7. Wind rose for the Zouaghi site for the period from 03/15/2017 to 3/15/2018

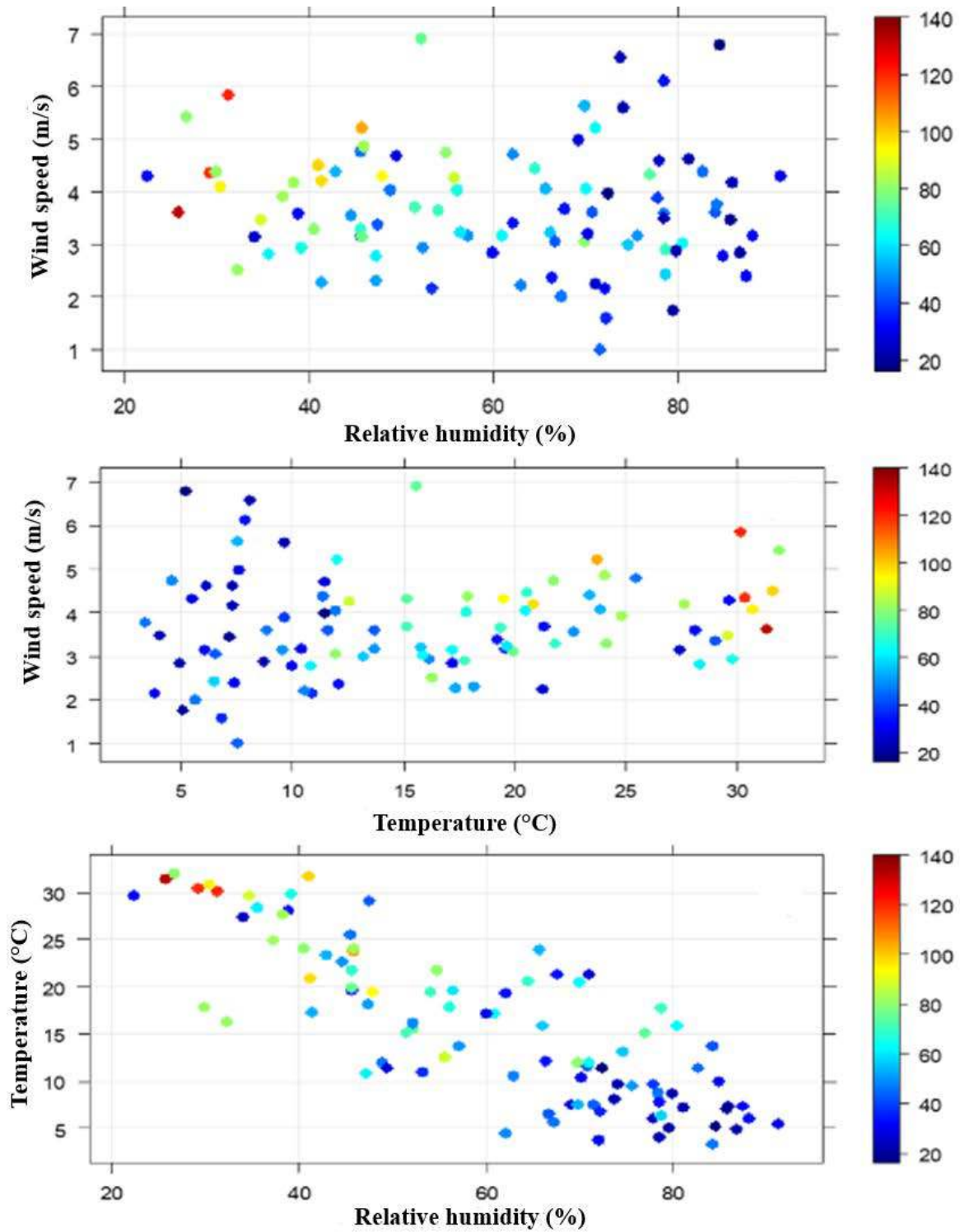


Fig. S8. Scatterplot of daily PM_{2.5} concentrations versus relative humidity (%), temperature (°C) and wind speed (m/s)

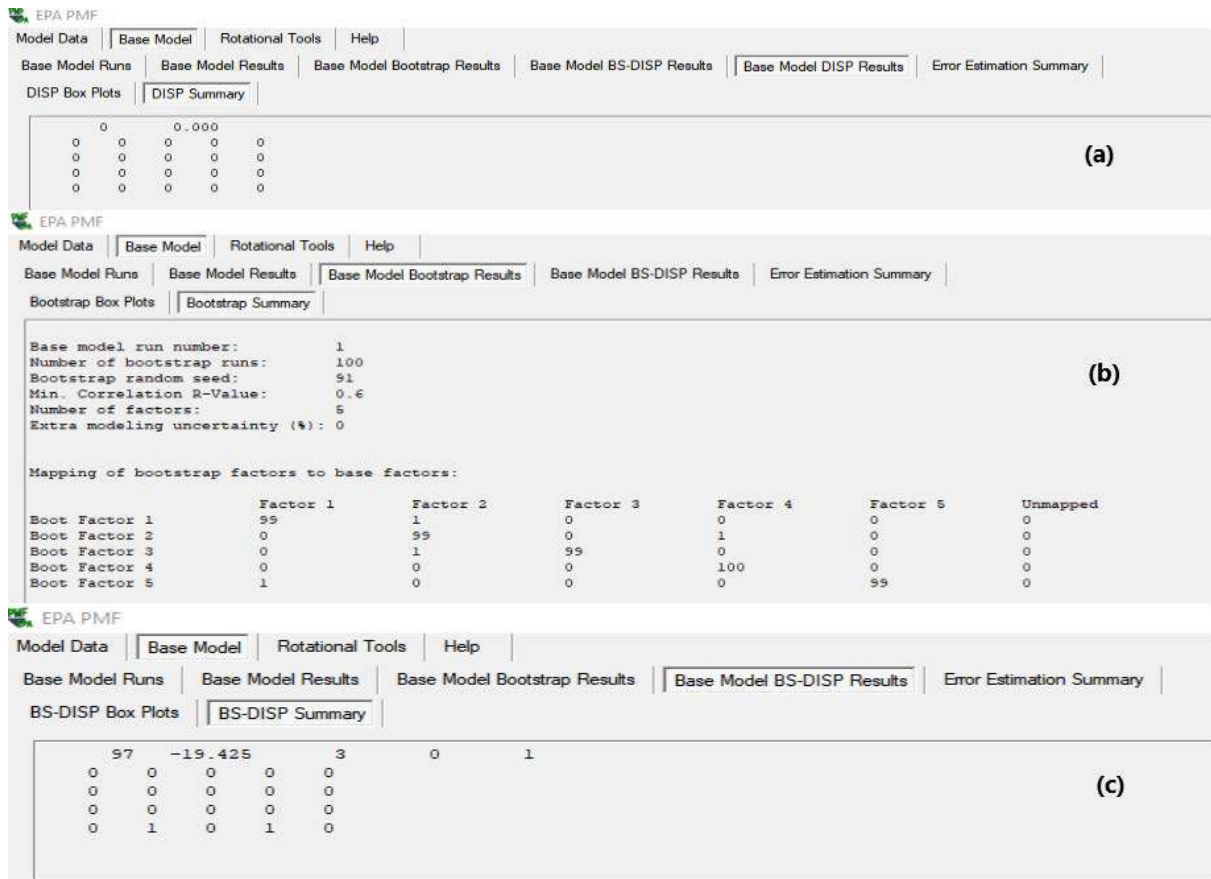


Fig. S9. The results of error estimation (a) DISP, (b) BS, and (c) BS DISP for the five-factor solution

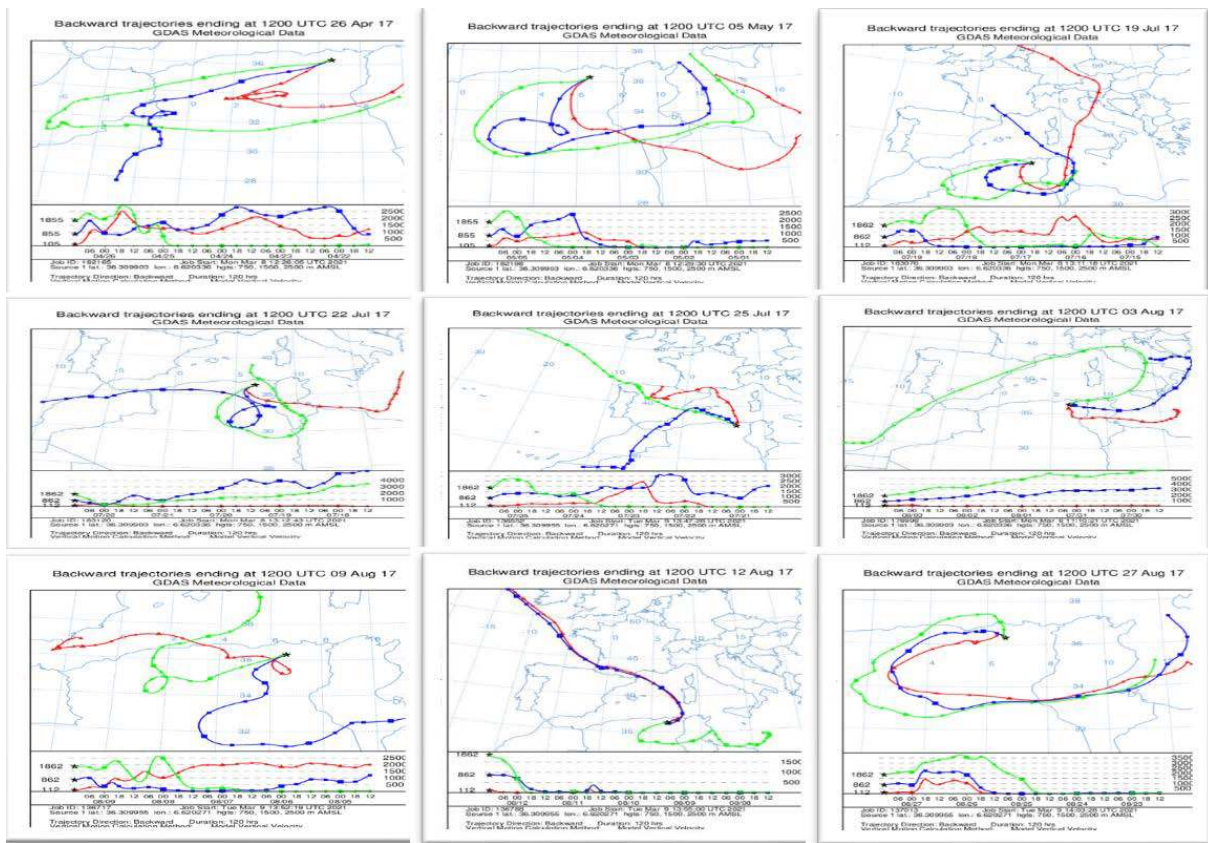


Fig. S10. 85th percentiles of sea salt back trajectories developed by HYSPLIT at altitudes of 750, 1500 and 2500 m above sea level for 5 days

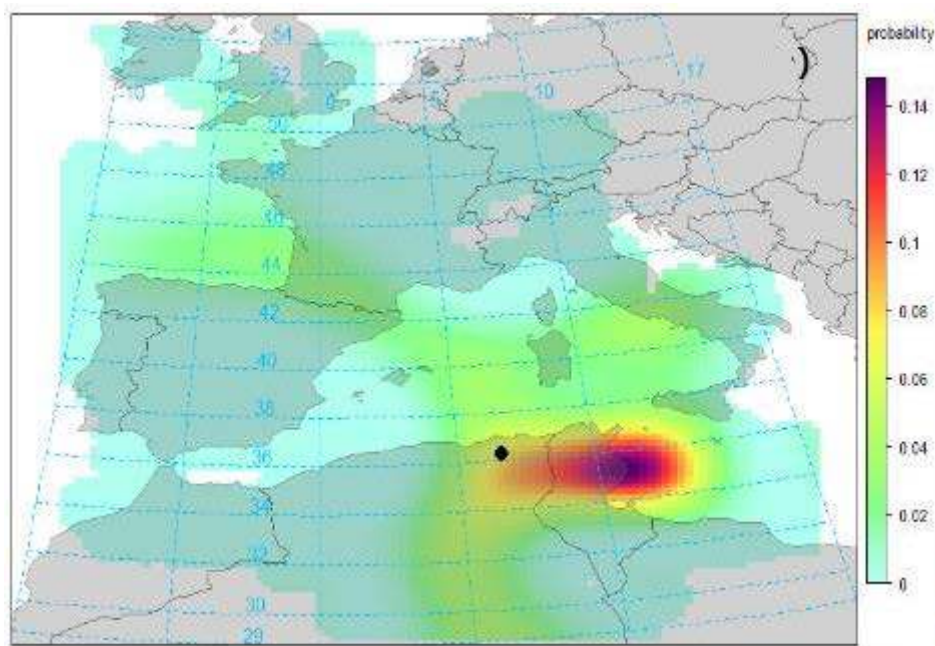


Fig. S11. PSCF probabilities (percentile 90) for sea salts contributions

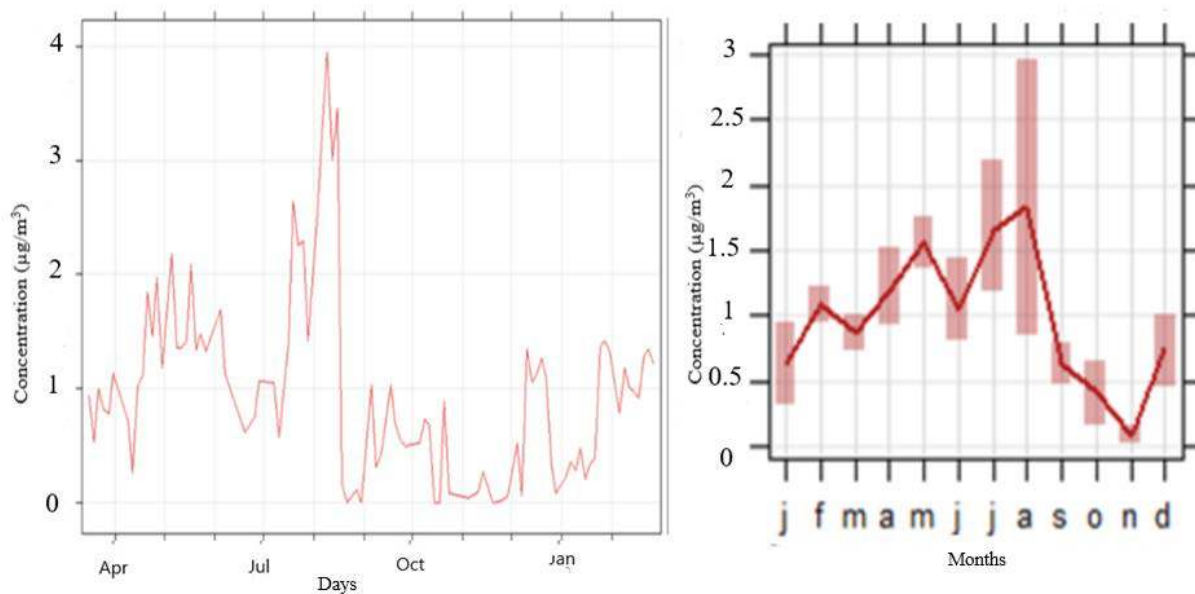


Fig. S12. Daily and monthly concentrations of the "sea salts" factor resolved by PMF

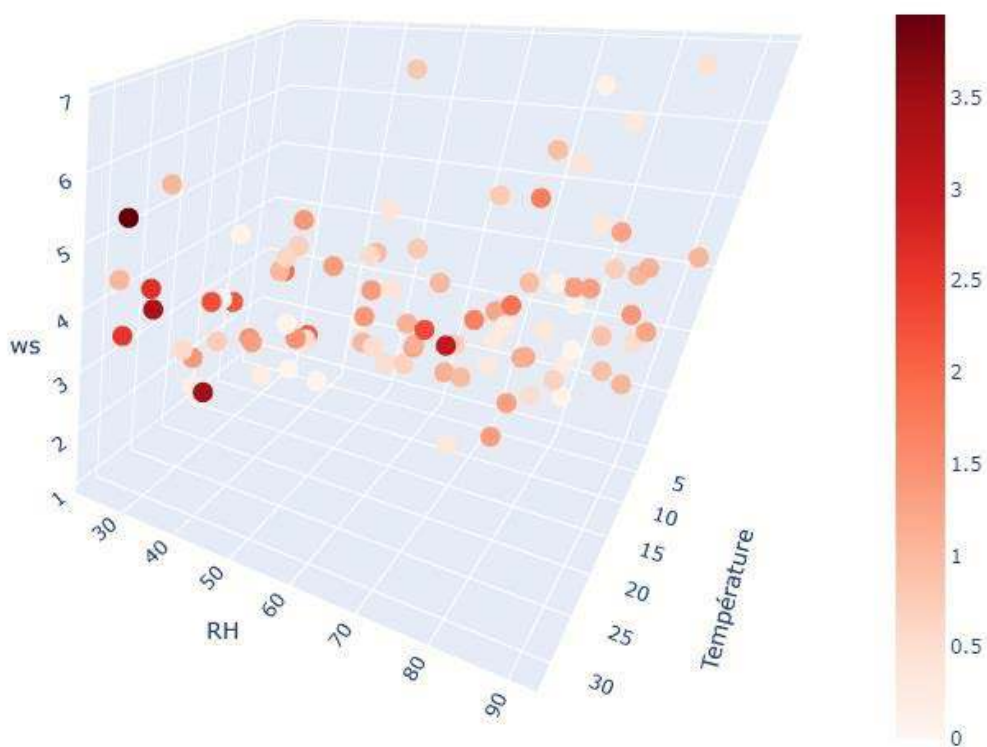


Fig. S13. Contributions ($\mu\text{g}/\text{m}^3$) of "sea salts" as a function of wind speed (m/s), relative humidity (%) and temperature ($^{\circ}\text{C}$)

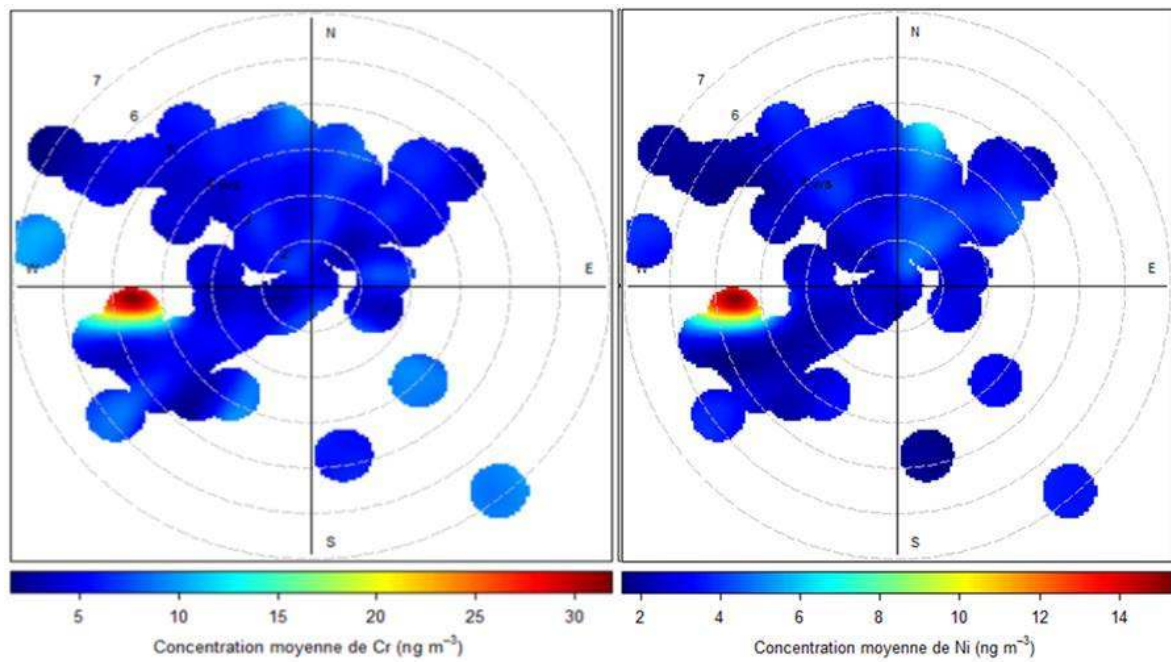


Fig. S14. Spatial distribution of Cr and Ni concentrations as a function of wind direction and speed

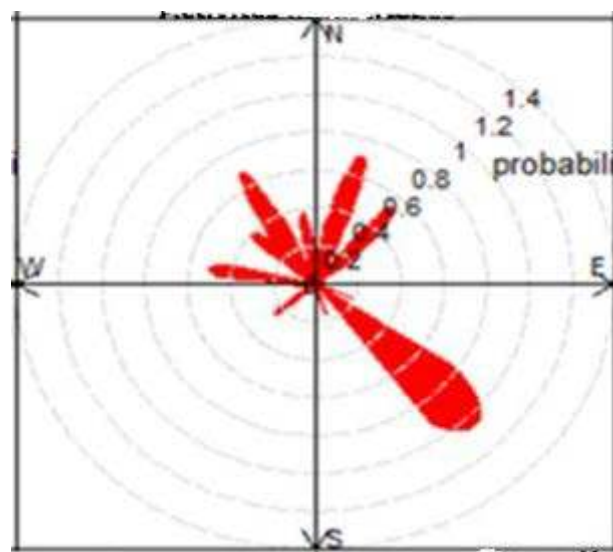


Fig. S15. 75th percentile of the conditional probability function (CPF) for the “industrial emissions” factor contributing to $\text{PM}_{2.5}$

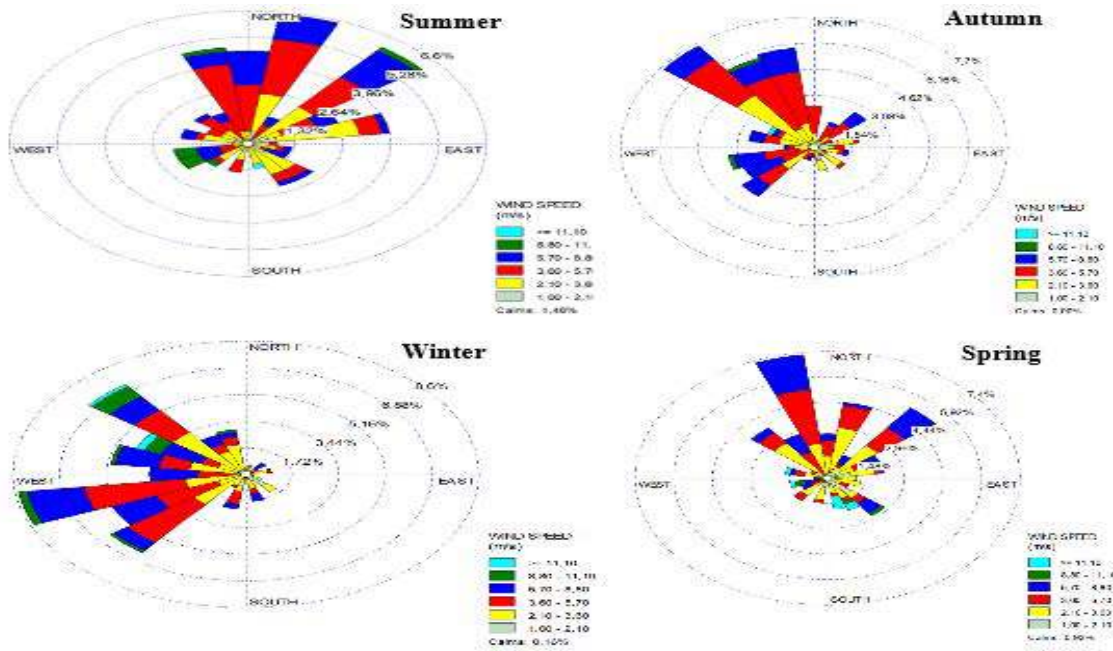


Fig. S16. Seasonal wind roses for the Zouaghi site

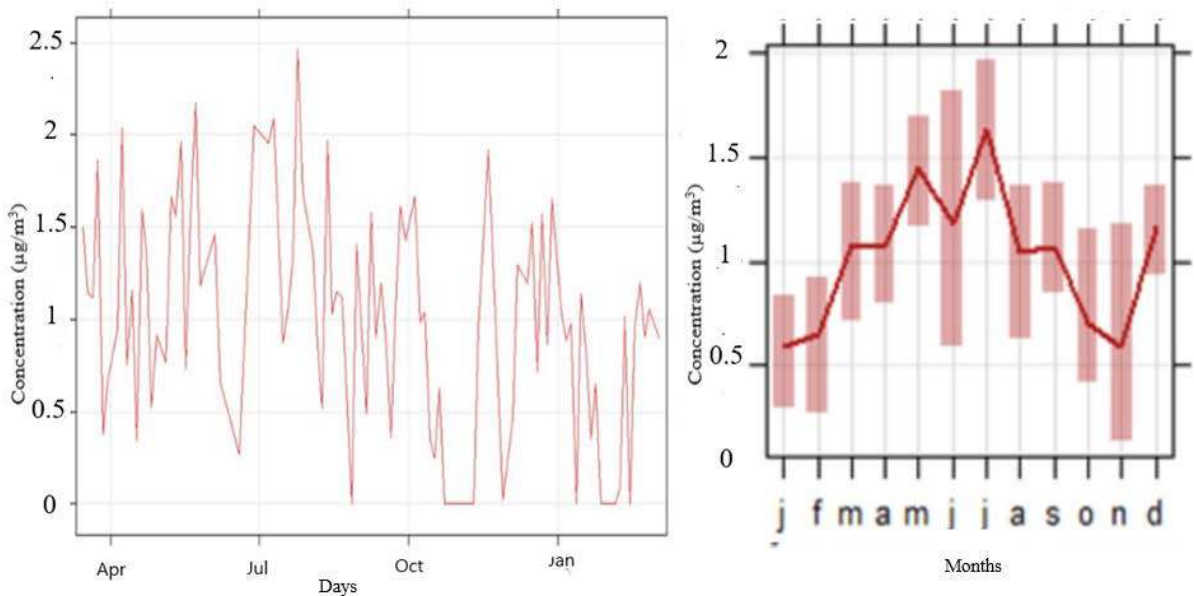


Fig. S17. Daily and monthly contribution of the "industrial activities" factor resolved by PMF

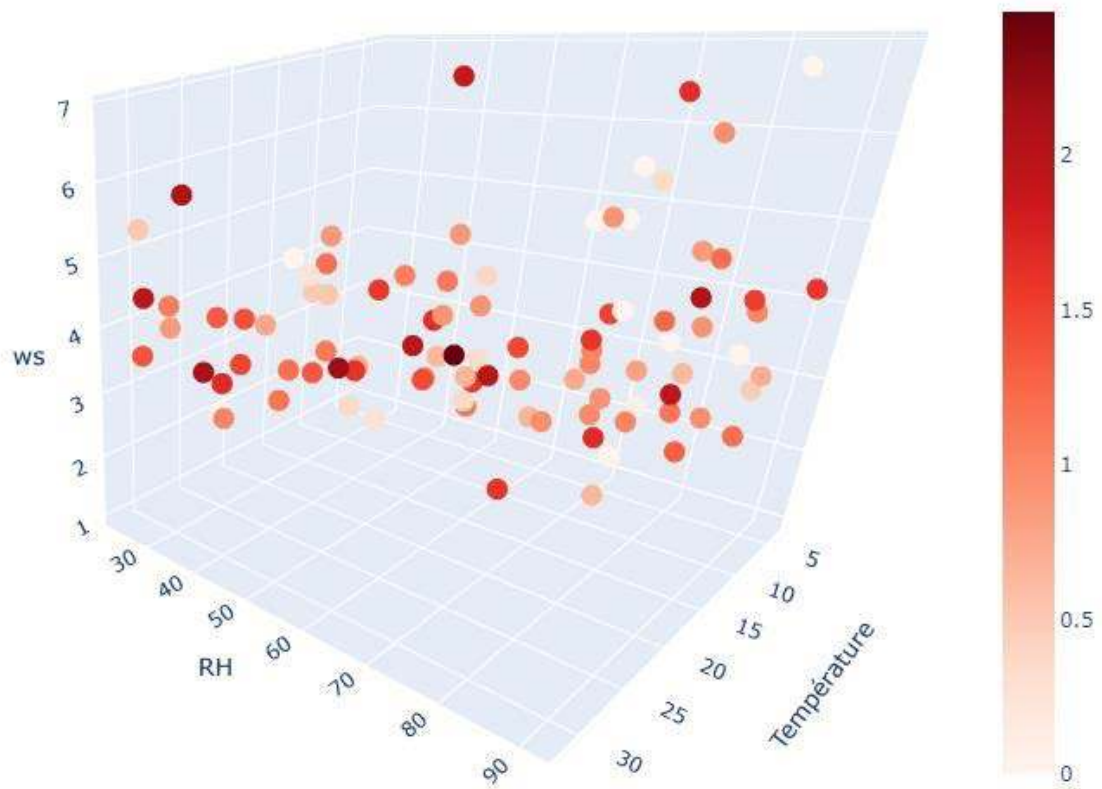


Fig. S18. Contributions ($\mu\text{g}/\text{m}^3$) of “industrial activities” as a function of wind speed (m/s), relative humidity (%) and temperature ($^{\circ}\text{C}$)

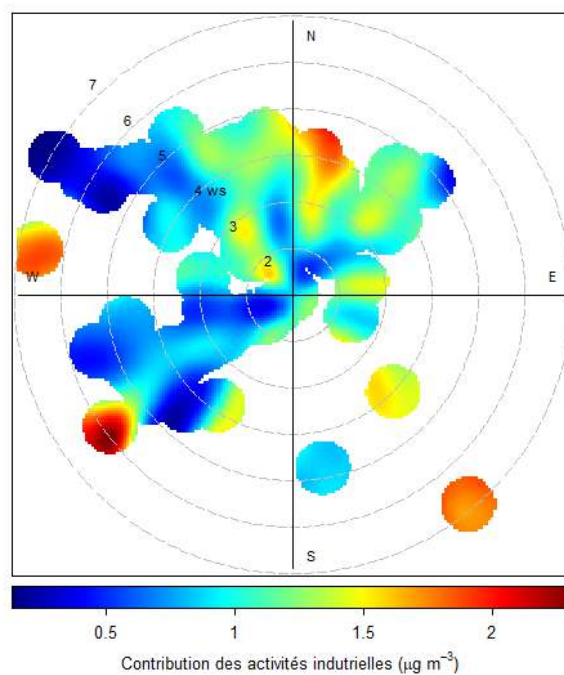


Fig. S19. Contribution of “industrial activities” as a function wind speed and direction

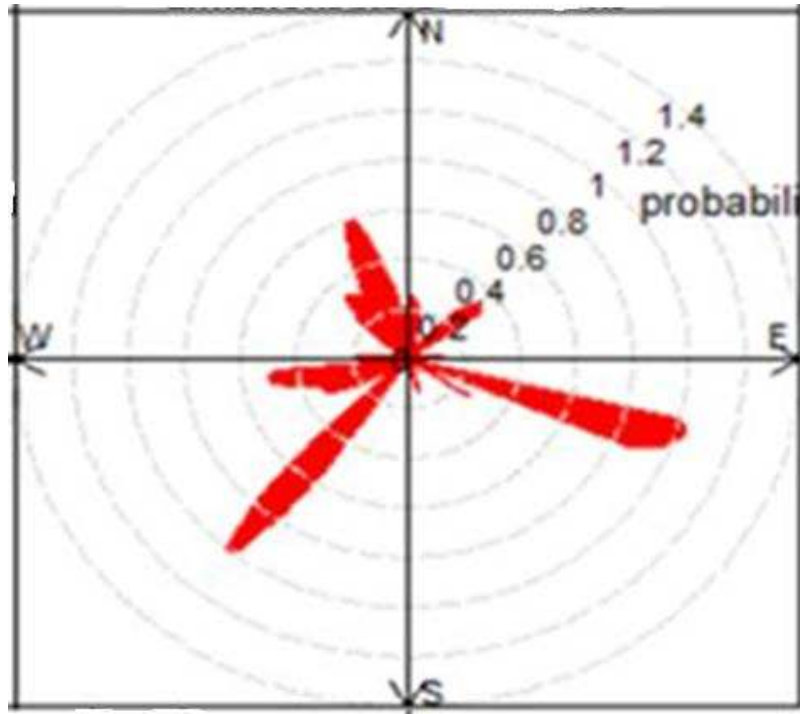


Fig. S20. 75th percentile of the conditional probability function (CPF) for the “non-exhaust emissions” factor contributing to PM_{2.5}

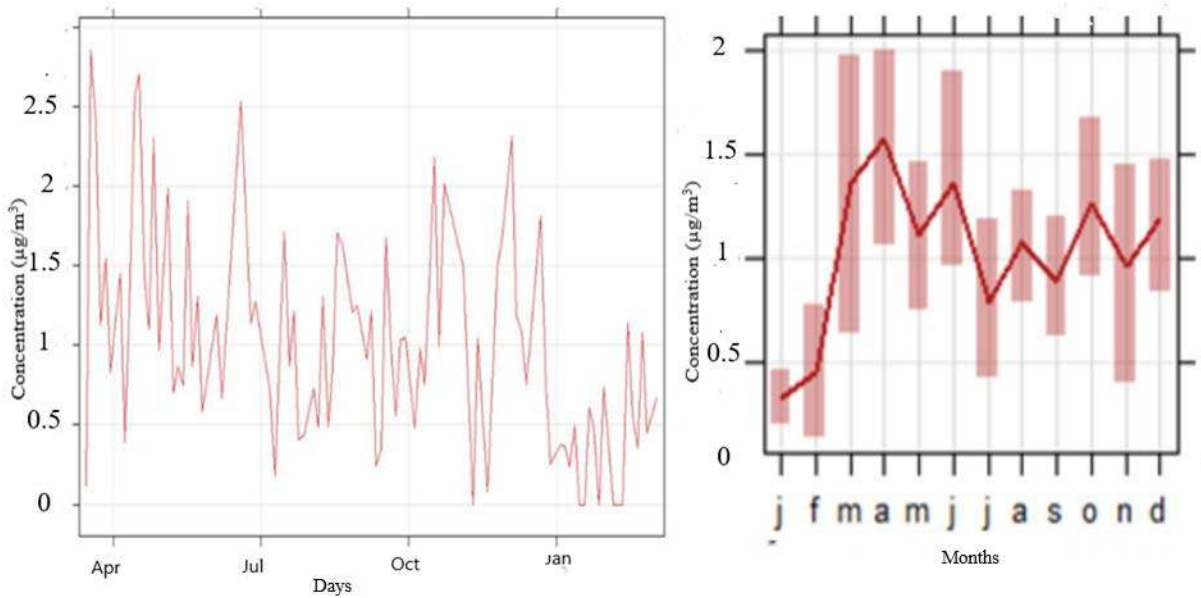


Fig. S21. Daily and monthly contribution of the “non-exhaust emissions” factor resolved by PMF

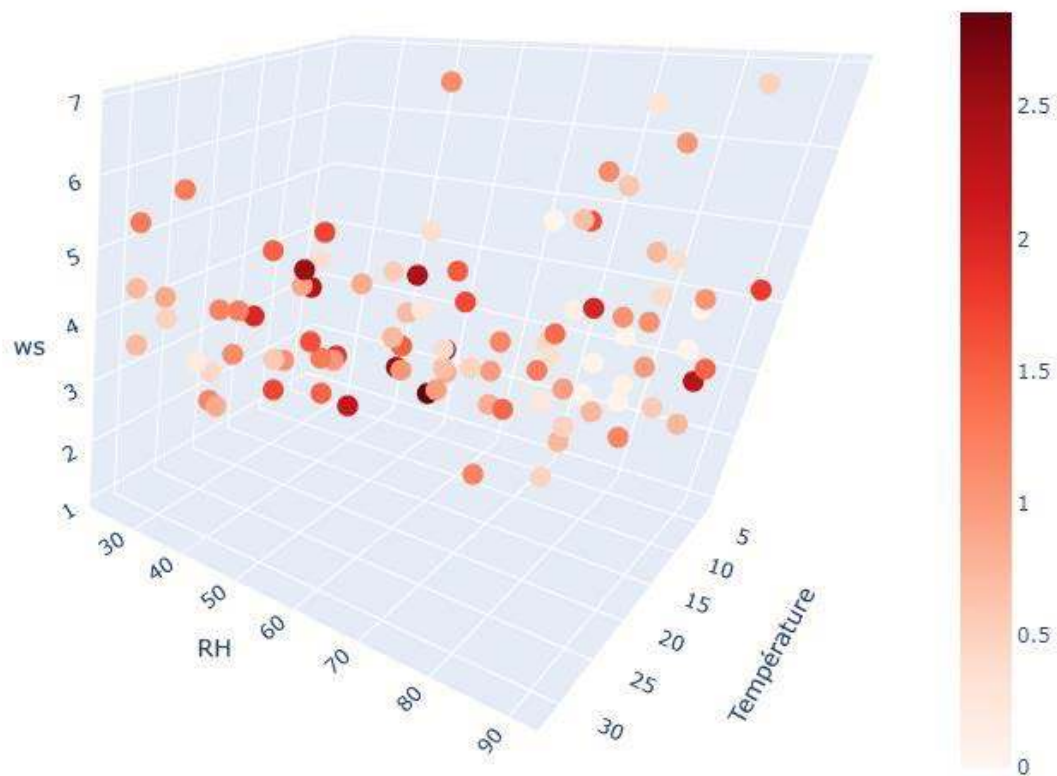


Fig. S22. Contributions ($\mu\text{g}/\text{m}^3$) of “non-exhaust emissions” as a function of wind speed (m/s), relative humidity (%) and temperature ($^{\circ}\text{C}$)

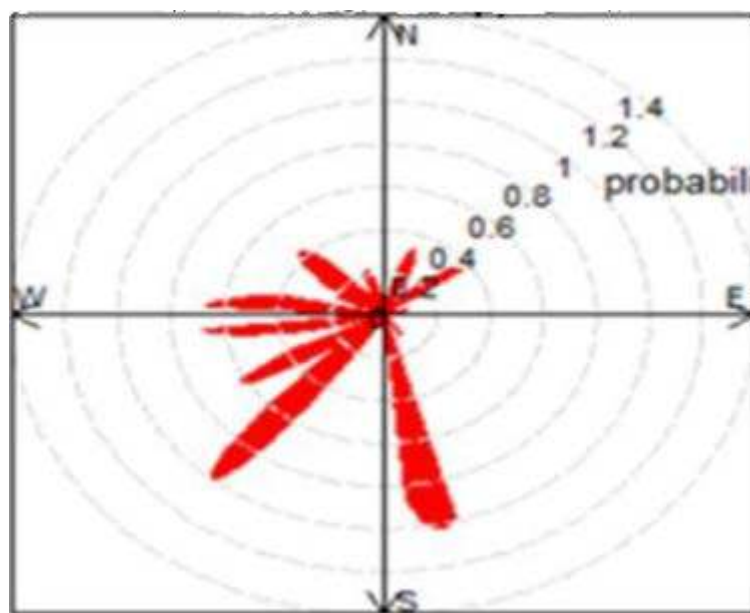


Fig. S23. 75th percentile of the conditional probability function (CPF) for “exhaust emissions” factor contributing to $\text{PM}_{2.5}$

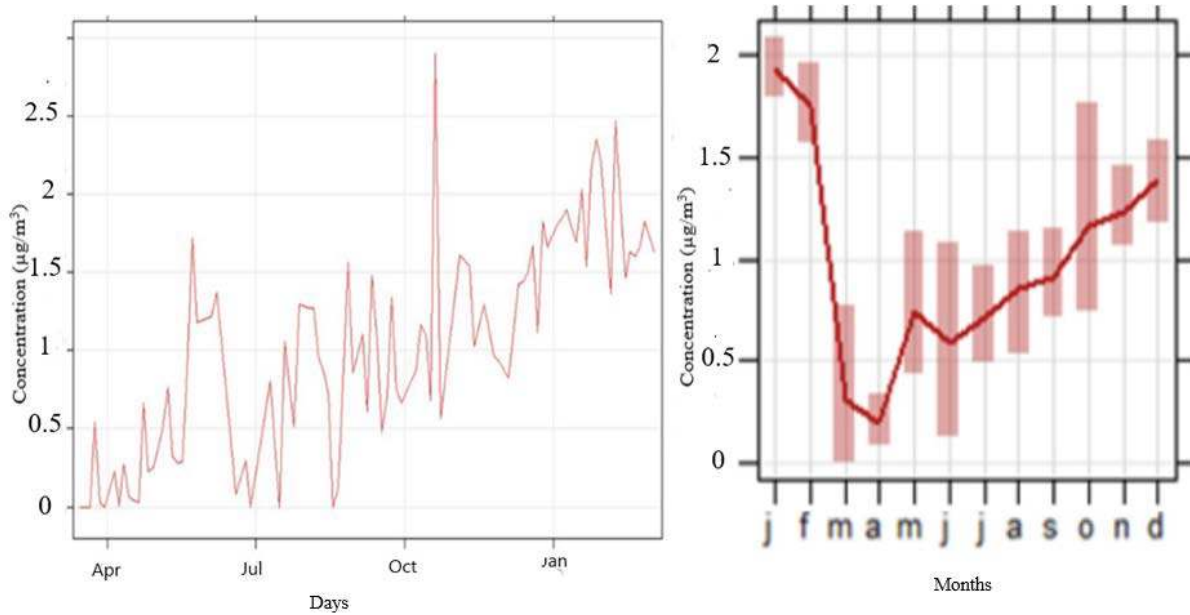


Fig. S24. Daily and monthly contribution of the "exhaust emissions" factor resolved by PMF

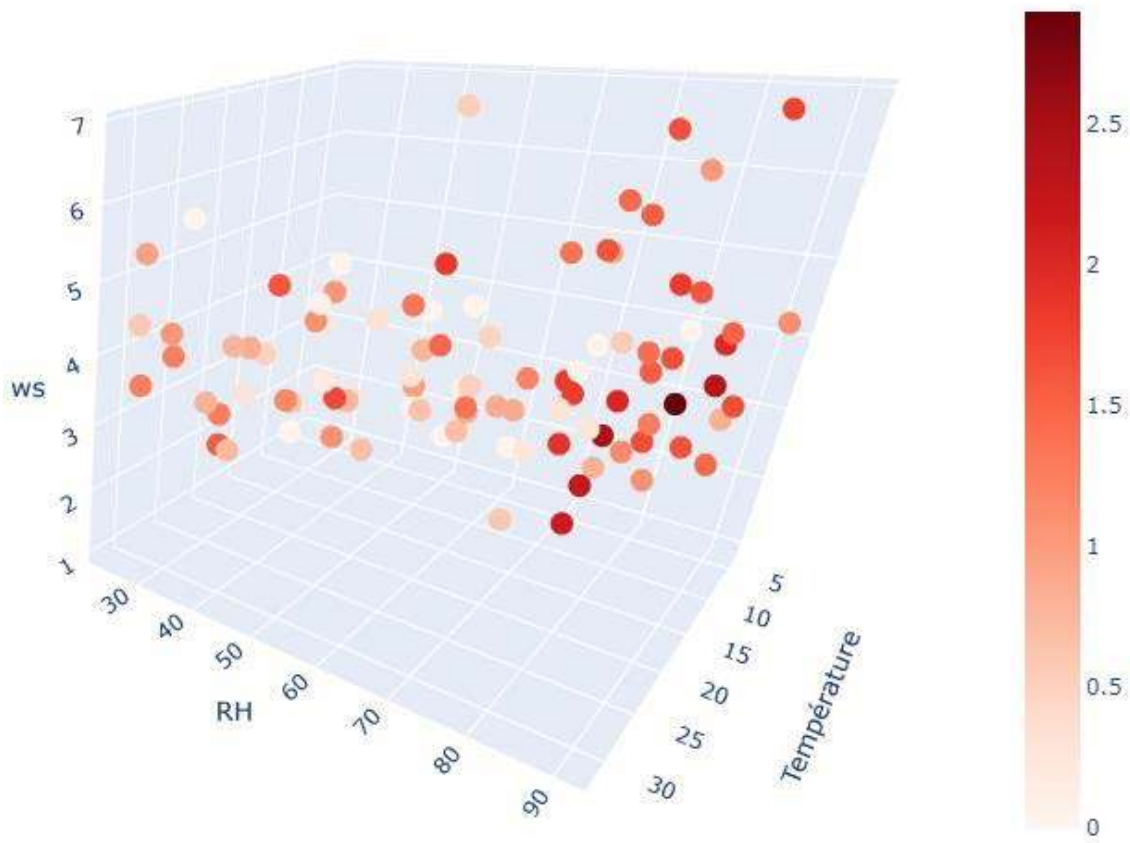


Fig. S25. Contributions ($\mu\text{g}/\text{m}^3$) of "exhaust emissions" as a function of wind speed (m/s), relative humidity (%) and temperature ($^{\circ}\text{C}$)

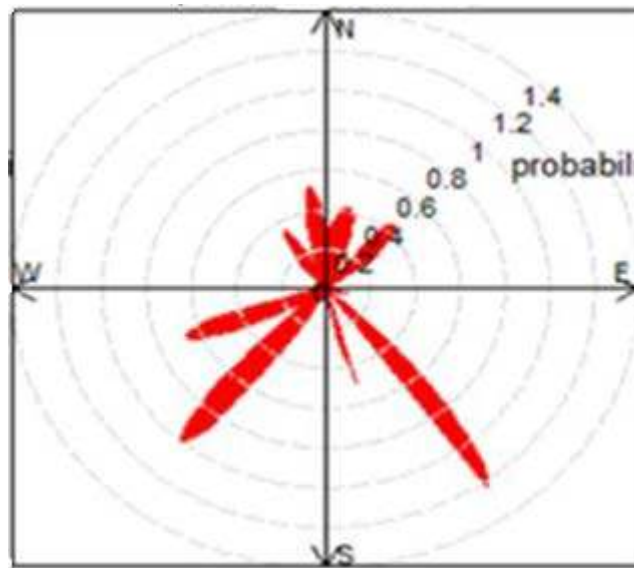


Fig. S26. 75th percentile of the conditional probability function (CPF) for the “mineral dust” factor contributing to PM_{2.5}

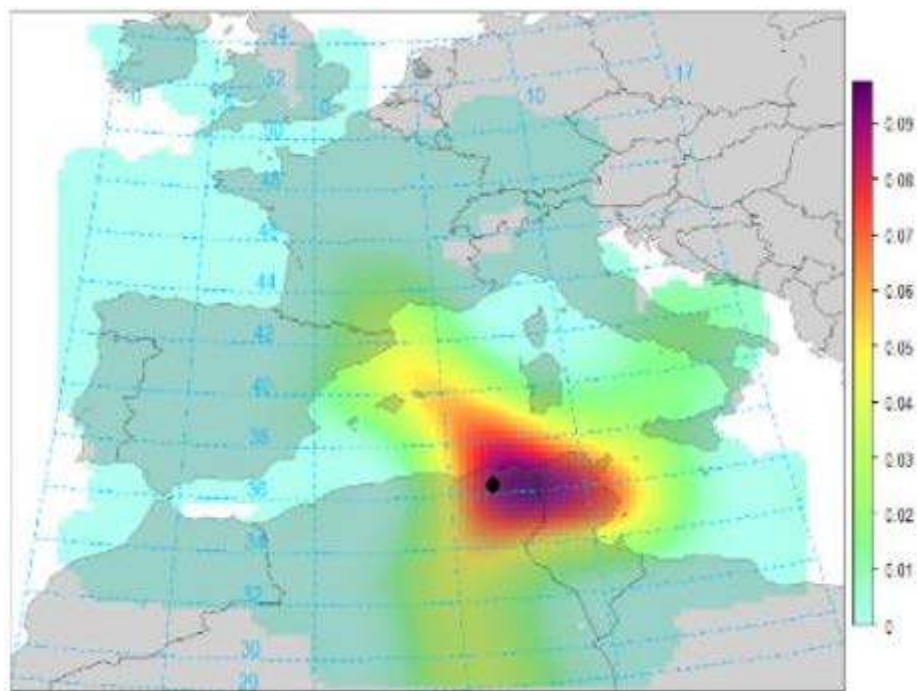


Fig. S27. 90th percentile of PSCF probabilities for the “mineral dust” contributions

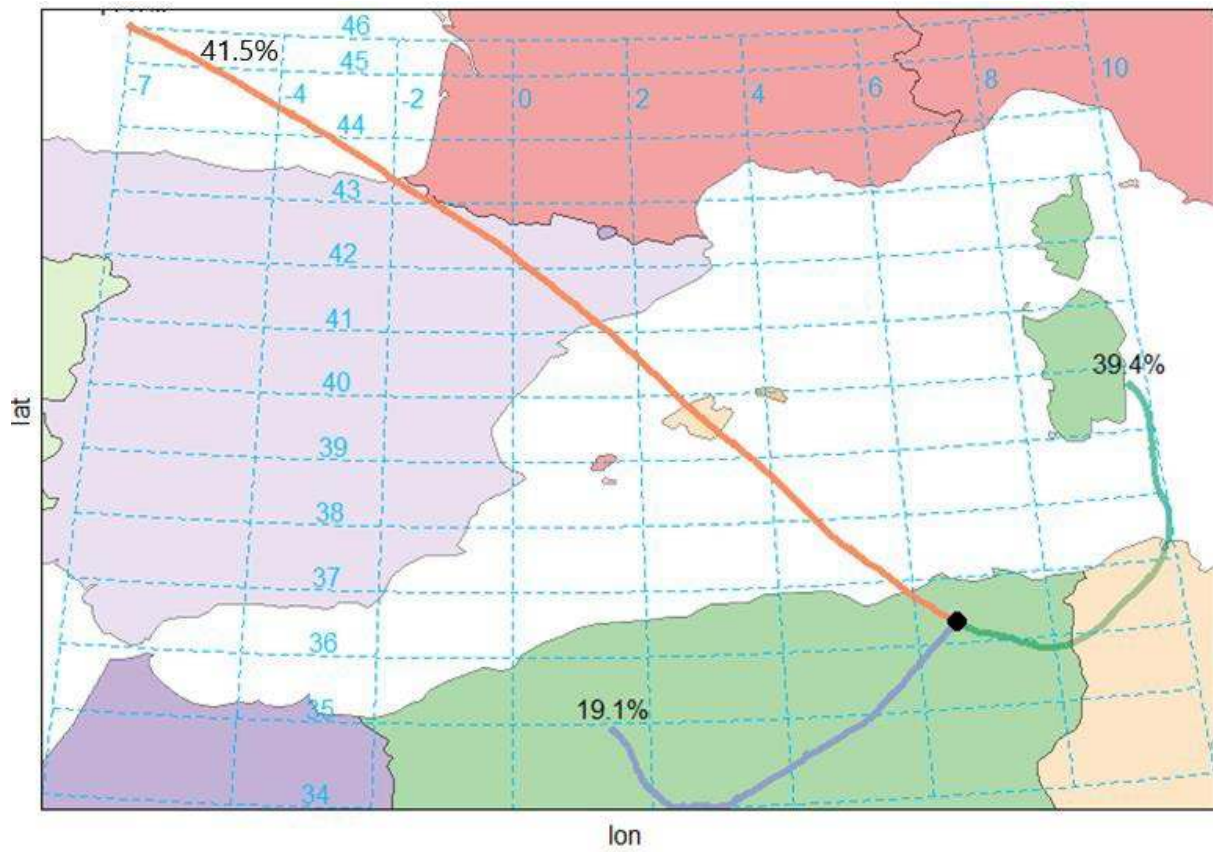


Fig. S28. Synoptic wind rose (clusters of back-trajectories calculated for our site for the study period)

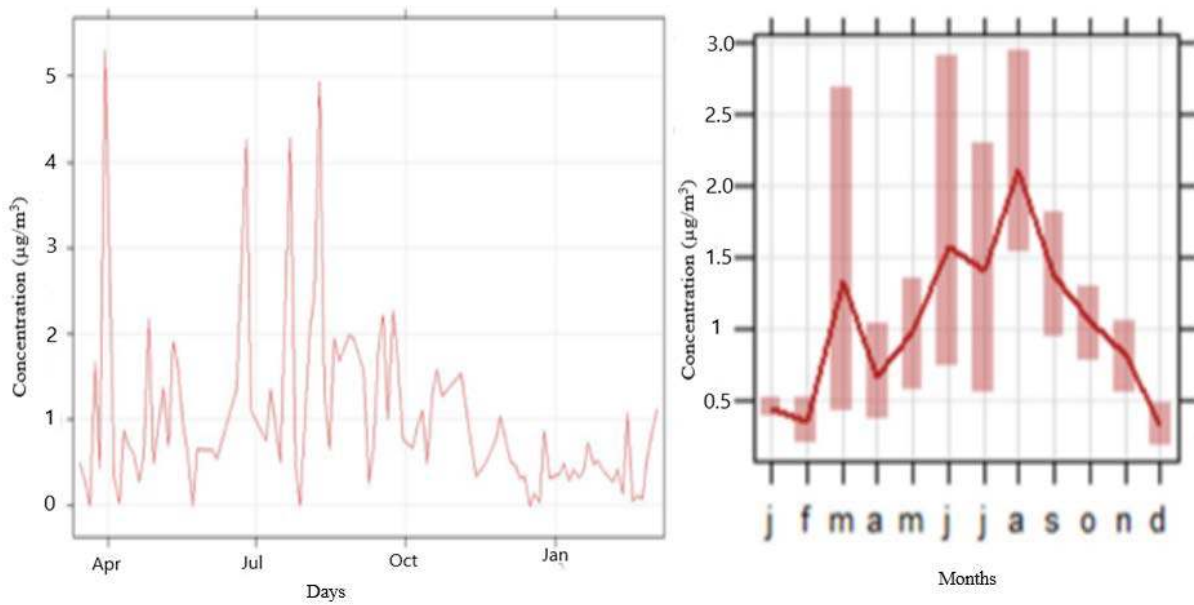


Fig. S29. Daily and monthly concentration of the "mineral dust" factor resolved by PMF

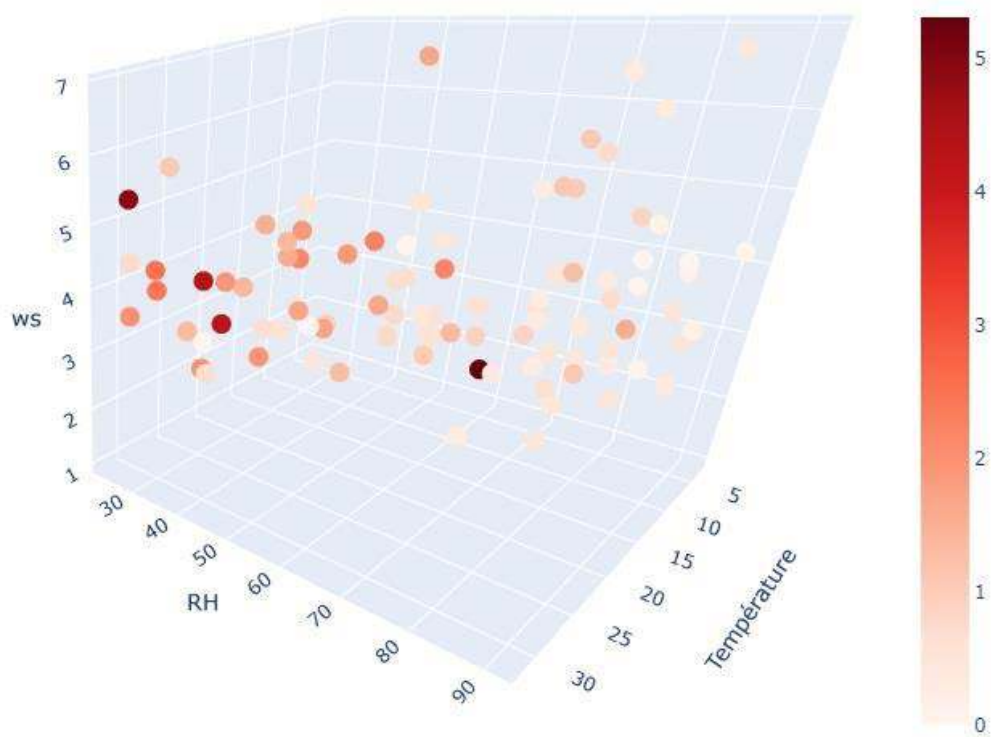


Fig. S30. Contributions ($\mu\text{g}/\text{m}^3$) of “mineral dust” as a function of wind speed (m/s), relative humidity (%) and temperature ($^{\circ}\text{C}$)

Factor Profiles (conc. of species) from Base Run #1 (Convergent Run)

| | Sea-salt | Industrial e | Non-exhaust | Exhaust | Mineral dust | | Sea-salt | Industrial e | Non-exhaust | Exhaust | Mineral dust |
|---------|-------------|--------------|-------------|----------|--------------|-------|----------|--------------|-------------|----------|--------------|
| 1 PM2.5 | 5,7995 | 7,2829 | 9,3096 | 6,0815 | 9,974 | | | | | | |
| 1 Na | 0,66525 | 0,10187 | 0 | 0 | 0 | | | | | | |
| 1 Mg | 0,043967 | 0,007336 | 0,0046574 | 0,015968 | 0,032069 | | | | | | |
| 1 P | 0 | 0,005763 | 0,0087976 | 0,004326 | 0,0082171 | | | | | | |
| 1 K | 0,094687 | 0,013319 | 0,0094812 | 0,004778 | 0,046773 | | | | | | |
| 1 Ca | 0,19822 | 0 | 0,06991 | 0,2533 | 0,57767 | | | | | | |
| 1 Sr | 0,0021654 | 0,000253 | 0,0002923 | 0,001785 | 0,0010173 | | | | | | |
| 1 Zr | 0,0041576 | 0,002918 | 0,0028646 | 0,002962 | 0,00058641 | | | | | | |
| 1 Ba | 0,04214 | 0 | 0 | 0,057337 | 0 | | 0,027338 | 0,036779 | 0,064337 | 0,021733 | |
| 1 Pb | 0 | 0 | 0,0080946 | 0,028764 | 0,0020408 | | | | | | |
| 1 Al | 0,25673 | 0,12004 | 0,099676 | 0,16212 | 0,097498 | | | | | | |
| 1 Ti | 0,003299 | 0,003242 | 0,0013406 | 0,004085 | 0,0062146 | Ca/Al | 0,772095 | 0 | 0,701372 | 1,562423 | 5,92494205 |
| 1 V | 0,000089098 | 0,00014 | 0 | 0 | 0,00030028 | | | | | | |
| 1 Cr | 0,00024465 | 0,003811 | 0,00013323 | 0,000227 | 0,00059013 | Ti/Al | 0,01285 | 0,027007 | 0,01345 | 0,025196 | 0,063740795 |
| 1 Mn | 0,0003818 | 0,001033 | 0,00073132 | 0,001111 | 0,001599 | Mn/Al | 0,001487 | 0,008602 | 0,007337 | 0,006854 | 0,016400336 |
| 1 Fe | 0,02247 | 0,037771 | 0,019884 | 0,01727 | 0,073576 | Fe/Al | 0,087524 | 0,314653 | 0,199486 | 0,106526 | 0,754641121 |
| 1 Ni | 0 | 0,00123 | 0,00094036 | 0,000197 | 0,00017775 | Zn/Al | 0,009137 | 0 | 0,12968 | 0 | 0 |
| 1 Cu | 0,00025244 | 0,000417 | 0,00034076 | 0 | 0,00032744 | Pb/Al | 0 | 0 | 0,081209 | 0,177424 | 0,020931711 |
| 1 Zn | 0,0023458 | 0 | 0,012926 | 0 | 0 | Zn/Pb | #DIV/0! | #DIV/0! | 1,596867 | 0 | 0 |
| 1 Mo | 0,00023708 | 0,007541 | 0,0092419 | 0,012087 | 0 | V/Ni | #DIV/0! | 0,113841 | 0 | 0 | 1,689338959 |
| 1 S | 0,0011189 | 0,066587 | 0,079397 | 0,077006 | 0,0033523 | Mn/Fe | 0,016992 | 0,027338 | 0,036779 | 0,064337 | 0,02173263 |
| | | | | | | Fe/Ca | 0,113359 | #DIV/0! | 0,284423 | 0,06818 | 0,127366836 |
| | | | | | | Na/Mg | 15,13067 | 13,88726 | 0 | 0 | 0 |
| | | | | | | Na/Sr | 307,2181 | 403,0305 | 0 | 0 | 0 |
| | | | | | | Sr/Mg | 0,049251 | 0,034457 | 0,06276 | 0,111774 | 0,031722224 |
| | | | | | | K/Fe | 4,21393 | 0,352625 | 0,476826 | 0,276653 | 0,635710014 |
| | | | | | | k/Al | | | | | 0,479732918 |

Factor Profiles (% of species sum) from Base Run #1 (Convergent Run)

| | Factor 1 | Factor 2 | Factor 3 | Factor 4 | Factor 5 |
|---------|-------------|----------|-------------|----------|-------------|
| 1 PM2.5 | 15,08420574 | 18,94245 | 24,21379804 | 15,81767 | 25,94186878 |
| 1 Na | 86,72046094 | 13,27954 | 0 | 0 | 0 |
| 1 Mg | 42,27722172 | 7,053576 | 4,478402722 | 15,3543 | 30,83649609 |
| 1 P | 0 | 21,26222 | 32,45987529 | 15,95986 | 30,31804597 |
| 1 K | 56,01521551 | 7,879293 | 5,608916338 | 2,826465 | 27,67010968 |
| 1 Ca | 18,03475571 | 0 | 6,360658721 | 23,04613 | 52,55845692 |
| 1 Sr | 39,2812051 | 4,585166 | 5,302436617 | 32,37697 | 18,45422091 |
| 1 Zr | 30,8237281 | 21,63355 | 21,23764949 | 21,95753 | 4,347542427 |
| 1 Ba | 42,36155091 | 0 | 0 | 57,63845 | 0 |
| 1 Pb | 0 | 0 | 20,80906132 | 73,94459 | 5,246353414 |
| 1 Al | 34,87876054 | 16,30836 | 13,54175724 | 22,02526 | 13,24585906 |
| 1 Ti | 18,14541634 | 17,83135 | 7,37367237 | 22,46753 | 34,18202619 |
| 1 V | 16,83101359 | 26,44474 | 0 | 0 | 56,72424476 |
| 1 Cr | 4,886930432 | 76,12945 | 2,661294672 | 4,534368 | 11,78795935 |
| 1 Mn | 7,862729673 | 21,2652 | 15,06069006 | 22,88182 | 32,9295567 |
| 1 Fe | 13,14257974 | 22,09205 | 11,63004252 | 10,10113 | 43,03419878 |
| 1 Ni | 0 | 48,3196 | 36,95032869 | 7,745596 | 6,984475015 |
| 1 Cu | 18,8720433 | 31,17431 | 25,47471667 | 0 | 24,47893305 |
| 1 Zn | 15,36033735 | 0 | 84,63966265 | 0 | 0 |
| 1 Mo | 0,814523726 | 25,90686 | 31,75192688 | 41,52669 | 0 |
| 1 S | 0,49190807 | 29,274 | 34,90573337 | 33,85457 | 1,473789816 |

Factor Profiles (% of factor total) from Base Run #1 (Convergent Run)

| | Factor 1 | Factor 2 | Factor 3 | Factor 4 | Factor 5 |
|---------|-------------|----------|-------------|----------|-------------|
| 1 PM2.5 | 81,25672091 | 95,1246 | 96,58955866 | 90,43362 | 92,12997974 |
| 1 Na | 9,320809309 | 1,330561 | 0 | 0 | 0 |
| 1 Mg | 0,616021079 | 0,095812 | 0,048321755 | 0,237449 | 0,296221809 |
| 1 P | 0 | 0,075269 | 0,091277423 | 0,064323 | 0,075901469 |
| 1 K | 1,326658356 | 0,173964 | 0,098369954 | 0,071047 | 0,432042866 |
| 1 Ca | 2,777257905 | 0 | 0,725334713 | 3,766642 | 5,335945999 |
| 1 Sr | 0,030339392 | 0,003301 | 0,00303269 | 0,02654 | 0,009396815 |
| 1 Zr | 0,058252081 | 0,038113 | 0,029720982 | 0,044041 | 0,005416678 |
| 1 Ba | 0,590423005 | 0 | 0 | 0,852617 | 0 |
| 1 Pb | 0 | 0 | 0,083983613 | 0,427729 | 0,018850899 |
| 1 Al | 3,597040772 | 1,567886 | 1,034164824 | 2,41077 | 0,900590412 |
| 1 Ti | 0,046222247 | 0,042344 | 0,013909079 | 0,060742 | 0,057404349 |
| 1 V | 0,001248351 | 0,001828 | 0 | 0 | 0,002773691 |
| 1 Cr | 0,003427788 | 0,049779 | 0,001382296 | 0,003376 | 0,005451039 |
| 1 Mn | 0,005349395 | 0,013487 | 0,007587638 | 0,016522 | 0,014769986 |
| 1 Fe | 0,314826885 | 0,493341 | 0,206301751 | 0,25681 | 0,679622558 |
| 1 Ni | 0 | 0,016062 | 0,009756483 | 0,002931 | 0,001641879 |
| 1 Cu | 0,003536934 | 0,005447 | 0,003535475 | 0 | 0,003024568 |
| 1 Zn | 0,032866974 | 0 | 0,134110664 | 0 | 0 |
| 1 Mo | 0,003321725 | 0,098491 | 0,095887153 | 0,179737 | 0 |
| 1 S | 0,015676894 | 0,869717 | 0,823764844 | 1,145101 | 0,030965243 |

Factor Contributions (avg = 1) from Base Run #1 (Convergent Run)

| | | Factor 1 | Factor 2 | Factor 3 | Factor 4 | Factor 5 |
|---|----------------|----------|----------|----------|----------|----------|
| 1 | 03/15/17 00:00 | 0,93715 | 1,4984 | 0,11435 | -0,2 | 0,47951 |
| 1 | 03/18/17 00:00 | 0,52385 | 1,1434 | 2,858 | -0,2 | 0,26622 |
| 1 | 03/21/17 00:00 | 0,99544 | 1,1193 | 2,3837 | -0,08507 | -0,02621 |
| 1 | 03/24/17 00:00 | 0,81526 | 1,8623 | 1,1277 | 0,53744 | 1,6621 |
| 1 | 03/27/17 00:00 | 0,78304 | 0,37965 | 1,5466 | 0,02531 | 0,4353 |
| 1 | 03/30/17 00:00 | 1,1348 | 0,65938 | 0,83198 | -0,2 | 5,3051 |
| 1 | 04/05/17 00:00 | 0,98103 | 0,95328 | 1,4495 | 0,22683 | 0,31756 |
| 1 | 04/08/17 00:00 | 0,71112 | 2,0384 | 0,38657 | 0,002102 | 0,02629 |
| 1 | 04/11/17 00:00 | 0,25796 | 0,75311 | 1,2367 | 0,27056 | 0,8706 |
| 1 | 04/14/17 00:00 | 1,0213 | 1,1582 | 2,5707 | 0,063247 | 0,67492 |
| 1 | 04/17/17 00:00 | 1,1165 | 0,34443 | 2,7012 | 0,045225 | 0,60349 |
| 1 | 04/20/17 00:00 | 1,8487 | 1,5917 | 1,4327 | 0,026261 | 0,27075 |
| 1 | 04/23/17 00:00 | 1,4618 | 1,3573 | 1,0976 | 0,66658 | 0,55577 |
| 1 | 04/26/17 00:00 | 1,9763 | 0,52581 | 2,3063 | 0,22356 | 2,1654 |
| 1 | 04/29/17 00:00 | 1,1742 | 0,91238 | 0,96905 | 0,25254 | 0,48131 |
| 1 | 05/05/17 00:00 | 2,1851 | 0,7672 | 1,9837 | 0,49408 | 1,3727 |
| 1 | 05/08/17 00:00 | 1,3608 | 1,6652 | 0,7019 | 0,76625 | 0,70186 |
| 1 | 05/11/17 00:00 | 1,3589 | 1,563 | 0,86635 | 0,32125 | 1,8989 |
| 1 | 05/14/17 00:00 | 1,4202 | 1,965 | 0,74219 | 0,28068 | 1,656 |
| 1 | 05/17/17 00:00 | 2,0926 | 0,73193 | 1,9089 | 0,28978 | 0,96572 |
| 1 | 05/20/17 00:00 | 1,3359 | 1,5556 | 0,86396 | 0,87856 | 0,60757 |
| 1 | 05/23/17 00:00 | 1,4809 | 2,1739 | 1,3083 | 1,7182 | -0,06721 |
| 1 | 05/26/17 00:00 | 1,3235 | 1,184 | 0,58105 | 1,1752 | 0,64791 |
| 1 | 06/04/17 00:00 | 1,6949 | 1,4638 | 1,1923 | 1,2203 | 0,64 |
| 1 | 06/07/17 00:00 | 1,1288 | 0,6624 | 0,67129 | 1,3721 | 0,53894 |
| 1 | 06/19/17 00:00 | 0,60801 | 0,26618 | 2,5335 | 0,082798 | 1,3506 |
| 1 | 06/25/17 00:00 | 0,7492 | 1,469 | 1,1391 | 0,29259 | 4,2639 |
| 1 | 06/28/17 00:00 | 1,0655 | 2,046 | 1,2757 | -0,2 | 1,1016 |
| 1 | 07/07/17 00:00 | 1,054 | 1,9566 | 0,71997 | 0,58861 | 0,74941 |
| 1 | 07/10/17 00:00 | 0,56699 | 2,0862 | 0,17481 | 0,80526 | 1,3505 |
| 1 | 07/16/17 00:00 | 1,4164 | 0,87679 | 1,7157 | -0,17947 | 0,49944 |
| 1 | 07/19/17 00:00 | 2,6392 | 1,0747 | 0,87465 | 1,0534 | 2,5299 |
| 1 | 07/22/17 00:00 | 2,2553 | 1,3368 | 1,2125 | 0,78214 | 4,273 |
| 1 | 07/25/17 00:00 | 2,3021 | 2,4626 | 0,40385 | 0,50698 | 0,45765 |
| 1 | 07/28/17 00:00 | 1,4236 | 1,6782 | 0,43323 | 1,2925 | -0,00714 |
| 1 | 08/03/17 00:00 | 2,5532 | 1,3723 | 0,73031 | 1,2705 | 2,0465 |
| 1 | 08/06/17 00:00 | 3,3523 | 0,86483 | 0,48617 | 1,2684 | 2,4291 |
| 1 | 08/09/17 00:00 | 3,9549 | 0,51984 | 1,3022 | 0,95831 | 4,9371 |
| 1 | 08/12/17 00:00 | 3,0057 | 1,9693 | 0,48436 | 0,86339 | 1,3287 |
| 1 | 08/15/17 00:00 | 3,4636 | 1,031 | 0,86107 | 0,72388 | 0,65667 |
| 1 | 08/18/17 00:00 | 0,15934 | 1,1515 | 1,7025 | -0,00011 | 1,9498 |
| 1 | 08/21/17 00:00 | -0,15608 | 1,1172 | 1,6292 | 0,13028 | 1,679 |
| 1 | 08/27/17 00:00 | 0,1057 | -0,03606 | 1,2073 | 1,5641 | 1,9938 |
| 1 | 08/30/17 00:00 | -0,2 | 1,4111 | 1,253 | 0,85778 | 1,939 |
| 1 | 09/05/17 00:00 | 1,0341 | 0,49112 | 0,90837 | 1,1039 | 1,583 |
| 1 | 09/08/17 00:00 | 0,31234 | 1,5803 | 1,2149 | 0,60974 | 0,25427 |
| 1 | 09/11/17 00:00 | 0,42517 | 0,90328 | 0,24051 | 1,4741 | 0,66438 |
| 1 | 09/14/17 00:00 | 0,71059 | 1,2001 | 0,34979 | 1,0591 | 1,8876 |
| 1 | 09/17/17 00:00 | 1,0331 | 0,9081 | 1,6755 | 0,48258 | 2,2076 |
| 1 | 09/20/17 00:00 | 0,68132 | 0,3603 | 0,98283 | 0,70023 | 0,99901 |
| 1 | 09/23/17 00:00 | 0,55183 | 1,0662 | 0,55491 | 1,3409 | 2,261 |
| 1 | 09/26/17 00:00 | 0,4856 | 1,6144 | 1,035 | 0,74748 | 1,7224 |
| 1 | 09/29/17 00:00 | 0,5088 | 1,4314 | 1,0473 | 0,66025 | 0,76331 |
| 1 | 10/05/17 00:00 | 0,51793 | 1,6672 | 0,48331 | 0,80293 | 0,66176 |
| 1 | 10/08/17 00:00 | 0,72921 | 0,98718 | 0,98201 | 0,86858 | 0,92607 |

| | | | | | | |
|---|----------------|----------|----------|----------|---------|----------|
| 1 | 10/11/17 00:00 | 0,68027 | 1,0374 | 0,74918 | 1,1656 | 1,0979 |
| 1 | 10/14/17 00:00 | -0,03952 | 0,35242 | 1,4791 | 1,1052 | 0,47947 |
| 1 | 10/17/17 00:00 | -0,02238 | 0,24553 | 2,1811 | 0,68237 | 1,2991 |
| 1 | 10/20/17 00:00 | 0,88274 | 0,63216 | 0,99228 | 2,9001 | 1,5746 |
| 1 | 10/23/17 00:00 | 0,07709 | -0,2 | 2,0167 | 0,56208 | 1,274 |
| 1 | 11/04/17 00:00 | 0,040628 | -0,16409 | 1,4919 | 1,6077 | 1,5423 |
| 1 | 11/10/17 00:00 | 0,085167 | -0,19811 | -0,19126 | 1,5404 | 0,73106 |
| 1 | 11/13/17 00:00 | 0,26406 | 0,95832 | 1,0364 | 1,0262 | 0,3236 |
| 1 | 11/19/17 00:00 | -0,2 | 1,9166 | 0,078076 | 1,2953 | 0,49822 |
| 1 | 11/25/17 00:00 | 0,015858 | 0,63012 | 1,5035 | 0,96284 | 0,76455 |
| 1 | 11/28/17 00:00 | 0,061548 | 0,021175 | 1,6593 | 0,93622 | 1,0309 |
| 1 | 12/04/17 00:00 | 0,51746 | 0,46225 | 2,3128 | 0,82172 | 0,51477 |
| 1 | 12/07/17 00:00 | 0,058561 | 1,2975 | 1,184 | 1,0909 | 0,4635 |
| 1 | 12/10/17 00:00 | 1,3456 | 1,2405 | 1,0868 | 1,4208 | 0,31263 |
| 1 | 12/13/17 00:00 | 1,051 | 1,1949 | 0,7524 | 1,437 | 0,33626 |
| 1 | 12/16/17 00:00 | 1,1221 | 1,5223 | 1,0653 | 1,4926 | -0,2 |
| 1 | 12/19/17 00:00 | 1,2599 | 0,71255 | 1,4694 | 1,6694 | 0,13583 |
| 1 | 12/22/17 00:00 | 1,0803 | 1,5763 | 1,8078 | 1,116 | 0,029742 |
| 1 | 12/25/17 00:00 | 0,33827 | 0,86636 | 0,73815 | 1,8251 | 0,86499 |
| 1 | 12/28/17 00:00 | 0,082406 | 1,6535 | 0,25041 | 1,659 | 0,31853 |
| 1 | 01/03/18 00:00 | 0,22467 | 1,022 | 0,37087 | 1,8067 | 0,35138 |
| 1 | 01/06/18 00:00 | 0,34943 | 0,88891 | 0,36664 | 1,8487 | 0,48096 |
| 1 | 01/09/18 00:00 | 0,28235 | 0,97626 | 0,23797 | 1,9026 | 0,28217 |
| 1 | 01/12/18 00:00 | 0,4782 | -0,05929 | 0,49506 | 1,7751 | 0,41457 |
| 1 | 01/15/18 00:00 | 0,20102 | 1,1394 | -0,2 | 1,6911 | 0,31277 |
| 1 | 01/18/18 00:00 | 0,32572 | 0,80309 | -0,2 | 2,031 | 0,3836 |
| 1 | 01/21/18 00:00 | 0,39137 | 0,35355 | 0,60927 | 1,5367 | 0,7286 |
| 1 | 01/24/18 00:00 | 1,3682 | 0,64605 | 0,47797 | 2,1517 | 0,47974 |
| 1 | 01/27/18 00:00 | 1,4218 | -0,15233 | -0,17412 | 2,356 | 0,50585 |
| 1 | 01/30/18 00:00 | 1,329 | -0,2 | 0,7369 | 2,2032 | 0,40507 |
| 1 | 02/05/18 00:00 | 0,77747 | -0,14064 | -0,04998 | 1,3658 | 0,26728 |
| 1 | 02/08/18 00:00 | 1,1813 | 0,080923 | -0,05342 | 2,4706 | 0,41848 |
| 1 | 02/11/18 00:00 | 1,0073 | 1,0197 | -0,05395 | 1,9838 | 0,13387 |
| 1 | 02/14/18 00:00 | 0,96775 | -0,02375 | 1,1387 | 1,4658 | 1,0745 |
| 1 | 02/17/18 00:00 | 0,92064 | 0,95505 | 0,57306 | 1,634 | 0,054568 |
| 1 | 02/20/18 00:00 | 1,2785 | 1,1944 | 0,35167 | 1,5977 | 0,10355 |
| 1 | 02/23/18 00:00 | 1,3466 | 0,90795 | 1,078 | 1,6581 | 0,071953 |
| 1 | 02/26/18 00:00 | 1,2241 | 1,0547 | 0,44765 | 1,8229 | 0,5492 |
| 1 | 03/04/18 00:00 | 1,7405 | 0,89931 | 0,67116 | 1,6301 | 1,1145 |

