

# Influence of Saharan Dust Transport Events on PM<sub>2.5</sub> Concentrations and Composition over Athens

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**Abstract** The evaluation of the contribution of natural sources to PM<sub>10</sub> and PM<sub>2.5</sub> concentrations is a priority especially for the countries of European south strongly influenced by Saharan dust transport events. Daily PM<sub>2.5</sub> concentrations and composition were monitored at an urban site at 14 m above ground level, at the National Technical University of Athens campus from February to December 2010. The typical dust constituents Si, Al, Fe, K, Ca, Mg, and Ti were determined by wavelength dispersive X-ray fluorescence spectrometry (WDXRF). Sulfur, a tracer of anthropogenic origin and major constituent of PM<sub>2.5</sub>, was determined by both WDXRF and ionic chromatography. The contribution of dust and sulfates in PM<sub>2.5</sub> was calculated from the analytical determinations. An annual mean of 20 µg/m<sup>3</sup>

was calculated from the mean daily PM<sub>2.5</sub> concentrations data. Twenty-two per cent of daily concentrations of PM<sub>2.5</sub> reached or exceeded the EU annual target concentration of 25 µg/m<sup>3</sup>. The exceedances occurred during 13 short periods of 1–4 days. Back-trajectory analysis was performed for these periods in order to identify the air masses origin. From these periods, ten periods were associated to Saharan dust transport events. The most intense dust transport event occurred between February 17th and 20th and was responsible for the highest recorded PM<sub>2.5</sub> concentration of 100 µg/m<sup>3</sup> where the dust contribution in PM<sub>2.5</sub> reached 96 %. The other dust transport events were less intense and corresponded to less pronounced enhancements of PM<sub>2.5</sub> concentrations, and their contribution ranged from 15 to 39 % in PM<sub>2.5</sub> concentrations. Air masses originated from northwest Africa while the influence of central Sahara was quite smaller.

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## 1 Introduction

Particulate matter and especially PM<sub>10</sub> and PM<sub>2.5</sub> levels are monitored in ambient air quality networks because of their potential impact on health and climate. The presence of particulate matter in urban environments has several undesirable health effects, and new limits were set for European countries by the

directive for the quality of air in Europe (Directive 2008/50/EP): Daily and annual  $\text{PM}_{10}$  limits were set in 40 and  $50 \mu\text{g}/\text{m}^3$ , respectively, since 1 Jan 2005. The latter limit value cannot be exceeded by more than 35 times per calendar year. For  $\text{PM}_{2.5}$ , the annual limit value was set in  $25 \mu\text{g}/\text{m}^3$  (target value since 1 Jan 2010 and limit value since 1 Jan 2015). Furthermore, this directive also sets a  $\text{PM}_{2.5}$  exposure reduction obligation that applies to mean urban background levels.

The Mediterranean region is strongly affected by dust transport from Sahara with maxima occurring during spring and autumn mainly in the eastern basin (Engelstaedter and Washington 2007; Fotiadi et al. 2006; Gerasopoulos et al. 2007; Israelevich et al. 2002; Querol et al. 2009; Rodriguez et al. 2001; Sciare et al. 2003). Dust is transported into PBL, also often advected above the planetary boundary layer (PBL), and lifted up to 2–10 km (Gobbi et al. 2007; Kalivitis et al. 2007; Koukouli et al. 2010; Papayannis et al. 2005, 2008), and diffuses into PBL by subsidence, fumigation, and sedimentation. The long-range transport of Saharan dust is responsible for a high background of particulate matter in the area mainly for the southern European countries (Blanco et al. 2003; Gerasopoulos et al. 2006; Gobbi et al. 2007; Moreno et al. 2005; Pappalardo et al. 2010; Querol et al. 2004; Rodriguez et al. 2001; Samoli et al. 2011; Viana et al. 2007). Desert dust represents the main natural contribution in the particulate matter suspended in the atmosphere and is mainly associated with the coarse fraction, and its contribution could be determinant at exceeding the legal limits. The evaluation of dust contribution in  $\text{PM}_{10}$  in southern European countries urban environments has shown that a significant part of limit exceedances were associated with Saharan dust transport (Borge et al. 2007). For example, 34 % of the exceedance days occurred during African dust intrusions for sampling stations in Spain (Moreno et al. 2005); 25 % of the Saharan advection days led to a  $\text{PM}_{10}$  increase  $>30 \mu\text{g}/\text{m}^3$  in Rome (Gobbi et al. 2007), and Mitsakou et al. (2008) have reported that, for Greece, dust transport may contribute by more than 20 % to the annual number of exceedances— $\text{PM}_{10}$  concentrations  $>$  EU limits—depending on the specific monitoring location. Moreover, African dust outbreaks over Europe imply important health effects (Perez et al. 2008; Polymenakou et al. 2008; Samoli et al. 2011).

A heavily charged atmosphere by locally emitted and long-range transported particulate matter together with the absence of precipitation from spring to late autumn are situations frequently met in Athens. Monitoring of concentration levels and quantitative estimations on particle composition together with their source identification are necessary in order to acquire data for the estimation of their impact and to answer if the above limits are realistic for the large urban areas in Greece. For the above reasons, the identification of the contribution of the long-ranged transported aerosol particles and the quantification of their contribution to urban  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  levels are necessary.

Results from systematic monitoring of  $\text{PM}_{10}$  concentrations and  $\text{PM}_{10}$  composition determinations are the most frequent in literature for Greek urban environments and have shown that the EU limit of  $50 \mu\text{g}/\text{m}^3$  is very often exceeded (Chaloulakou et al. 2003; Gerasopoulos et al. 2006; Grivas et al. 2008; Terzi et al. 2010; Theodosi et al. 2011). Results on  $\text{PM}_{2.5}$  concentration levels and composition are few and recent and have also shown frequent exceedances of the annual target value of  $25 \mu\text{g}/\text{m}^3$  (Karageorgos and Rapsomanikis 2007; Lazaridis et al. 2008; Manoli et al. 2002; Sillanpaa et al. 2006; Theodosi et al. 2011; Vassilakos et al. 2005). Quantitative data on the concentrations and composition of  $\text{PM}_{2.5}$  based on longer time series are necessary in order to clarify and complete the picture, and the present work may contribute to this effort.

The present paper reports daily  $\text{PM}_{2.5}$  concentrations for Athens from monitoring at an urban site at 14 m above ground level, at the National Technical University of Athens campus from February to December 2010. The main purpose of the present work was to evaluate the contribution of Saharan dust episodes in the observed  $\text{PM}_{2.5}$  daily concentrations. Experimental results of  $\text{PM}_{2.5}$  sampling on polytetrafluoroethylene (PTFE) membranes by using TCR TECORA sampler are presented and discussed. Chemical analysis was performed in order to determine the elements characteristic of the dust. Sulfur was chosen to be shown in comparison to the elements characteristic of the dust, mainly because sulfur is a major element-constituent of  $\text{PM}_{2.5}$  (Sillanpaa et al. 2006; Theodosi et al. 2011) having both local and regional sources. The comparison of the temporal variability of the elemental concentrations of the dust constituents to that of sulfur/sulfates in the present

study was also chosen in order to show the contrast between the variability of the concentrations of elements characterized by sporadic and abrupt increases of their concentrations (during dust transport episodes) to that corresponding to a more homogenous concentration temporal variability as is the case of sulfur/sulfates. The temporal variability of  $PM_{2.5}$  concentrations is discussed focusing on maxima and limit exceedances. The temporal variability of crustal origin elements and sulfur is also presented and associated with that of  $PM_{2.5}$  concentrations. For the periods where maxima were observed for  $PM_{2.5}$  concentrations, elemental concentrations together with back-trajectory analysis were considered simultaneously in order to explain the exceedances observed. The percentage of the dust contribution and the contribution of sulfates in the observed  $PM_{2.5}$  concentrations was deduced from the experimental results. Finally, the source regions of the dust transported over Athens are also discussed.

## 2 Materials and Methods

### 2.1 $PM_{2.5}$ Sampling, Concentrations, and Elemental Composition Determinations

Twenty-four-hour aerosol samples were collected at the top of the building of the School of Mining and Metallurgical Engineering at the National Technical University of Athens campus at 14 m above ground level. A hundred sixteen samples corresponding to about 10–15 samples per month have been collected from February to December 2010. The sampling point is fully exposed to wind and free all around of other obstacles (Remoundaki et al. 2011).  $PM_{2.5}$  sampling was carried out using a TCR TECORA (Sentinel PM) operating at 38.33 l/min, constructed, and then calibrated in order to comply with European Standard EN14907 for standard sampling of  $PM_{2.5}$ .

The sampling device operates with autonomy of 16 samples charged in a charging cassette by programming the sampling span and duration. Aerosol samples were collected on PTFE membranes ( $PM_{2.5}$  air monitoring membranes, Whatman).

Sampling material and filter-keeping Petri dishes were pretreated by soaking in dilute nitric acid solution and thorough rinsing by ultra-pure water (18 M $\Omega$ /cm) and dried under the laminar flow hood of the laboratory. In order to determine  $PM_{2.5}$  concentrations, the

membranes were weighted before and after sampling according to the procedure described in EN12341 (Annex C) using a Mettler Toledo MS105 with a resolution of 10  $\mu$ g in the air conditioned weighing room of the laboratory.

The pre-weighted membranes were charged to the filter supports and sampler cassette under the laminar flow hood. Filter blanks and blank field samples were also prepared and analyzed together with samples. The elemental composition determinations have been carried out by WDXRF (Thermo ARL Advant XP, sequential XRF) with Rh X-ray tube at 30 kV, 30 mA. Single element standards purchased from Micromatter have been used for calibration for each element. Two NIST standards SRM 2783 have been also used as calibration points and for calibration verification. The elements Si, Al, Fe, K, Ca, Mg, Ti, and S have been determined. The detection limits were calculated to be Al, 100; Si, 300; Fe, 300; Ca, 40; S, 20; Ti, 40; K, 20; and Mg, 100 ng. The estimated precision of the method was estimated to be <5 %.

Following WDXRF analysis, one half of the filter was extracted with 10 mL of ultrapure water for ion chromatography analysis, and acid digestion was performed to the second part for trace metals analysis. In this paper, only results on sulfates are presented, as mass closure results for  $PM_{2.5}$  composition including the ionic content, organic and elemental carbon, and trace metals are in progress. The extracted samples were kept refrigerated and analyzed by ion chromatography within 1 week after extraction. Ions were determined by a Metrohm 732 IC Separation Center connected to a 732 IC conductivity detector and 753 Suppressor Module for anions determination. A Metrosep Anion Dual 2 column was used for anions determinations (eluent, 2 mmol/L  $NaHCO_3$ , 1.3 mmol/L  $Na_2CO_3$  with chemical suppression; flow, 0.8 ml/min). The calculated detection limit for sulfates was 14  $\mu$ g/L. The determinations of S performed by WDXRF and IC can be compared on the basis of a linear regression which resulted to the following equation,  $S_{(IC)} = 1.0145 \times S_{(WDXRF)} - 18.8$ , ( $r^2 = 0.97$ ) meaning a very satisfactory agreement between both methods and the presence of the totality of sulfur as sulfates.

### 2.2 Air Mass Trajectories

Air mass backward-trajectories were calculated for the Athens site using the hybrid single-particle Lagrangian

integrated trajectory model to gather information about the origin of the observed aerosols corresponding to the period under study. The calculations were made for the arrival heights of 1, 2, and 3.5 km asl over Athens, Greece.

### 3 Results and Discussion

#### 3.1 PM<sub>2.5</sub> Concentrations

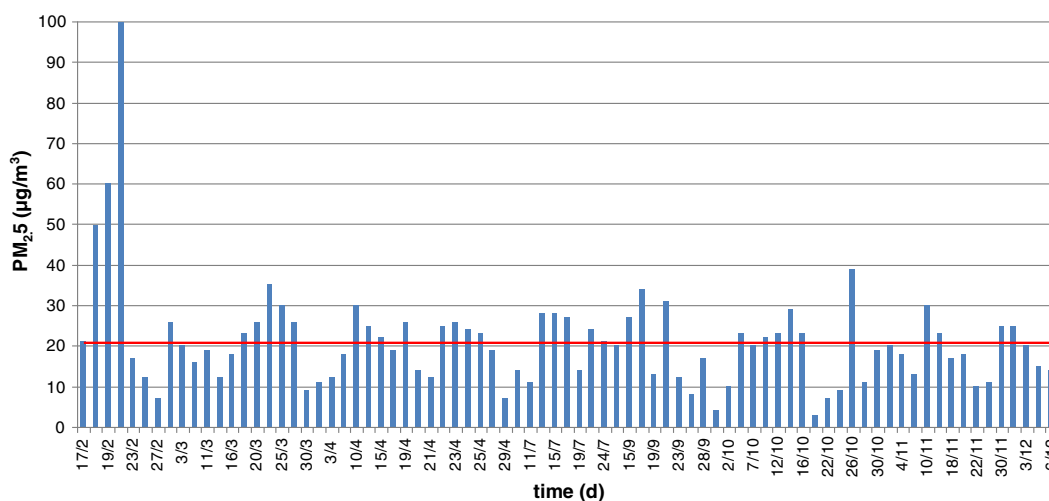
PM<sub>2.5</sub> concentrations arithmetic mean and median were calculated to be 21(±11) and 20  $\mu\text{g}/\text{m}^3$ , respectively, corresponding to 24-h samples collected between February and December 2010 (minimum and maximum values were equal to 3 and 100  $\mu\text{g}/\text{m}^3$ , respectively). Other studies report for Athens similar or higher values—21  $\mu\text{g}/\text{m}^3$ , for a suburban sampling point and year 2003 (Vassilakos et al. 2005), 23.5 and 29.4  $\mu\text{g}/\text{m}^3$  for downtown sampling points and years 2005–2006 (Theodosi et al. 2011), 40.5 and 32  $\mu\text{g}/\text{m}^3$  for high traffic downtown sampling points at ground level and 25 m above ground level, respectively, and two campaigns on years 2003 and 2004 (Karageorgos and Rapsomanikis 2007). Because of the choice of the sampling site at 14 m above ground level, it is expected that our results may represent lower values than those reported for ground level and downtown sampling sites. During the warm season (April to September), the mean and median values were equal to 21  $\mu\text{g}/\text{m}^3$  while, during the cold season (October to March), the mean and

median values were equal to 22 and 18  $\mu\text{g}/\text{m}^3$ , respectively, indicating the absence of a significant seasonal variability which is in agreement with Vassilakos et al. (2005) and Theodosi et al. (2011).

Figure 1 presents the temporal variability of PM<sub>2.5</sub> mean daily concentrations for the sampling period. On the same figure, the annual target value of 25  $\mu\text{g}/\text{m}^3$  is also shown. From this figure, the following can be pointed out: (1) unusually high PM<sub>2.5</sub> concentrations occurred between February 17th and 20th: PM<sub>2.5</sub> concentration reached 100  $\mu\text{g}/\text{m}^3$ . (2) Most of the recorded values approach 20  $\mu\text{g}/\text{m}^3$ , reflecting the mean and median values reported for the year 2010. (3) PM<sub>2.5</sub> concentrations equal and/or higher than 25  $\mu\text{g}/\text{m}^3$  correspond to 13 periods of 1–5 days representing 22 % of the values determined.

#### 3.2 Temporal Variability of Elemental Concentrations

Table 1 presents the correlation coefficients between the elements derived from the data of the whole period studied. The elements Al, Si, Fe, Ca, K, Mg, and Ti are highly correlated. Sulfur does not correlate with any of the other elements. Figure 2a presents the temporal variability of the daily concentrations of four major elements of crustal origin: Al, Si, Fe, and Ca. Figure 2b presents the temporal variability of the daily concentrations of Al, K, Mg, and Ti (Al was added in the figure to facilitate the comparison between the variability of a major dust constituent to that of the elements K, Mg, and Ti which also have a significant



**Fig. 1** Temporal variability of PM<sub>2.5</sub> mean daily concentrations for the sampling period

**Table 1** The correlation coefficients ( $r^2$ ) between the elements

	Al	Si	Ca	Fe	K	Ti	Mg	S
Al	1.000							
Si	0.998	1.000						
Ca	0.965	0.970	1.000					
Fe	0.983	0.986	0.942	1.000				
K	0.960	0.968	0.938	0.978	1.000			
Ti	0.981	0.985	0.949	0.995	0.971	1.000		
Mg	0.961	0.961	0.962	0.920	0.907	0.928	1.000	
S	-0.084	-0.070	-0.058	-0.055	0.060	-0.069	-0.055	1.000

crustal origin). A very similar temporal variability with simultaneous concentration maxima was obtained from our data for the elements Al, Fe, Ca, Ti, K, and Mg as it can be seen from Fig. 3a, b. The maximum of February 20th corresponds to 15.17  $\mu\text{g}/\text{m}^3$  of Si; 6.91  $\mu\text{g}/\text{m}^3$  of Al; 6.28  $\mu\text{g}/\text{m}^3$  of Ca; 6.44  $\mu\text{g}/\text{m}^3$  of Fe; and 0.69  $\mu\text{g}/\text{m}^3$  of Ti. This is in good agreement with concentration values reported for crustal origin elements for intense dust events. For example, Kocak et al. (2007) reported for dust events arithmetic mean concentration values of Ca, Fe, and Ti at 3.5, 1.2, and 0.1  $\mu\text{g}/\text{m}^3$ , respectively, whereas the recorded concentration maxima were much higher, e.g., 7  $\mu\text{g}/\text{m}^3$  for Fe. Maxima for Ca, and Ti in the coarse fraction were reported for dust events to be 18.6, 8.5, and 0.29  $\mu\text{g}/\text{m}^3$ , respectively (Koulouri et al. 2008). In a previous paper on an intense Saharan dust transport event recorded for  $\text{PM}_{10}$  in our sampling site, maxima for Si, Al, Ca, and Fe were 9, 5, 9, and 2.3  $\mu\text{g}/\text{m}^3$ , respectively (Remoundaki et al. 2011).

The concentrations of the elements of crustal origin presented a high variability as can be seen from Fig. 2a, b, for example, Si concentrations ranged between 5 and 15,165  $\text{ng}/\text{m}^3$  for the whole sampling period. Crustal origin elements concentrations maxima correspond to ten short periods occurring during spring and autumn, and as it will be presented in the following section, these periods are associated with dust transport events from African regions.

Figure 2c presents the temporal variability of the daily concentrations of sulfur selected to represent a tracer of anthropogenic origin and a significant constituent of  $\text{PM}_{2.5}$ . Sulfur concentrations present a different temporal variability than that of the elements of crustal origin and of a shorter range (between 350 and 2,921  $\text{ng}/\text{m}^3$ ). Sulfur concentration annual mean value was calculated to be 1,398  $\text{ng}/\text{m}^3$  ( $\pm 635$ ) (4.2  $\mu\text{g}/\text{m}^3$  of

sulfates). A seasonal pattern of sulfur concentrations can also be seen from Fig. 2c with lower values during autumn and winter—1,076 ( $\pm 529$ )  $\text{ng}/\text{m}^3$  (3.2  $\mu\text{g}/\text{m}^3$  as sulfates) and higher values during spring and summer—1,699 ( $\pm 632$ )  $\text{ng}/\text{m}^3$  (5.1  $\mu\text{g}/\text{m}^3$  as sulfates). These values are lower to those reported for Athens and Thessaloniki downtown sites as expected (Karageorgos and Rapsomanikis 2007; Terzi et al. 2010; Theodosi et al. 2011). The seasonal pattern is also in agreement with these studies, since the higher values reported for the warm season are mainly due to enhanced photochemistry, lack of precipitation, low air masses renovation at regional scale, or the increment of the summer mixing layer depth, favoring the regional mixing of polluted air masses (Mihalopoulos et al. 2007; Theodosi et al. 2011).

In the following section, the relationship between  $\text{PM}_{2.5}$  concentrations maxima and  $\text{PM}_{2.5}$  composition will be examined as a function of the origin of the corresponding air masses.

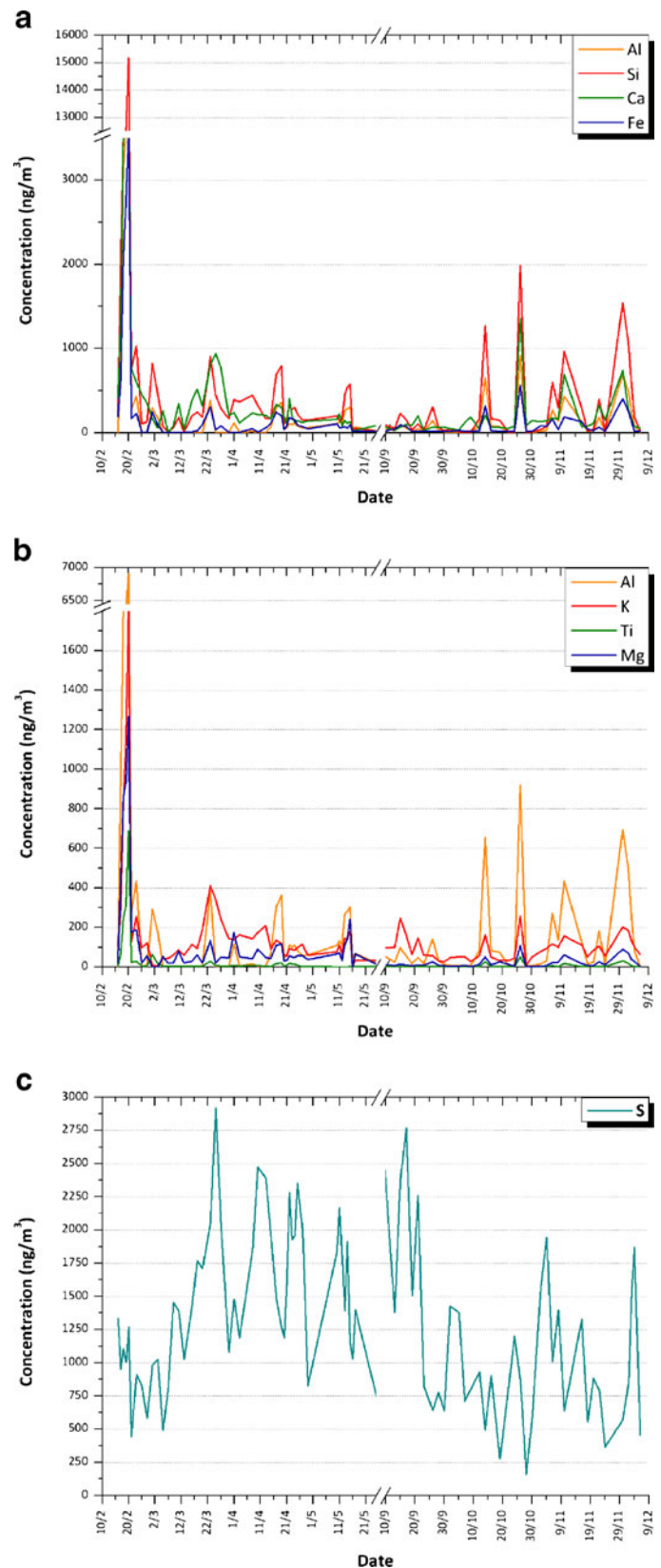
### 3.3 Relationship between $\text{PM}_{2.5}$ Concentrations Maxima and Composition and the Origin of Air Masses

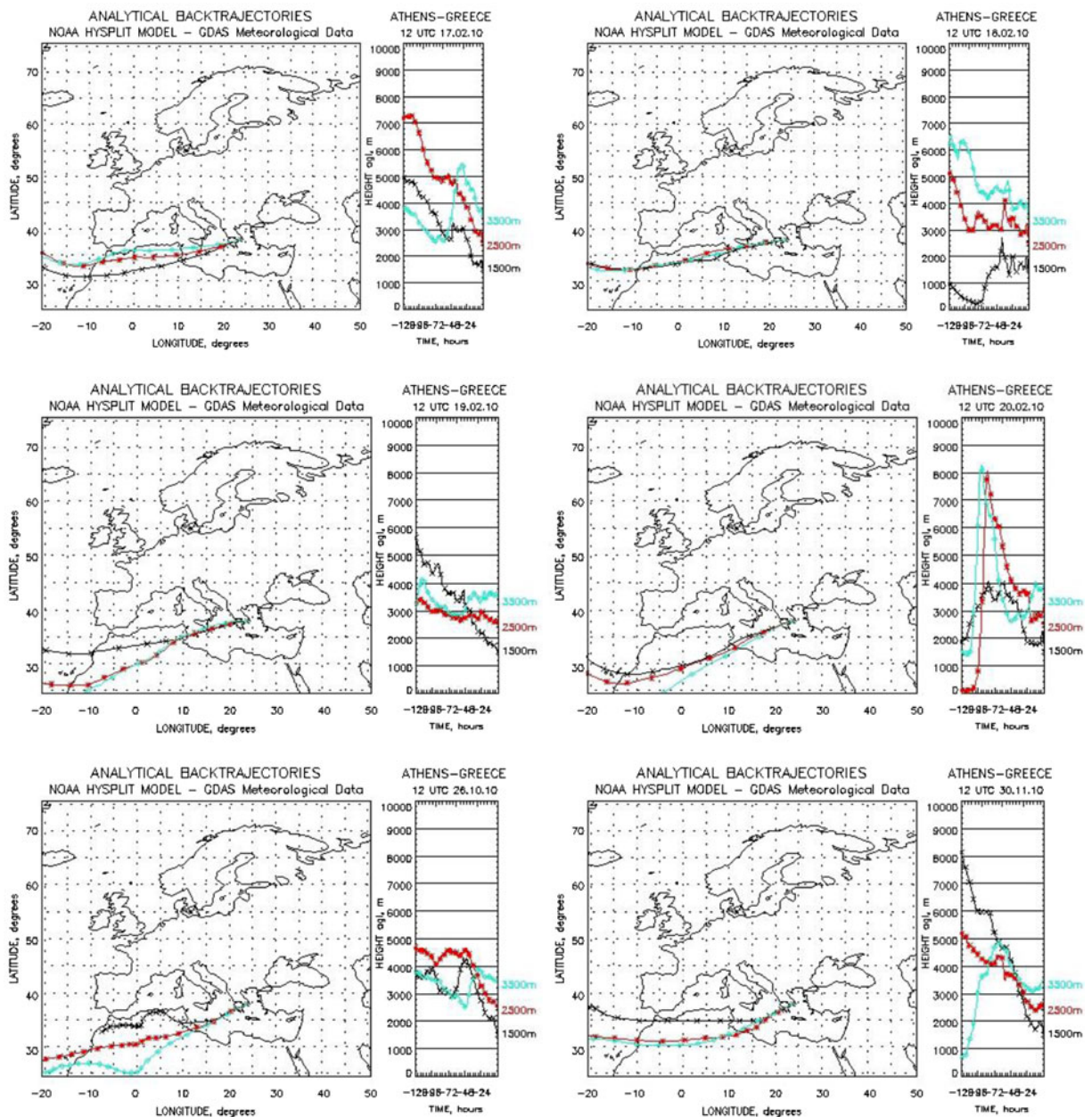
More than half of the air masses arriving in Athens originate from the north sector, which covers central and eastern Europe, as well as part of the western Turkey. This northern contribution reaches almost 85 % during summer. On the other hand, Sahara dust events are very frequent in the area during spring and autumn, contributing up to 25 % of the prevailing air masses (Borge et al. 2007; Gerasopoulos et al. 2011; Kassomenos et al. 2010; Markou and Kassomenos 2010; Mihalopoulos et al. 2007).

Table 2 reports the periods where  $\text{PM}_{2.5}$  concentrations maxima were recorded. In the same table are



**Fig. 2** Temporal variability of **a** Al, Si, Ca, and Fe concentrations in  $PM_{2.5}$ ; **b** Al, K, Mg, and Ti concentrations in  $PM_{2.5}$ ; **c** S concentrations in  $PM_{2.5}$  in nanograms per cubic meter





**Fig. 3** Air mass trajectories ending over Athens on 18, 19, and 20 February, 26 October, and 30 November 2010

reported the corresponding Si and S daily concentrations as tracers of dust and anthropogenic origin, respectively, and the origin of the air masses from the air mass trajectory analysis. The set of trajectories evaluated for the present study corresponds to the triple of the dates of sampling (116 samples, 348 trajectories) as, in addition to the date of sampling, the trajectories corresponding to 2 days before that date were also calculated. For each date, the calculations were made for the arrival heights

of 1, 2, and 3.5 km asl over Athens. According to our data, 13 periods of  $PM_{2.5}$  daily concentrations maxima were recorded with the highest values observed between February 17th and 20th where  $PM_{2.5}$  mean daily concentration reached  $100 \mu g/m^3$ . The corresponding air mass trajectories are shown on Fig. 3 where the air masses present significant homogeneity in altitude. This episode of high intensity may be considered comparable to that which occurred between 29 March and 2

**Table 2** PM<sub>2.5</sub> concentration maxima (in micrograms per cubic meter), Si and S mean daily concentrations (in nanograms per cubic meter), and origin of the corresponding air masses

No	Date	PM <sub>2.5</sub> , µg/m <sup>3</sup>	Si, ng/m <sup>3</sup>	S, ng/m <sup>3</sup>	Origin of air masses
1	17 Feb 2010	21	2,103	951	NW Africa
	18 Feb 2010	50	6,790	1,105	
	19 Feb 2010	60	8,306	1,006	
	20 Feb 2010	100	15,165	1,268	
2	1 March 2010	26	820	980	NW Africa
	2 March 2010	20	469	1,026	
3	20 March 2010	26	184	1,711	Western Europe/Mediterranean/NW Africa on 22 March local circulations
	23 March 2010	35	907	2,051	
	25 March 2010	30	448	2,921	
	27 March 2010	26	288	2,043	
4	10 April 2010	30	322	2,475	Northern Europe
	13 April 2010	25	169	2,390	
	15 April 2010	22	173	1,961	
5	17 April 2010	19	690	1,486	NW Africa
	19 April 2010	26	794	1,270	
6	22 April 2010	25	319	2,281	NW Europe/NW Africa on 24 April
	23 April 2010	26	276	1,928	
	24 April 2010	24	302	1,960	
	25 April 2010	23	221	2,352	
	27 April 2010	19	148	2,005	
7	13 May 2010	26	623	1,372	NW Africa
	14 May 2010	24	515	1,851	
	15 May 2010	26	735	1,448	
8	13 July 2010	28	—	—	Northern and Western Europe
	15 July 2010	28	—	—	
	17 July 2010	27	—	—	
9	15 September 2010	27	232	2,384	Northern Europe
	17 September 2010	34	152	2,768	
	21 September 2010	31	103	2,258	
10	14 October 2010	29	1,273	493	Northern Africa (Libya)
11	26 October 2010	39	1,986	865	NW Africa
12	10 November 2010	30	966	639	NW Africa
13	30 November 2010	25	1,541	569	NW Africa
	2 December 2010	25	494	847	NW Africa
	3 December 2010	19	261	1,523	Northern Africa

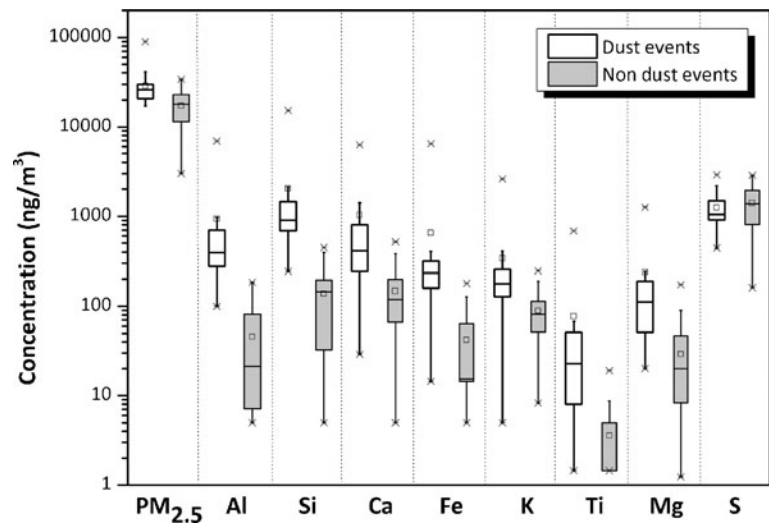
April 2009 recorded in our sampling site for PM<sub>10</sub> (Remoundaki et al. 2011; Papayannis et al. 2012) in terms of origin and intensity, since both dust transport events originated from western Saharan sources and resulted to very high particulate concentrations, 100 µg/m<sup>3</sup> for PM<sub>2.5</sub> in the present study and 156 µg/m<sup>3</sup> for PM<sub>10</sub> in Remoundaki et al. (2011).

As it can be seen in Table 2, PM<sub>2.5</sub> concentrations maxima are associated—for ten from the 13 cases—to

increases of Si concentrations and long-range transport from African regions. The net increases of Si (and the other crustal origin elements or elements with a main crustal origin) concentrations influenced by Saharan dust transport events have been already presented in Fig. 2 and reported below in Fig. 4, a graphical representation of key values from summary statistics. For dust events, mean Si (2,000 ng/m<sup>3</sup>) and median Si (1,000 ng/m<sup>3</sup>) silicium concentrations are



**Fig. 4** PM<sub>2.5</sub> and elemental concentrations (nanograms per cubic meter) for dust events and the periods corresponding to absence of dust event. *Multiplication sign*: percentiles (1–99 %), *small open square*: mean, *box*: 25–75 % percentile, *dash*: median



tenfold higher than those corresponding to non-dust events (mean and median Si concentrations about 200 ng/m<sup>3</sup>, Fig. 4).

The events corresponding to 26 October and 30 November (cases 11 and 13 on Table 2) may also be considered of significant intensity. The corresponding air masses also present significant homogeneity in altitude as it can be seen on Fig. 3.

Dust may be mixed with anthropogenic emissions from North African regions increasing sulfur concentrations during dust outbreaks. Moreover, when during the same period, e.g., case 3 on Table 2, the air masses origin changes including both transports from European continental sources and Sahara, there is mixing of air masses of significantly different origins, and high S concentrations were observed simultaneously with high Si concentrations. Finally, case 6 on Table 2 is an example of air masses having different origins in different altitudes. In this case, the air masses originated mainly from western Europe, but, in higher altitudes, air masses travelled from Sahara (on 24 April). The increase in Si concentrations was less pronounced compared with cases with homogeneous altitude air masses originating from northern Africa.

When the air masses originated from northern and western Europe, e.g., cases 4 and 9 (Table 2), sulfur concentrations were much higher than the mean annual value of 1,398 ng/m<sup>3</sup>.

Summarizing the above, it is apparent that, although dust is mainly associated with the coarse fraction (Gerasopoulos et al. 2006; Kocak et al. 2007; Koulouri et al. 2008) when intense dust transport events occur, as

was the case in February, they can mask any contribution of local sources in PM<sub>2.5</sub> concentrations and any difference between background and urban sites. This was already pointed out for other Greek urban sites (Gerasopoulos et al. 2006; Koulouri et al. 2008; Theodosi et al. 2011). Moreover, the less intense dust transport events contribute significantly in PM<sub>2.5</sub> concentrations. Long-range transported sulfur from the northern and western sectors is added to local emissions doubling the mean sulfur annual concentration.

Previous works (Borge et al. 2007; Kassomenos et al. 2010; Markou and Kassomenos 2010; Gerasopoulos et al. 2011) have shown that, for the air masses arriving over Athens, the south sector is related to the highest aerosol loadings over the area, since it includes the transport of dust particles from desert and arid locations of north Africa with dominant patterns that retain influences from NW Africa (Morocco, Algeria, Tunisia) while the influence of Central Sahara is quite smaller. For the south sector, a high percentage of the air masses (about 89 %) originate from NW Africa (Gerasopoulos et al. 2011). The above findings are confirmed in this work as the dominant pattern was from NW Africa, and only in two cases of Saharan dust transport did the air masses originate from Northern Africa (cases 10 and 13, Table 2).

Figure 4 is a graphical representation of key values from summary statistics. The values presented are the minimum, 25th percentile, median, 75th percentile, and the maximum. Figure 4 reports values of PM<sub>2.5</sub> and elemental concentrations for dust events and the period corresponding to absence of dust event. PM<sub>2.5</sub> mean

concentration for dust events is equal to  $29 \mu\text{g}/\text{m}^3$  while, in absence of dust, event is equal to  $17 \mu\text{g}/\text{m}^3$ . The difference between the concentrations of crustal origin elements and elements with a major crustal component for dust and non-dust periods is also obvious. Sulfur concentrations do not present such a difference since sulfur has a net anthropogenic origin and also a significant local origin, on top of which, long-range transported sulfur is added.

Elemental ratios have been extensively used in order to be compared with those characteristic of different origins. The elemental ratios Al/Si, Ca/Si, Fe/Si, Ti/Fe, and Ti/Ca are reported on Table 3. Al/Si and Fe/Si ratio values can be considered as typical for Saharan dust particles showing little variability (Chiapello et al. 1997; Formenti et al. 2001, 2003; Guieu et al. 2002). The ratios of Ti/Fe and Ti/Ca also present values similar to those reported in literature for long-range transported Saharan dust (Borbely-Kiss et al. 2004; Ganor and Foner 1996).

**Table 3** Elemental ratios Al/Si, Fe/Si, Ca/Si, Ti/Fe, and Ti/Ca for the periods corresponding to dust transport events

Date	Al/Si	Ca/Si	Fe/Si	Ti/Fe	Ti/Ca
18 Feb 2010	0.46	0.67	0.31	0.10	0.05
19 Feb 2010	0.46	0.59	0.31	0.12	0.06
20 Feb 2010	0.47	0.53	0.33	0.11	0.07
21 Feb 2010	0.46	0.41	0.42	0.11	0.11
22 Feb 2010	0.37	1.01	0.22	0.14	0.03
23 Feb 2010	0.42	0.58	0.23	0.13	0.05
2 March 2010	0.36	0.20	0.30	–	–
3 March 2010	0.39	0.13	0.20	–	–
23 March 2010	0.42	0.89	0.34	0.09	0.04
17 April 2010	0.44	0.49	0.35	0.07	0.05
19 April 2010	0.46	0.38	0.26	0.11	0.07
13 May 2010	0.65	0.30	0.10	–	–
14 May 2010	0.53	0.21	0.10	–	–
15 May 2010	0.54	0.18	0.11	0.06	0.03
14 Oct 2010	0.51	0.16	0.25	0.09	0.13
26 Oct 2010	0.46	0.68	0.29	0.09	0.04
10 Nov 2010	0.45	0.72	0.19	0.10	0.03
30 Nov 2010	0.45	0.48	0.26	0.08	0.04
2 Dec 2010	0.46	0.26	0.24	0.07	0.07
3 Dec 2010	0.45	0.28	0.24	0.05	0.04
Mean	0.44	0.57	0.24	0.10	0.05
SD	0.12	0.43	0.09	0.05	0.03

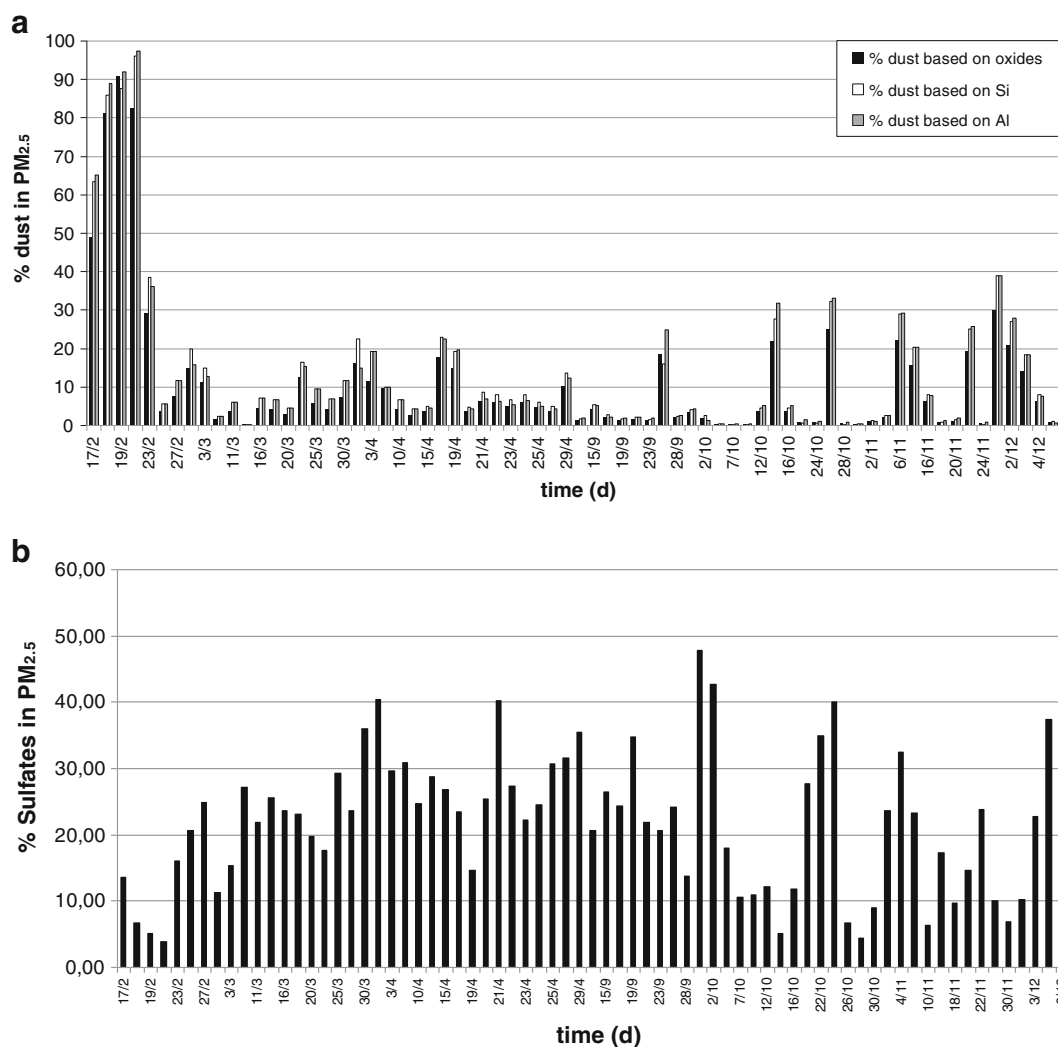
The values of Ca/Si ratio gave an arithmetic mean of 0.57 which falls within the values reported for African dust. Saharan dust originating from Western Sahara is known to be rich in calcite characterized by high Ca/Si ratios, 0.58–0.80 (Avila et al. 1997; Coz et al. 2009; Krueger et al. 2004; Moreno et al. 2005). Dust coming from the Moroccan Atlas region also contains high amounts of dolomite and calcite (Avila et al. 1997). These regions are also dominant origins of Saharan dust transported during the reported period of study, and this may be reflected also in the mean value of Ca/Si ratio. Finally, the above results on elemental ratios are also in very good agreement with those presented in a previous work on an intense Saharan dust transport event recorded over Athens in our sampling site, which was also originated from Western Sahara (Remoundaki et al. 2011).

### 3.4 Percentage Contribution of Dust and Sulfates in $\text{PM}_{2.5}$ Concentrations

The amount of dust was calculated from our data using the elemental determinations results. Since, for each aerosol sample, only the results from the elemental determinations are available and not the exact mineralogical composition, the calculation of the percentage contribution of dust in  $\text{PM}_{2.5}$  concentrations based on the elemental composition present uncertainties. The amount of dust was calculated from our data by three different ways of calculation which are considered as typical in literature:

1. Dust calculation using the typical Al percentage given in literature for Saharan dust:  $\text{Al}/\text{dust} = 7.1\%$  (Dulac et al. 1996; Guieu et al. 2002; Mendez et al. 2010).
2. Dust calculation on Si using the typical Al percentage given in literature for Saharan dust:  $\text{Al}/\text{dust} = 7.1\%$  and the ratio  $\text{Al}/\text{Si} = 0.45$  also typical in literature and in very good agreement to that calculated from our results and reported on Table 3 ( $0.44 \pm 0.12$ ). This results in  $\text{Si}/\text{dust} = 15.8\%$ .
3. From the concentrations of metals of crustal origin after conversion to their common oxides. Ca was multiplied by a factor of 1.95 to account for CaO and  $\text{CaCO}_3$  which are considered as its most abundant forms (Remoundaki et al. 2011; Terzi et al. 2010).

The results of the percentage of dust in  $\text{PM}_{2.5}$  concentrations are presented in Fig. 5a. From this figure, it



**Fig. 5** **a** Percentage of dust in  $PM_{2.5}$  concentrations, **b** percentage of sulfates in  $PM_{2.5}$  concentrations

is apparent that the three ways of calculation gave similar results with higher values corresponding to the calculations based on Al and Si and values systematically lower when the sum of the common oxides was considered. The dust event of February resulted to 82.3 % of dust contribution in  $PM_{2.5}$  when calculated considering the sum of the elements common oxides and to 96–97 % contribution of dust when calculated on Si and Al. For events of significant intensity as those of 26 October and of 30 November, dust contribution was calculated to be of the order of 25–33 % and of 30–39 % when calculated on the basis of oxides and Si and Al, respectively. For the other less intense dust events, our findings are in the same order with 8–21 % reported for  $PM_{2.5}$  for urban sites in Spain

(Querol et al. 2004) and 7.6–19 % reported for  $PM_{10}$  for Athens suburb estimated by modeling (Mitsakou et al. 2008). In absence of dust events, the percentage contribution of dust in  $PM_{2.5}$  was calculated to be 5 ( $\pm 5$ )%. Our results are in good agreement with those reported for  $PM_{2.5}$  and the fine mode for southern European urban sites, 4.4 %–5.2 % (Sillanpaa et al. 2006) and the Mediterranean, 6–10 % (Koulouri et al. 2008).

The percentage contribution of sulfates in  $PM_{2.5}$  is presented in Fig. 5b. Sulfate contribution in  $PM_{2.5}$  is less variable with a mean contribution of 22 % ( $\pm 10$ ) ranging from 5 to 48 %. The mean percentage of 22 % of sulfates in  $PM_{2.5}$  calculated from our results is in very good agreement with the values reported by Sillanpaa et al.

(2006) for PM<sub>2.5</sub> and six urban sites in Europe, 19 % (Duisburg), 20 % (Prague), 14 % (Amsterdam), 26 % (Helsinki), 24 % (Barcelona), and 31 % (Athens).

#### 4 Conclusions

The following conclusions can be drawn from this study.

1. Monitoring of PM<sub>2.5</sub> mean daily concentrations together with quantitative analysis of crustal origin elements and sulfur for the year 2010 allowed the estimation of the contribution of major Saharan dust events in PM<sub>2.5</sub> concentrations and the comparison of this contribution with that of sulfates.
2. Our results have shown that the mean annual value of PM<sub>2.5</sub> concentrations reached 20 µg/m<sup>3</sup>. Twenty-two percent of the reported values were found to be equal and/or higher of the annual target value of 25 µg/m<sup>3</sup> corresponding to 13 short periods of 1–5 days.
3. Ten of the 13 periods of enhanced PM<sub>2.5</sub> concentrations were associated with dust transport events from Sahara. The other periods were associated with local emissions together with long-range transport from northern and eastern Europe.
4. The most intense dust event was recorded from 17 to 20 February, where PM<sub>2.5</sub> reached 100 µg/m<sup>3</sup> and 96 % contribution of dust in PM<sub>2.5</sub> concentrations.
5. The contribution of dust in PM<sub>2.5</sub> concentrations for the other recorded dust transport events ranged between 15 % and 39 %.
6. The air masses corresponding to Saharan dust events originated from NW Africa while the influence of central Sahara is quite smaller.
7. Sulfur, a tracer of anthropogenic origin, has shown a different and less pronounced temporal variability than that of the elements of crustal origin. A mean contribution of 22 % in PM<sub>2.5</sub> concentrations was calculated for sulfates from our results, confirming that sulfates are a major constituent of PM<sub>2.5</sub> as expected and as already reported in literature.

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