

Identification of sources of atmospheric particulate matter and trace metals in Constantine, Algeria

Ahmed Terrouche · Hocine Ali-Khodja ·
Amina Kemmouche · Mokhtar Bouziane ·
Ahmed Derradji · Aurélie Charron

Received: 4 October 2014 / Accepted: 12 December 2014 / Published online: 17 February 2015
© Springer Science+Business Media Dordrecht 2015

Abstract Very few previous investigations of respirable PM have been undertaken in the city of Constantine, Algeria. In general, air quality in Algeria and other Maghreb countries has not been extensively studied because of the insufficient number of researchers involved in this field and the unreliability of the few air quality monitoring networks installed in Algeria. It is therefore of utmost importance to address this problem in order to estimate the concentration levels of PM₁₀ and their origins. The purpose of this study was to identify the different sources of PM₁₀ and some chemical elements (Pb, Cu, Zn, Fe, K, Ca, Na, Mg) at a traffic site at Zouaghi, in the south of Constantine, using different statistical methods: factor analysis (FA) to categorise the different trace elements according to their origin, and the enrichment factor (EF) to identify terrigenous elements and those having a marine origin. We also used back-trajectories clustering to identify potential distant sources that contribute to particulate pollution and metallic elements in our site.

Keywords PM₁₀ · PM_{2.5} · Traffic site · Factor analysis · Back-trajectories · Enrichment factors

Introduction

Motor vehicles strongly affect air quality within urban areas (Pastuszka et al. 2010). Several studies have provided

evidence that the exposure to high concentrations of aerosols is associated with adverse health effects (Pateraki et al. 2012). Multi-city extensive studies conducted in the United States and in Europe reported positive associations between PM₁₀ and death (Rückerl et al. 2011). In a multicentre study involving four European cities, consistent positive associations were found between coarse particles central sites concentrations and prevalence of respiratory symptoms (Karakatsani et al. 2012). Another particulate matter health study in China revealed a 10 $\mu\text{g}/\text{m}^3$ increase in 2-day moving-average PM₁₀ was associated with a 0.35 % increase of total mortality, 0.44 % increase of cardiovascular mortality and 0.56 % increase of respiratory mortality (Chen et al. 2012). In a study that examines the relation of lung cancer incidence with long-term residential exposures to ambient particulate matter, it was established that a 10- $\mu\text{g}/\text{m}^3$ increase in 72 months average PM₁₀ was positively associated with lung cancer (Puett et al. 2014). Moreover, adverse health effects may be caused by mineral dusts originating from the Sahara (Morman and Plumlee 2014).

Lim et al. (2012), in the framework of the WHO-driven evaluation of the Global Burden of Disease, evidenced that particulate atmospheric pollution is the fourth cause of worldwide mortality in developing countries and the 11th one in central Europe. REVHIHAAP (Review of evidence on health aspects of air pollution) (WHO 2013) and HRAPIE (Health risks of air pollution in Europe) (Henschel and Chan 2013) evidenced that mean life expectancy of European citizens is reduced by 9 months due to increase on premature mortality due to cardiovascular, respiratory and cerebro-vascular causes. These reports also indicate that atmospheric particulate matter (PM) is the main pollutant causing these health outcomes.

Studies on traffic-related airborne particulate matter, especially PM_{2.5}, are scarce or even not available in many cities in the developing world and particularly in Africa (Han 2006;

A. Terrouche · H. Ali-Khodja (✉) · A. Kemmouche ·
M. Bouziane · A. Derradji
Laboratoire de Pollution et Traitement des Eaux, Département de
Chimie, Faculté des Sciences Exactes, Université Constantine 1,
Constantine, Algeria
e-mail: hocine_ak@yahoo.fr

A. Charron
IFSTTAR, LTE, 25, avenue François Mitterrand,
Case 24 Cité des mobilités, F-69675 Bron Cedex, France

Eliasson et al. 2009). This is in spite of the high levels of atmospheric particulate pollution observed in African cities (Val et al. 2013).

In the developing countries, the particulate matter forms the major contributor to air pollution and hence the pressure to understand its sources better. According to Lioussé et al. (2014), emissions from combustion of fossil fuels are expected to increase significantly in African cities in the near future. Atmospheric particulate pollution is more severe in developing countries than in developed countries because of rapid urbanisation and a sudden expansion in the number of vehicles. Petkova et al. (2013) reported that annual PM levels in Northern Africa exceeded annual and 24 h WHO guidelines, with annual PM₁₀ levels exceeding 150 $\mu\text{g}/\text{m}^3$ in different sites of Cairo while mean PM₁₀ levels across a network of four monitoring stations that was established in Algiers ranged from approximately 38 to 129 $\mu\text{g}/\text{m}^3$ between 2002 and 2003. Laid et al. (2006) reported that the daily average level of PM₁₀ at the Mustapha hospital in downtown Algiers was 61 $\mu\text{g}/\text{m}^3$ for the period extending from 1 October 2001 to 30 September 2002. Winter was characterised by a significantly higher average PM₁₀ concentration than summer (74 vs. 48 $\mu\text{g}/\text{m}^3$). In a more recent study, Bouchlaghem and Blaise (2012) reported annual average concentrations of 58, 80, 89, 90 and 87 $\mu\text{g}/\text{m}^3$ in Sousse, Bizerte, Sfax industrial site, Tunis and Sfax centre, respectively.

The Mediterranean area is affected by natural mineral dust transport from the Sahara (Rodríguez et al. 2007). According to the study of Bouchlaghem et al. (2009), daily PM₁₀ levels recorded at seven monitoring stations in the Tunisian coasts during dust episodes (up to 700 $\mu\text{g}/\text{m}^3$) were higher than the average levels during non-dust episodes (up to 100 $\mu\text{g}/\text{m}^3$).

Algerian cities are growing rapidly, offering employment, better living conditions, access to universities, and other living standards that are not available in rural areas. The rapid urbanisation, however, has some environmental collateral problems such as air pollution, congestion and imbalance in fragile ecosystems. The results of a study carried out in a residential area with heavy traffic in Constantine from 25 March 2010 to 24 May 2010 showed that the average PM₁₀ concentration was 49 $\mu\text{g}/\text{m}^3$ (Terrouche et al. 2014). PM₁₀ concentrations ranged from 7.59 to 159.4 $\mu\text{g}/\text{m}^3$. Mean PM levels for days with African dust intrusions reached 68 $\mu\text{g}/\text{m}^3$ for PM₁₀, and for the rest of the days 45 $\mu\text{g}/\text{m}^3$. Ali-Khodja et al. (2008) reported that dust storm events from the Sahara led to a peak daily concentration of total suspended particles (TSP) of 9591 $\mu\text{g}/\text{m}^3$ in the town of Didouche Mourad which is located 13 km north of Constantine on 15 November 2002. Mean TSP concentrations were 300 and 117 $\mu\text{g}/\text{m}^3$ with and without dust events, respectively. The present study aims to investigate further

the ambient air levels of atmospheric particulates in a heavy traffic hotspot site in the vicinity of a roundabout.

Constantine has a population of 0.6 million (1,300,000 with the agglomeration), making it the third largest city in the country after Algiers and Oran. The province of Constantine has an area of 2,187 km² and contains seven industrial areas of which three are under construction and 11 business parks. Industrial sources include food industry, bricks and related clay products, cement manufacturing, glass manufacturing, lime manufacturing, limestone and clay quarries, pharmaceutical industry and mechanical industry. The total area occupied by industrial areas and business parks that has the province has now reached 1,000 ha. Ongoing projects of the expansion of the existing parks and the creation of new ones will, in the short-term, make Constantine the province that has the largest business and industrial parks in the country with a total area of over 2,340 ha.

The vehicle fleet, at national level, is currently estimated at 8 million vehicles. According to the Ministry of Transport, this number will increase to 21 million vehicles by 2025–2030. Roads can no longer withstand the flow of vehicles that is increasing rapidly. Road networks are becoming heavily congested as never before within cities. Moreover, imported vehicles do not meet any emission standard.

A study in 239 U.S. cities showed a PM_{2.5}/PM₁₀ ratio between 44 and 71 %, while it is around 50 % in Cairo (WHO 2006). PM_{2.5}/PM₁₀ ratio varies from 40 to 80 % across Europe, by region and type of measurement site (Querol et al. 2004). It is more significant when the particles have an anthropogenic origin, especially in connection with the production of fine particles of combustion. Conversely, the PM_{2.5}/PM₁₀ ratio is lower in regions where the sources of coarse particles are the most important as is the case in our site where the PM_{2.5}/PM₁₀ ratio is of the order of 47 % due to the significant contribution of resuspended coarse particles.

The distribution of trace elements in the different particle sizes may lead to estimation of sources of particles in the air. Traffic, for example, is a major source of PM causing higher levels of particles in urban air. A study of the size and composition of particles emitted by motor vehicles showed that coarse particles are mainly composed of abrasion and resuspension particles (street dust, tire wear, break wear), although the combustion process is the main source of fine particles (Handler et al. 2008).

The main objectives of this study were: (1) to assess the temporal variations of atmospheric particulates and associated metallic elements; (2) to identify distant sources of PM₁₀ by calculating 3 days back-trajectories; (3) to identify potential pollution sources of PM₁₀, PM_{2.5} and metallic elements based on factor analysis and cluster analysis.

Methods

Site description

The sampling site was located at the entrance to the campus of the Faculty of Earth Sciences located at Zouaghi, Constantine, nearby National Road 79, which is one of the busiest traffic highways in the city of Constantine, (36°22'N, 6°40'E, 640 m.a.s.l), Algeria (Fig. 1). The sampling device was placed about 5 m above the ground and about 6 m from the road. The position of the sampling site was such that it could be considered a traffic station because it is directly influenced by vehicular emissions. Sampling was scheduled at midnight.

Measurement of fine particulate matter

A portable low-volume air sampler Model Minivol TAS with a rate of 5 l/min was used. Ambient air particulates were trapped by a quartz filter (47 mm in diameter with a porosity of 0.2 μm). After each sampling interval, the collection media were returned to the weighing laboratory and allowed to

equilibrate for 24 h in a desiccator before weighing to a precision less than ± 0.01 mg using a Shimadzu balance (model AUW120D). The initial weights were determined after a similar period of desiccation.

Analysis of trace elements

Each filter was digested according to the method of Kuvarega and Taru (2008). A Shimadzu-7000 AAS supporting an acetylene flame was used to analyse metallic elements Fe, Na, Mg, Ca, Zn and K, while Pb and Cu were analysed using polarography (VA Computrace797). To minimise the effects of matrices, the standard addition technique was used for the determination of all metals. The results of the analysis of ten blank samples were used to estimate the element concentration produced by the filter and sample preparation.

Factor analysis

Factor analysis attempts to identify underlying variables, or factors, that explain the pattern of correlations within a set of observed variables. The primary interest of this type of analysis is to replace the original variables, generally correlated



Fig. 1 Fine particulate matter site sampling locations

with more easily treated uncorrelated variables (Dagnelie 1975). This technique tries to explain a set of data in a number smaller than the number of starting dimensions. The technique is to summarise the data-matrix with minimal and controlled loss of initial information by a reduced number of factors as differentiated as possible. This is a linear transformation whose general pattern is written in the form:

$$X_i = A_{i1}F_1 + A_{i2}F_2 + \dots + A_{ik}F_k + U_i$$

with X as the variable,

F , the common factor,

U , single factor,

and A , coefficient used to combine k factors.

The data were processed using IBM SPSS Statistics 20.0 software. The main sources of PM10 were identified by performing a Varimax rotation which has reduced the initial number of variables to a lesser number of independent variables (factors) and which estimated values of the factors (factor score) for each sample.

Results and Discussion

A total of 117 PM10 samples were collected every other day from 23 March 2011 to 22 November 2011. Sampling was scheduled at midnight in order to facilitate comparison between weekdays and weekends. Over this period, six daily measurements were missing. Occasional failures of the battery packs lead to gaps in the sampling schedule.

Moreover, 32 pairs of samples of PM10 and PM2.5 were collected simultaneously at the same sampling location at Zouaghi, Constantine, nearby the above-mentioned traffic site at random dates between 23 December 2011 and 8 January 2013 in order to identify the associated meteorological and source emissions characteristics.

Table 1 Statistical analysis of experimental results

	Average ($\mu\text{g}/\text{m}^3$)	Minimum ($\mu\text{g}/\text{m}^3$)	Maximum ($\mu\text{g}/\text{m}^3$)	Standard deviation (SD) ($\mu\text{g}/\text{m}^3$)
PM10	80.42	14.52	161.84	32.03
Pb	0.90	0.01	4.75	0.67
Cu	0.45	0.01	5.56	0.63
Na	5.61	1.57	27.77	3.05
K	4.08	0.01	23.82	5.95
Ca	3.01	0.01	26.94	5.14
Mg	1.83	0.07	6.80	1.20
Fe	3.11	0.01	10.89	2.57
Zn	1.29	0.02	22.20	2.56

PM levels

Table 1 shows some statistics of the experimental dataset. The minimum and maximum concentrations of PM10 were 14.52 and 161.84 $\mu\text{g}/\text{m}^3$, respectively. The average daily concentration of PM10 (80.42 $\mu\text{g}/\text{m}^3$) exceeds the annual limit of 40 $\mu\text{g}/\text{m}^3$ value within the EU, and the WHO guideline value of 20 $\mu\text{g}/\text{m}^3$. It is however very close to the Algerian annual limit of 80 $\mu\text{g}/\text{m}^3$. Table 2 shows the Algerian and EU standard limits and the WHO guideline values for particulate matter. The daily EU limit of 50 $\mu\text{g}/\text{m}^3$ was exceeded 85 times out of 117 samples which represents 73 % of the sampling period. The Algerian limit is more permissive because African dust outbreaks are expected to contribute to the increase of the daily PM10 concentration. Indeed, with the help of satellite data, Meloni et al. (2007) located the heaviest dust sources in Mauritania and Southern Algeria. They also identified Tunisia, northern and central Algeria, Morocco and Mali as the regions with the most frequent loading by using air masses back-trajectories. The more stringent WHO guideline value of 20 $\mu\text{g}/\text{m}^3$ may therefore be exceeded during Saharan dust intrusions. Such a value seems hard to meet in a region interested by the advection or resuspension of natural aerosols as mineral dust (Gobbi et al. 2007). Moreover, the measured PM10 in our study site may partly be due to nearby tram construction works and heavy traffic. This explains why, during weekends, the daily PM10 concentration decreases by 25 % compared with the average long-term concentration. The high Pb concentration can be explained by the fact that tetraethyl lead is still present in gasoline in Algeria.

During the study period implying simultaneous sampling of PM10 and PM2.5, the average concentration of PM10 was $105.2 \pm 53.7 \mu\text{g}/\text{m}^3$, while the average concentration of PM2.5 was $57.8 \pm 39.6 \mu\text{g}/\text{m}^3$.

Table 3 shows a summary of statistical data regarding PM concentrations, temperature, relative humidity and wind speed during the sampling period. Daily average concentrations of PM10 and PM2.5 vary between 10.9 to 193.2 $\mu\text{g}/\text{m}^3$ and 2.8 to 152.2 $\mu\text{g}/\text{m}^3$, respectively. The variation of PM

Table 2 PM standard limits and guideline values

	EU	WHO	Algerian limits
PM10	40 $\mu\text{g}/\text{m}^3$ annual mean	20 $\mu\text{g}/\text{m}^3$ annual mean	80 $\mu\text{g}/\text{m}^3$ annual mean
	50 $\mu\text{g}/\text{m}^3$ 24-h mean not be exceeded more than 7 times a calendar year	50 $\mu\text{g}/\text{m}^3$ 24-h mean	24-h mean not available
PM2.5	25 $\mu\text{g}/\text{m}^3$ annual mean (to be met in 2010)	10 $\mu\text{g}/\text{m}^3$ annual mean	–
	20 $\mu\text{g}/\text{m}^3$ annual mean (to be met in 2020)	25 $\mu\text{g}/\text{m}^3$ 24-h mean	

Table 3 Summary of meteorological parameters and PM concentrations during the study period (December 2011 to January 2013)

Parameter	Temperature	Humidity	Wind speed	Atmospheric pressure	PM10	PM2.5
Unit	°C	%	m/s	mm Hg	µg/m ³	µg/m ³
N	35	35	35	35	32	30
Average	13.7	36.8	1.6	863.7	122.6	57.8
SD	4.5	17.8	0.9	107.0	53.7	39.6
Minimum	5.9	13.4	0.4	688.3	10.9	2.8
Maximum	22.6	86.0	3.4	939.9	333.3	152.8

concentrations during the study period can be attributed to the variation of traffic density and meteorological parameters.

The formation of secondary particles formed in the air from precursor gases and favoured by high relative humidity is another source of particles in the study area that could affect the distributions of PM10 and PM2.5 (Kulshrestha et al. 2009).

PM10 and PM2.5 measured simultaneously exceed both standard limits set by the EU and the WHO guidelines (Table 2).

Factor analysis results

The factor analysis method was applied to identify likely sources of particulate emissions. Four major anthropogenic and natural emission sources were found accounting for 63.7 % of the variance (Table 4).

The first factor (F1) is strongly correlated with elements from anthropogenic sources such as Zn (0.76), Cu (0.57) and Ca (0.69). Thus, factor F1 was identified as anthropogenic emissions.

F2 is correlated with crustal elements such as Fe (0.86) and K (0.56). The factor was identified as soil dust.

F3 includes elements of marine source such as Mg (0.78) and Na (0.6). It was identified as a marine source factor.

F4 is strongly correlated with Pb (0.95). This factor was identified as an emission factor traffic.

Enrichment factors with respect to soil and seawater average concentrations

The enrichment factor element (FE) was originally developed to speculate on the origin of the elements in the atmosphere, precipitation or seawater (Goldberg 1972; Chester and Stoner 1973; Zoller et al. 1974).

The enrichment of an element in the atmospheric aerosol compared with a reference material can be measured through the enrichment factor (EF), which is defined as follows:

$$EF = (X/R)_{\text{sample}} / (X/R)_{\text{reference}}$$

where X =element studied, R =normalising factor

The choice of the reference element is determined by its stability and quantity (Guor-Cheng et al. 2006). For seawater, the most appropriate reference element is sodium, even if it can be associated with anthropogenic (waste incineration) or terrigenous sources in urban areas (Gordon 1980; Brewer

Table 4 Varimax rotated factor analysis with Kaiser normalisation for the traffic site

Element	Traffic site			
	F1	F2	F3	F4
Pb	0.08	0.01	0.01	0.95
Zn	0.76	-0.05	-0.03	0.06
Fe	-0.18	0.86	0.01	0.15
Mg	-0.15	-0.13	0.78	0.10
Ca	0.69	-0.01	0.4	0.07
K	0.36	0.56	-0.03	-0.20
Na	-0.20	-0.38	0.60	0.24
Cu	0.57	0.13	-0.26	-0.05
Var %	20.2	15.4	15	13.4
Cum %	20.2	35.6	50.6	63.7
Origin	Anthropogenic	Soil resuspension	Sea salt	Traffic

1975). In this study, Fe was the only element which can be related to crustal elements.

The comparison of the PM10 composition and the average composition of the soils may help to identify which trace elements are enriched in the aerosols with respect to the soils.

Enrichment factors in relation to soil and seawater average concentrations were calculated for all elements analysed in the total aerosol, namely K, Fe, Mg, Na, Ca, Cu, Zn and Pb (Fig. 2). Mg shows a low enrichment factor with regard to the sea concentration which indicates that it is of marine origin ($\log(EF) < 0.5$). The enrichment factor for K with regard to the soil concentration is also low ($\log(K) < 0.4$) indicating that it originates mainly from the soil.

Elements Pb, Cu and Zn are extremely enriched in the aerosols with a lesser extent with respect to Ca. These elements are derived from anthropogenic sources and cannot be associated neither with the soil nor with the sea.

Trajectories calculation and clustering

Backward air trajectories arriving at our observation sites were calculated using the web version of the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT-4) model. This model is a system for computing air mass trajectories and complex dispersion and deposition simulations (Draxler and Hess 1998). In this study, we calculated 3 days back-trajectories using the National Weather Service’s National Centres for Environmental Prediction (NCEP) model data available in NOAA’s Air Resources Laboratory (ARL) archives. The model output is a set of latitude–longitude coordinates of the air parcel estimated position for every hour.

A single trajectory can be insufficient to represent the transport history of a sampling volume even if it is small (Stohl et al. 2002). It has been demonstrated that clusters of back trajectories arriving at a specific location can serve as a surrogate of different synoptic circulation patterns (Borge et al. 2007).

Clustering is the division of data into groups of similar objects (Berkhin 2006). Cluster analysis is a multivariate statistical technique which involves splitting a data set into a

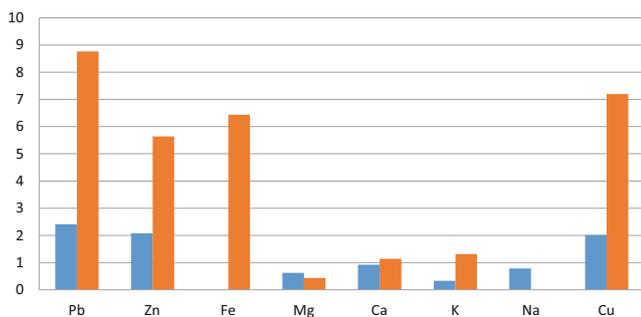


Fig. 2 Enrichment factors in relation with soil (■) and sea water (■)

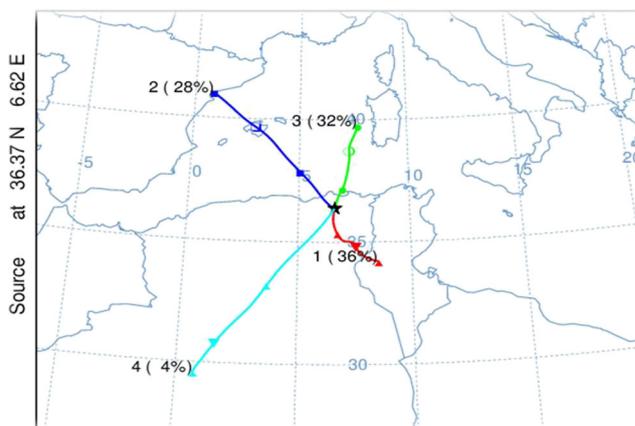


Fig. 3 Mean 3-day back-trajectory for the main trajectory clusters at Constantine

number of groups which need to be as homogeneous and as distinctly different from each other as possible.

Trajectories arriving at 12:00 GMT at Constantine, the midpoint of the sampling time used to collect air pollution data, were analysed. Each measured pollutant concentration was associated with its corresponding 72 h back-trajectory arriving (ending) at Zouaghi campus, Constantine. The choice of the 72-h back-trajectory length is supported by the lifetime of the different secondary species (Abdalmogith and Harrison 2005). For each 3 days (72 h) trajectory, 72 *x–y* coordinates (i.e. end points of the trajectory location at every hour) are utilised as input variables for the clustering algorithm. One hundred fourteen daily midday back-trajectories arriving at Constantine between 23 March 2011 and 22 November 2011 were assigned to four clusters using an automated K-means clustering algorithm.

The average back trajectory of each cluster is then calculated from its trajectory members. The average back trajectories for four clusters at Constantine are presented in Fig. 3.

The data set was split with relation to their origin (local, seawater and desert) and their distance (fast, slow) (Table 5).

Table 5 Average concentrations of PM10 and other metallic elements within each group (micrograms per cubic meter)

	PM10	Pb	Zn	Fe	Mg	Ca	K	Cu	Na
Group 4 (desert)	109.62	0.79	0.65	3.69	1.74	1.72	1.57	0.25	6.17
Group1 (local)	82.59	0.74	1.28	2.67	1.41	2.76	3.12	0.45	4.86
Continent	96.11	0.77	0.97	3.18	1.56	2.24	2.35	0.35	5.52
Group 2 (fast seawater)	70.20	0.88	1.24	2.53	1.97	2.32	5.05	0.36	6.50
Group 3 (slow sea water)	83.37	1.20	0.94	2.43	1.85	4.32	5.39	0.41	5.53
Sea water	76.79	1.04	1.10	2.48	1.91	3.32	5.22	0.39	6.02

Table 6 Matrix components after rotation (Varimax with Kaiser normalisation)

	PM2.5				PM10				
	F1	F2	F3	F4	F1	F2	F3	F4	
Pb	-0.08	0.86	-0.13	0.00	0.89	-0.09	-0.14	-0.07	Polluted soils
Cd	-0.04	0.91	-0.07	-0.04	0.91	-0.16	-0.13	-0.07	
Cu	0.60	-0.16	0.58	-0.15	-0.09	0.04	0.00	0.94	
Mg	0.80	0.03	-0.19	-0.03	-0.04	0.84	-0.13	-0.28	
Fe	-0.17	-0.13	0.84	0.11	-0.13	-0.07	0.87	-0.20	
Na	-0.05	-0.20	0.19	0.87	-0.13	0.67	-0.02	0.24	
Ca	0.19	0.42	-0.26	0.59	0.14	-0.19	0.65	0.45	
Zn	0.80	-0.12	0.22	0.14	0.87	-0.12	0.34	0.06	
Var %	22.38	20.62	16.83	15.29	27.44	21.05	16.88	14.09	
Cum%	22.38	42.99	59.82	75.12	27.44	48.49	65.37	79.47	
Possible sources		Exhaust	Crustal	Marine aerosol	Traffic	Marine aerosol	Crustal		

Factor loadings greater than ±0.5 are shown in bold

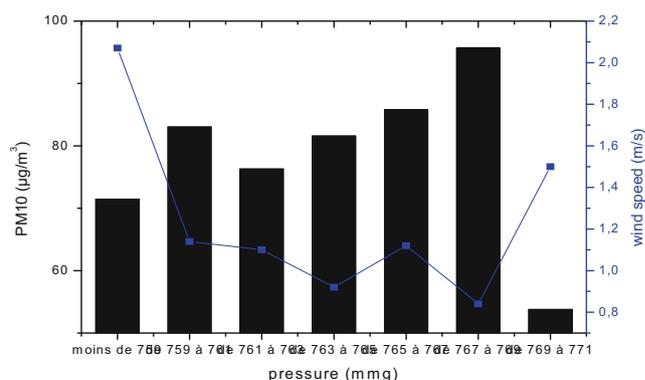


Fig. 4 The evolution of PM10 with atmospheric pressure and wind speed

The Saharan air mass intrusions are an important natural mineral particulate source and account for an increase (10–15 mg/m³) of the background PM10 levels for long periods (Alastuey et al. 2005). Such masses not only carry high levels of minerals of terrigenous origin such as Fe (Table 6), but they also travel past numerous salt marshes located 50 km south of the study site, such as Sebkhet Ezzemoul, Chott Tinsilt, Sebkhet Djendli, sweeping NaCl laden air and resulting in higher concentrations of Na.

Northerly air masses contain high concentrations of mineral elements such as Na (7 % higher than the average), Mg (4 % higher than the average), K (28 % higher than the average) and Ca (45 % higher than the average) which means that the Mediterranean basin is a major source of such elements. Soil dust and industry are not the only sources of K and Ca, respectively. These air masses also bring about anthropogenic elements such as Zn and Pb as they pass through the industrial area of Didouche Mourad and busy roads north of the city of Constantine.

Table 7 Average concentrations of metallic elements and PM2.5/PM10 ratios

	PM10 (µg/m ³)	PM2.5 (µg/m ³)	PM2.5/PM10
PM	122.63	57.83	0.47
Pb	0.79	0.44	0.79
Cu	0.51	0.22	0.60
Ca	0.10	0.01	0.49
Na	0.82	0.35	0.49
Cd	0.06	0.03	0.36
Zn	1.07	0.10	0.37
Mg	1.33	0.60	0.45
Fe	2.36	0.61	0.34

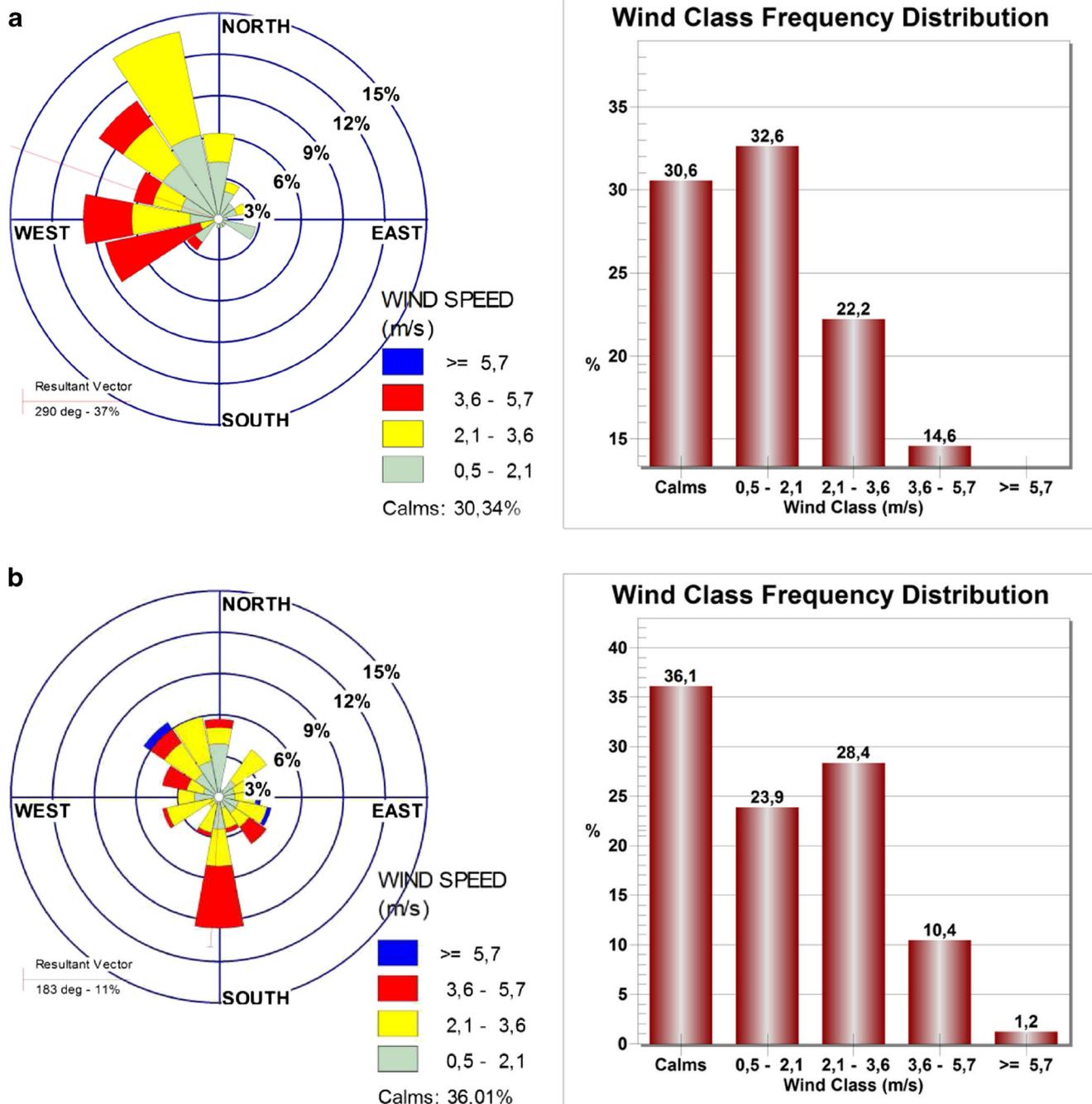


Fig. 5 Wind rose at Zouaghi, Constantine. **a** $PM_{2.5} < 20 \mu g/m^3$, **b** $PM_{2.5} > 20 \mu g/m^3$

The influence of atmospheric pressure and wind speed

Episodes of air pollution are often associated with anticyclonic weather systems (Unal et al. 2011). Generally, high-pressure systems can cause light winds and stable atmospheric conditions. To illustrate the influence of wind speed and atmospheric pressure on PM_{10} , we calculated the mean concentrations of PM_{10} for different ranges of air pressure and wind speed (Fig. 4). From this figure, we distinguish three

different episodes. The first episode is characterised by a low atmospheric pressure (< 759 mmHg), with an average

Table 8 Average frequencies of calm and westerly winds

PM ₁₀ concentration	$< 50 \mu g/m^3$	$> 50 \mu g/m^3$	$> 100 \mu g/m^3$
Frequency of calm winds	35.5	40.9	45.7
Frequency of westerly winds	1.5	3.5	3.7

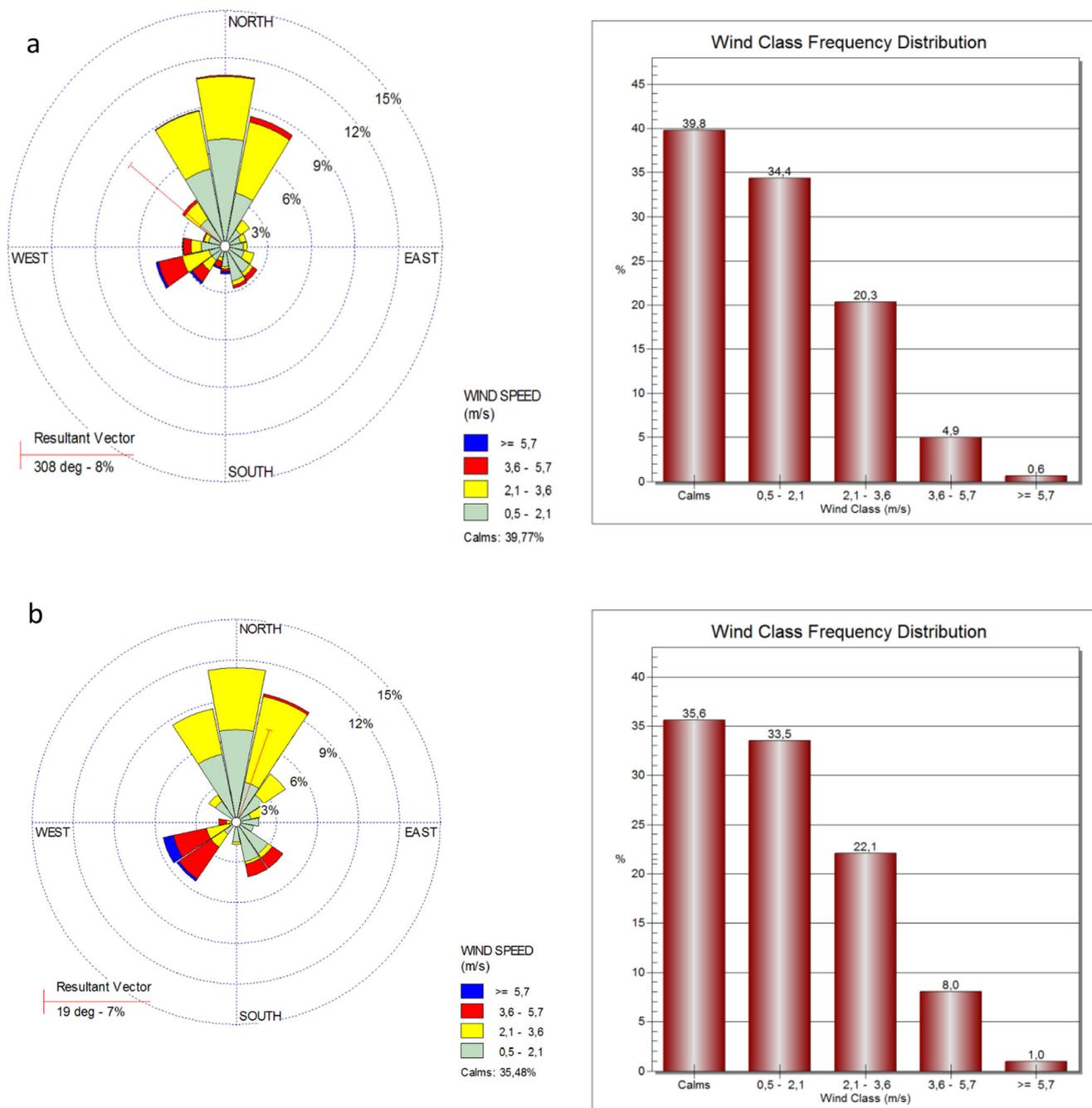


Fig. 6 Wind roses in Constantine. **a** All study period, **b** $PM_{10} < 50 \mu g/m^3$, **c** $PM_{10} > 50 \mu g/m^3$, $PM_{10} > 100 \mu g/m^3$

maximum wind speed of 2 m/s and an average PM_{10} concentration of $71 \mu g/m^3$ which is slightly lower than the average value. The second episode is characterised by a low concentration of PM_{10} ($53 \mu g/m^3$) and a high atmospheric pressure (769–771 mmHg) accompanied by moderate winds (1.5 m/s). In the third episode, high concentrations of PM_{10} ($95 \mu g/m^3$) coincide with anticyclonic weather conditions (high air pressure (767 to 769 mmHg) and weak winds (0.8 m/s).

Table 6 shows the results of factor analysis (FA) for $PM_{2.5}$ and PM_{10} with possible types of sources. Four factors were

extracted from the data of $PM_{2.5}$, which account for about 75.1 % of the total variance with factors F1 to F4 representing 22.4 %, 20.6 %, 16.8 % and 15.3 %, respectively.

F1 is defined by Mg and Zn and, to a lesser extent, by Cu. This factor is related to brake lining particles (Zn), tyre remains (Cu and Zn) (Rodriguez et al. 2004) and construction activities of the nearby tramway multimodal station (Mg) (Balasubramani et al. 2003). This factor is considered to be representative of road dust and brake lining.

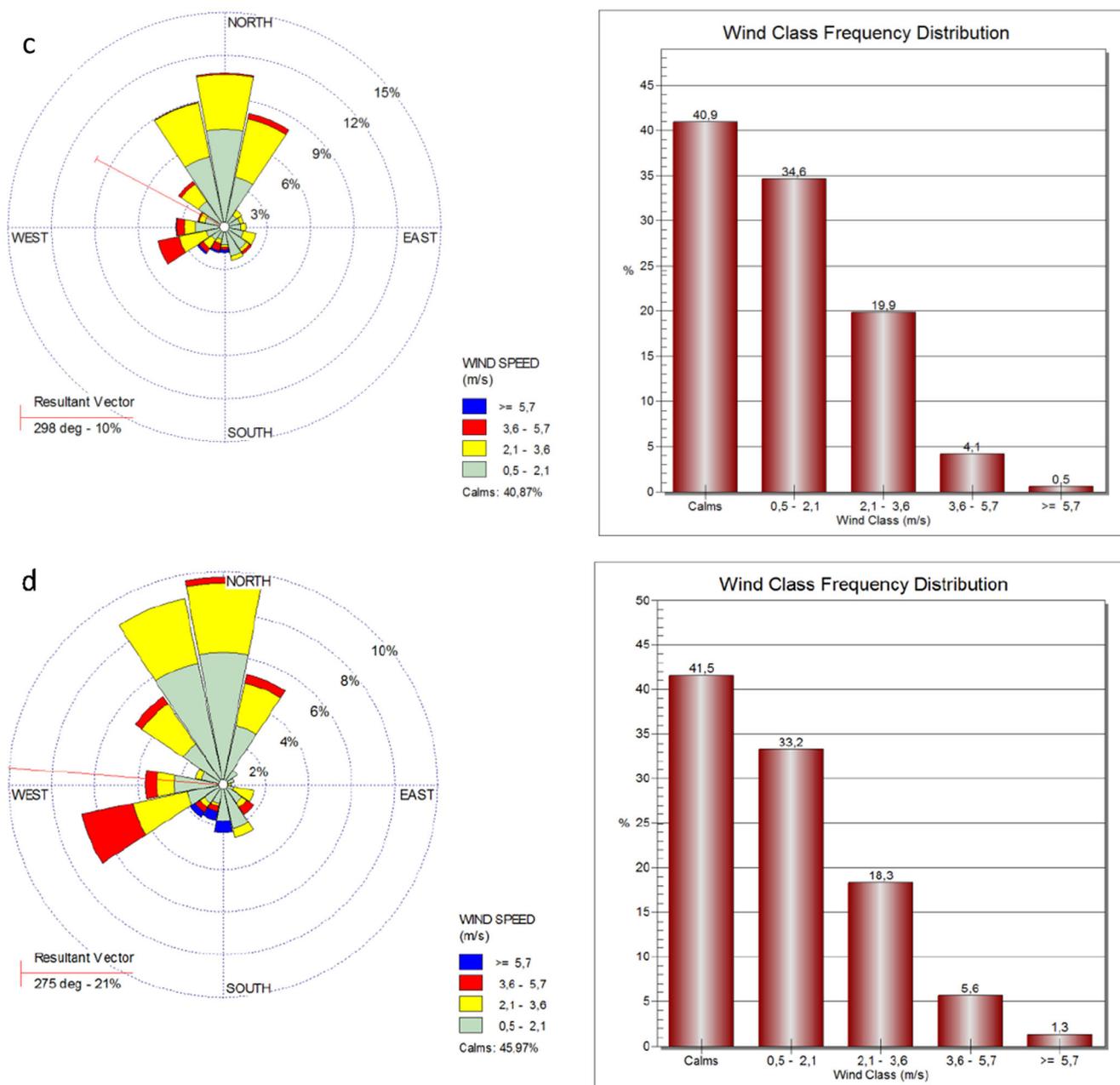


Fig. 6 (continued)

F2 represented combustion sources as it is associated with high loadings of Pb and Cd. Thus, the combustion factor includes vehicular emissions.

F3 reflects a high burden of Fe and a moderate Cu load. It is therefore assigned to a crustal origin.

Factor F4 was characteristic of sea salt shown by a high loading of Na whose origins can be the Mediterranean Sea at the north and the numerous salt marshes to the south.

Four factors were extracted from the PM₁₀ data that account for about 79.47 % of the total variance.

F1 is correlated with Pb, Zn and Cd. These compounds have been associated with vehicular releases, namely, exhaust gases of vehicles, the normal wear of lubricants, tires and mechanical parts.

F2 corresponds to high loads of Na and Mg. It is therefore associated with marine pollution.

F3 is associated with Fe and Ca. It represents the erosion of the earth's crust.

F4 contains a high load of Cu and a moderate load of Ca and can then be associated with the resuspension of road dust.

Table 9 Average PM concentrations in each group of backtrajectories

	Air mass type	PM _{2.5} μg/m ³	PM ₁₀	PM _{coarse}	Wind speed m/s	Humidity %	Temperature °C
Group 1	South-slow	100.7	215.4	137.8	1.9	46.7	15.1
Group 2	North-Slow	44.1	75.3	52.8	1.3	33.1	15.1
Group 3	North-west-fast	42.1	136.1	103.3	1.8	34.0	12.4
Group 4	North-fast	35.6	156.5	120.9	2.1	34.8	8.8

The PM_{2.5}/PM₁₀ ratio

The ratios of element concentrations in the fractions PM_{2.5}/PM₁₀ are shown in Table 7. PM_{2.5}/PM₁₀ elements with a ratio greater than 0.5 (mentioned in bold) are partitioned to the PM_{2.5} fraction rather than the PM₁₀ one. Samples with less than the detection limit were excluded in the calculation of PM_{2.5}/PM₁₀ ratios.

As shown in Table 7, the percentage of PM_{2.5} in PM₁₀ was 47.2 % which indicates that coarse particles originating from road dust resuspension and abrasion processes are the dominating fraction in particulate matter (Arkouli et al. 2010). The ratio is lower than the mean value of 0.72 observed by Artinano et al. (2004) in a study carried out in order to characterise PM₁₀ and PM_{2.5} fractions sampled at a representative urban site in Madrid. In another study, results obtained from the particulate matter (PM) data collected over the 1999–2008 period from representative urban traffic sites in the Madrid air basin show that PM₁₀ and PM_{2.5} mean annual values were 39.6±6.0 and 19.4±3.2 μg/m³, respectively (Salvadora et al. 2011). The corresponding ratio of 49 % is in agreement with the value found in our site. Gomiscek et al. (2004) reported the PM_{2.5} to PM₁₀ ratios calculated for various sites throughout Europe which vary from 40 % in Canada to 68 % in Germany and local Central and Eastern Europe, although comparison of the sites must be done carefully as they have distinct local features. The PM_{2.5} to PM₁₀

ratio found in our study is in the lower side of the bandwidth since 14 out of 15 European sites show ratios exceeding 49 %. Finally, the annual mean PM_{2.5}:PM₁₀ ratio in the industrial city of Bursa, the fourth largest city in Turkey, was 0.64 between May 2007 and April 2008 as average PM_{2.5} and PM₁₀ mass concentrations over the year were 53 and 83 μg/m³, respectively (Kendall et al. 2011). Therefore, a less significant proportion of PM₁₀—47.2 %—is in the respirable fine fraction (PM_{2.5}) in our site.

The results show that the terrestrial and marine elements (Fe, Ca, Mg, Na) are partitioned mostly to the PM₁₀ fraction. Zn and Cd are more abundant in the PM₁₀ than the PM_{2.5} because they are emitted more by a mechanical process (tire wear and mechanical parts) than by the combustion process. Lead and copper are partitioned to the PM_{2.5} fraction. They are often in the form of fine particles, which means that fuel combustion is the largest source of these elements.

Wind direction and speed

Figure 5 shows that winds from the same direction, which extends through National road 79, play a key role in the concentration of PM_{2.5} in our site. They accumulate a large number of pollutants emitted by vehicles before reaching the site. This explains why PM_{2.5} levels greater than 20 μg/m³ were recorded when the wind comes from the South, SSO, while in the days when the PM_{2.5} concentration is low (<20 μg/m³), no wind from the S, SSO is recorded (Fig. 5a).

We also note that the frequency of calm winds is higher in highly polluted days (PM_{2.5}>20 μg/m³) than the days with low pollution (PM_{2.5}<20 μg/m³), because calm conditions cause pollution stagnation at the emission point.

The strongest winds (>3.6 m/s) are often from SW (Sahara region), which mean that these air masses are often loaded with Saharan dust.

The frequency of westerly winds, the sector where the tramway construction site is located, is proportional to the concentration of PM₁₀. More frequent westerly winds lead to higher PM₁₀ levels (Table 8; Fig. 6).

Table 9 shows the average concentrations of PM₁₀, PM_{2.5} and coarse PM_{10-2.5} at Zouaghi, Constantine, during the occurrence of each transport path and the corresponding weather during the sampling period.

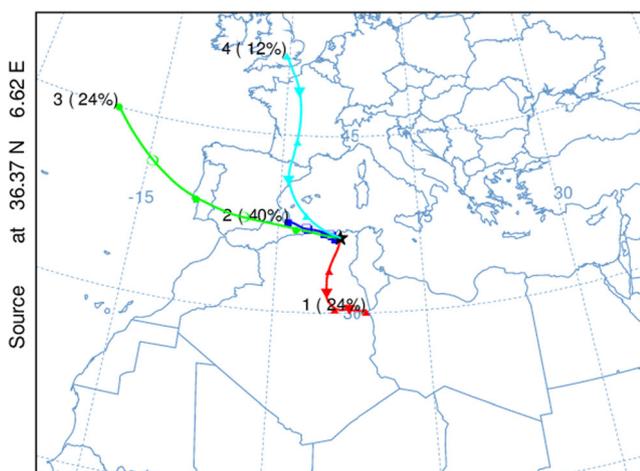


Fig. 7 Geometric centre of back-trajectories at 750 m above sea level. The percentage of retro-trajectories in each group is also included

When PM concentrations are analysed according to the type of group air mass, clear differences become apparent.

It can be found from Fig. 7 that groups 1 and 2 were associated with a short distance transport, compared with groups 3 and 4.

The group of slow North type is the most common and least polluted, unlike air masses coming from the south (group 1) which are significantly more polluted. These air masses are associated with higher concentrations of PM_{2.5}, PM₁₀ and coarse PM. High concentrations of coarse PM should be associated with long-range transport of Saharan particles. The occurrence of Group 2 was associated with low wind speeds and a low concentration of coarse PM, unlike other groups that are characterised by a higher wind speed, accompanied by a high load of coarse PM, as transport of this type of particles is strongly related to the wind speed. As this speed increases, the transport capacity is enhanced. The high concentration of PM_{2.5} in the slow southeast group type can be associated with the transport of pollutants from urban areas (traffic, industrial, fuel combustion), knowing that there are two major urban centres, the new city 8 km to the south and the city of El Khroub 10 km southeast.

Conclusions

PM₁₀ concentrations were measured at a sampling traffic site situated at Zouaghi, Constantine, between 23 March 2011 and 22 November 2011. The results presented in this work allow us to conclude that PM₁₀ and PM_{2.5} concentrations are excessive in light of the WHO and the EU standards. The latter seem hardly feasible in view of the contribution of natural aerosols to ambient PM levels. The average daily concentration of PM₁₀ (80.42 $\mu\text{g}/\text{m}^3$) was observed for the period extending from 23 March 2011 to 22 November 2011. During the study period implying simultaneous sampling of PM₁₀ and PM_{2.5}, the average concentration of PM₁₀ was 105.2 $\mu\text{g}/\text{m}^3$, while the average concentration of PM_{2.5} was 57.8 $\mu\text{g}/\text{m}^3$. Therefore, the PM_{2.5}/PM₁₀ ratio was equal to 0.47. Sources of coarse particles are the most important due to the significant contribution of resuspended coarse particles and Saharan dust intrusions.

In this work, some statistical techniques have been successfully used to identify and characterise PM₁₀ and PM_{2.5} sources. The application of Varimax rotated factor analysis, a multivariate technique, has allowed us to qualitatively identify anthropogenic, soil resuspension, sea salt and traffic as the main PM₁₀ sources, at a traffic site in Constantine (Algeria).

Enrichment factors in relation to soil and seawater average concentrations indicate that Mg is of marine origin and K

originates mainly from the soil while Pb, Cu and Zn are derived from anthropogenic sources.

To identify external sources and their geographical origin, air mass back-trajectories have been calculated with the HYSPLIT 4 model. Results point at the Sahara desert as a major source of PM₁₀ and Fe. The contribution to Na results from long-range transport of air masses originating from the North (Mediterranean Sea) or from the South (salt marshes).

On the other hand, salt marshes located south of the study station are identified as the path followed by dust plumes originated in the desert region. They have been derived for Na.

The Mediterranean Sea has been identified as the major source of Na, Mg, K and Ca. Iron is of crustal origin, either from nearby sources such as the soil surrounding the site or distant sources such as the Sahara to the south. Calcium and potassium have also an anthropogenic origin. Anthropogenic sources are related to the construction works of the tram on the other side of the road along the measurement site. Zn, Cu and Pb are derived from anthropogenic sources: traffic and industry. Traffic is the major source of the high Pb levels observed since gasoline still contains lead additives in Algeria.

References

- Abdalmogith SS, Harrison MR (2005) The use of trajectory cluster analysis to examine the long-range transport of secondary inorganic aerosol in the UK. *Atmos Environ* 39(35):6686–6695. doi:10.1016/j.atmosenv.2005.07.059
- Alastuey A, Querol X, Castillo S, Avila A, Estarellas C, Torres C, Exposito F, Garcia O, Diaz JP, Van dingenen R et al (2005) Characterisation of TSP and PM_{2.5} at Izaña and Sta. Cruz de Tenerife (Canary Islands, Spain) during a Saharan dust episode. *Atmos Environ* 39(26):4715–4728. doi:10.1016/j.atmosenv.2005.04.018
- Ali-Khodja H, Belaala A, Demmane-Debbih W, habbas B, Boumagoura N (2008) Air quality and deposition of trace elements in Didouche Mourad, Algeria. *Environ Monit Assess* 138:219–231. doi:10.1007/s10661-007-9792-1
- Arkouli M, Ulke AG, Endlicher W, Baumbach G, Schultz E, Vogt U, Müller M, Dawidowski L, Faggi A, Wolf-Benning U, Scheffknecht G (2010) Distribution and temporal behavior of particulate matter over the urban area of Buenos Aires. *Atmos pollut res* 1:1–8. doi:10.5094/APR.2010.001
- B. Artinano B, Salvador P, Alonso DG, Querol X, Alastuey A (2004) Influence of traffic on the PM₁₀ and PM_{2.5} urban aerosol fractions in Madrid (Spain). *Sci Total Environ* 334–335: 111– 123. doi: 10.1016/j.scitotenv.2004.04.032
- Balasubramani R, Qian WB, Decesari S, Facchini MC, Fuzzi S (2003) Comprehensive characterization of PM_{2.5} aerosols in Singapore. *J Geophys Res* 108(D16):AAC7.1–AAC7. doi:10.1029/2002JD002517
- Berkhin P (2006) A Survey of clustering data mining techniques. In: Kogan J, Nicholas C, Teboulle M (eds) *Grouping multidimensional data*. Springer, Berlin Heidelberg, pp 25–71
- Borge R, Lumbreras J, Vardoulakis S, Kassomenos P, Rodríguez E (2007) Analysis of long-range transport influences on urban PM₁₀

- using two-stage atmospheric trajectory clusters. *Atmos Environ* 41(21):4434–4450. doi:10.1016/j.atmosenv.2007.01.053
- Bouchlaghem K, Nsom B (2012) Effect of atmospheric pollutants on the air quality in Tunisia. *Sci World J*. doi:10.1100/2012/863528
- Bouchlaghem K, Nsom B, Latrache N, Haj Kacem H (2009) Impact of Saharan dust on PM10 concentration in the Mediterranean Tunisian coasts. *Atmos Res* 92:531–539. doi:10.1016/j.atmosres.2009.02.009
- Brewer P (1975) Minor elements in sea water. In: Riley JP, Chester R (eds) *Chemical oceanography*, vol 1, 2nd edn. Academic Press, New York, pp 416–496
- Chen R, Kan H, Chen B, Huang W, Bai Z, Song G, Pan G (2012) Association of particulate air pollution with daily mortality: the China Air Pollution and Health Effects study. *Am J Epidemiol* 175(11):1173–1181. doi:10.1093/aje/kwr425
- Chester J, Stoner J (1973) Pb in particulates from the lower atmosphere of the eastern Atlantic. *Nature* 245:27–28. doi:10.1038/245027b0
- Dagnelie P (1975) *Analyse statistique à plusieurs variables*. Presses Agronomiques, Gembloux
- Draxler R, Hess G (1998) An overview of the Hysplit_4 modelling system for trajectories dispersion, and deposition. *Aust Meteorol Mag* 47:295–308
- Eliasson I, Jonsson P, Holmer B (2009) Diurnal and intra-urban particle concentrations in relation to windspeed and stability during the dry season in three African cities. *Environ Monit Assess* 154:309–324. doi:10.1007/s10661-008-0399-y
- Gobbi GP, Barnaba F, Ammannato L (2007) Estimating the impact of Saharan dust on the year 2001 PM10 record of Rome, Italy. *Atmos Environ* 41:261–275. doi:10.1016/j.atmosenv.2006.08.036
- Goldberg E (1972) Baseline studies of heavy metal, halogenated hydrocarbon, and petroleum hydrocarbon pollutants in the marine environment and research recommendations, deliberations of the International Decade of Ocean Exploration (IDOE) Baseline Conference. National Science Foundation, New York
- Gomiscek B, Hauck H, Stopper S, Preining O (2004) Spatial and temporal variations of PM1, PM2.5, PM10 and particle number concentration during the AUPHEP-project. *Atmos Environ* 38:3917–3934. doi:10.1016/j.atmosenv.2004.03.056
- Gordon GE (1980) Receptor models. *Envir Sci Technol* 14(7):792–800. doi:10.1021/es60167a006
- Guor-Cheng F, Yuh-Shen W, Shih-Yu C, Shih-Han H, Rau JY (2006) Size distributions of ambient air particles and enrichment factor analyses of metallic elements at Taichung Harbor near the Taiwan Strait. *Atmos Res* 81(4):320–333. doi:10.1016/j.atmosres.2006.01.007
- Han X, Naeher LP (2006) A review of traffic-related air pollution exposure assessment studies in the developing world. *Environ Int* 32:106–120. doi:10.1016/j.envint.2005.05.020
- Handler M, Puls C, Zbiral J, Marr I, Puxbaum H, Limbeck A (2008) Size and composition of particulate emissions from motor vehicles in the Kaisermühlentunnel, Vienna. *Atmos Environ* 42(9):2173–2186. doi:10.1016/j.atmosenv.2007.11.054
- Henschel C, Chan G (2013) Pollution in Europe—HRAPIE project. WHO Regional Office for Europe. http://www.euro.who.int/__data/assets/pdf_file/0017/234026/e96933.pdf
- Karakatsani A, Analitis A, Perifanou D, Ayres JG, Harrison RM, Kotronarou A, Kavouras IG, Pekkanen J, Hämeri K, Kos GPA, de Hartog JJ, Hoek G, Katsouyanni K (2012) Particulate matter air pollution and respiratory symptoms in individuals having either asthma or chronic obstructive pulmonary disease: a European multicentre panel study. *Environ Health* 11:75. doi:10.1186/1476-069X-11-75
- Kendall M, Pala US, Gucer S (2011) Airborne particulate matter (PM2.5 and PM10) and associated metals in urban Turkey. *Air Qual Atmos Health* 4:235–242. doi:10.1007/s11869-010-0129-9
- Kulshrestha A, Gursumeeran S, Jamson M, Ajay T (2009) Metal concentration of PM2.5 and PM10 particles and seasonal variations in urban and rural environment of Agra, India. *Sci Total Environ* 407:6196–6204. doi:10.1016/j.scitotenv.2009.08.050
- Kuvarega AT, Taru P (2008) Ambient dust speciation and metal content variation in TSP, PM 10 and PM 2.5 in urban atmospheric air of Harare (Zimbabwe). *Environ Monit Assess* 144:1–14. doi:10.1007/s10661-008-0436-x
- Laïd Y, Atek M, Oudjehane R, Filleul L, Baough L, Zidouni N, Boughedaoui M, Tessier J-F (2006) Impact sanitaire de la pollution de l'air par les PM10 dans une ville du sud: le cas d'Alger. *Int J Tuberc Lung Dis* 10(12):1406–1411
- Lim SS, Vos TD, Flaxman AD, Danaei G, Shibuya K et al (2012) A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease study 2010. *Lancet* 380:2224–2260. doi:10.1016/S0140-6736(12)61766-8
- Liousse C, Assamoi E, Criqui P, Granier C, Rosset R (2014) Explosive growth in African combustion emissions from 2005 to 2030. *Environ Res Lett*. doi:10.1088/1748-9326/9/3/035003
- Meloni D, di Sarra A, Biavati G, DeLuisi JJ, Monteleone F, Pace G, Piacentino S, Sferlazzo DM (2007) Seasonal behaviour of Saharan dust events at the Mediterranean island of Lampedusa in the period 1999–2005. *Atmos Environ* 41:3041–3056. doi:10.1016/j.atmosenv.2006.12.001
- Morman SA, Plumlee GS (2014) Dust and human health. In: Knippertz P, Stuu J-BW (eds) *Mineral dust: a key player in the earth system*. Springer, New York, pp 385–409
- Pastuszka JS, Rogula-Kozłowska W, Zajusz-Zubek E (2010) Characterization of PM10 and PM2.5 and associated heavy metals at the crossroads and urban background site in Zabrze, Upper Silesia, Poland, during the smog episodes. *Environ Monit Assess* 168:613–627. doi:10.1007/s10661-009-1138-8
- Pateraki S, Asimakopoulos DN, Flocas HA, Maggos T, Vasilakos C (2012) The role of meteorology on different sized aerosol fractions (PM10, PM2.5, PM2.5–10). *Sci Total Environ* 419:124–135. doi:10.1016/j.scitotenv.2011.12.064
- Petkova EP, Jack DW, Volavka-Close NH, Kinney PL (2013) Particulate matter pollution in African cities. *Air Qual Atmos Health* 6(3):603–614. doi:10.1007/s10661-007-9792-1
- Puett RC, Hart JE, Yanosky JD, Spiegelman D, Wang M, Fisher JA, Hong B, Laden F (2014) Particulate matter air pollution exposure, distance to road, and incident lung cancer in the nurse's health study cohort. *Environ Health Perspect* 122(9):923–933. doi:10.1289/ehp.1307490
- Querol X et al (2004) Speciation and origin of PM10 and PM2.5 in selected European cities. *Atmos Environ* 38:6547–6555. doi:10.1016/j.atmosenv.2004.08.037
- Rodriguez S, Querol X, Alastueya A, Viana MM, Alarcon M, Mantilla E, Ruiz CR (2004) Comparative p M10–PM2.5 source contribution study at rural, urban and industrial sites during PM episodes in Eastern Spain. *Sci Total Environ* 328:95–113. doi:10.1016/S0048-9697(03)00411-X
- Rodriguez S, Querol X, Alastueya A, de la Rosa J (2007) Atmospheric particulate matter and air quality in the Mediterranean: a review. *Environ Chem Lett* 5:1–7. doi:10.1007/s10311-006-0071-0
- Rückerl R, Schneider A, Breitner S, Cyrys J, Peters A (2011) Health effects of particulate air pollution: a review of epidemiological evidence. *Inhal Toxicol* 23(10):555–592. doi:10.3109/08958378.2011.593587
- Salvadora P, Artinano B, Vianab MM, Querol X, Alastueyb A, Gonzalez-Fernandez I, Alonso R (2011) Spatial and temporal variations in PM10 and PM2.5 across Madrid metropolitan area in 1999–2008. *Procedia Environ Sci* 4:198–208. doi:10.1016/j.proenv.2011.03.024

- Stohl A, Eckhardt S, Forster C, James P, Spichtinger N, Seibert P (2002) A replacement for simple back trajectory calculations in the interpretation of atmospheric trace substance measurements. *Atmos Environ* 36(29):4635–4648. doi:10.1016/S1352-2310(02)00416-8
- Terrouche A, Ali-Khodja H, Talbi M, Bencharif-Madani F, Charron A, Derradji A. Roadside PM10 and associated metals in Constantine, Algeria. Accepted for publication in *Int J Environ Stud*.
- Unal YS, Toros H, Deniz A, Incecik S (2011) Influence of meteorological factors and emission sources on spatial and temporal variations of PM10 concentrations in Istanbul metropolitan area. *Atmos Environ* 45(31):5504–5513. doi:10.1016/j.atmosenv.2011.06.039
- Val S, Liousse C, Thierno Doumbia EH, Galy-Lacaux C, Cachier H, Marchand N, Badel A, Gardrat E, Sylvestre A, Baeza-Squiban A (2013) Physico-chemical characterization of African urban aerosols (Bamako in Mali and Dakar in Senegal) and their toxic effects in human bronchial epithelial cells: description of a worrying situation. *Part Fibre Toxicol* 10:10. doi:10.1186/1743-8977-10-10
- WHO (2006) Health risks of particulate matter from long range transboundary air pollution. WHO/Convention Task Force. European Centre for Environment and Health, Bonn Office. http://www.euro.who.int/data/assets/pdf_file/0006/78657/E88189.pdf
- WHO (2013) Review of evidence on health aspects of air pollution-REVHIIHAAP. WHO Regional Office for Europe.
- Zoller WH, Gladney ES, Duce RA (1974) Atmospheric concentrations and sources of trace metals at the South Pole. *Science* 183(4121): 198–2010