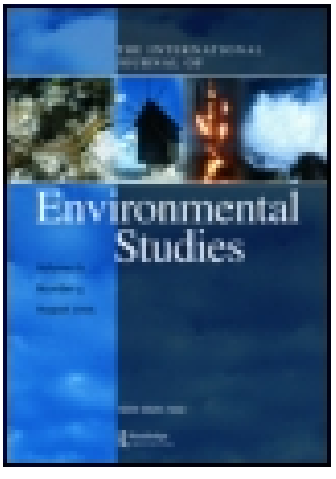

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Roadside PM10 and associated metals in Constantine, Algeria

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Samples of particulate matter less than or equal to 10 μm (PM10) were collected every other day using an Airmetrics MiniVol portable air sampler in Constantine, the third largest and most densely populated city of Algeria with a population of 600,000. The main objective of this study was to assess the concentrations of particulate matter (PM) with an aerodynamic diameter ≤10 μm (PM10), and dust-laden trace metals in a residential area with heavy traffic during two months, from 25 March 2010 to 24 May 2010. Furthermore, the present work examines the daily levels of metallic elements Fe, Zn and Cu. The PM10 concentrations ranged from 7.6 to 159.4 μg/m^3 during the study period. WHO's daily PM10 average guide value of 50 μg/m^3 was exceeded 10 times over 31 samples. HYSPLIT back trajectories were used to identify the source locations of the pollutants. Rain scavenging of atmospheric particulate matter led to a substantial decrease in PM10 concentrations. During the study period, Zn was detected in one sample at a concentration of 0.78 μg/m^3. It is believed that air masses originating from Iceland's Eyjafjöll volcano may have transported clouds of ashes rich in Zn to the sampling site on that occasion. A maximum of 2.92 μg/m^3 of Cu was observed when the volcanic cloud reached the sampling site four times during the corresponding 24-h sampling period.

Keywords: PM10; Metallic elements; Back-trajectories; Dust maps

Introduction

Human exposure to air pollution is believed to cause severe health effects, especially in urban areas where pollution levels are often high. Total suspended matter (TSP) includes particles too large to be inhaled (greater than about 10–15 μm), whereas fine particles (less than about 2.5–3.5 μm) are believed to cause health effects deeper in the lungs [1]. Accordingly, PM10 measurements were performed in this study for the first time in the city of Constantine in order to assess the exposure of the local resident population, to identify potential particulate matter sources and to assess the contribution of road traffic to observed PM10 and trace element levels.

Table 1 shows traffic related PM10 levels reported in developing and developed countries [2–8]. Levels are orders of magnitude higher in Egypt and the developing Asian countries compared with some European countries.

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Table 1. PM10 levels observed in different cities of the world.

City/country	Site		PM10	Source
Egypt	El-Zamalek	Winter 1999	127.2	[2]
	El-Qualaly		219.9	
	Helwan		88.1	
	Kaha		93.0	
	El-Maasara		186.1	
	Shobra		265.1	
Mumbai/India	Khodad	Oct–Nov–Dec	258	[3]
	Marol		528	
Kanpur/India	20 sites	May 2002	139	[4]
Calcutta/India	B.	?	535.9	[5]
	C.	?	1114.5	
	E.	?	909.2	
USA	Albany	07/9/2000–9/3/2001	34.9	[2]
	Birmingham		24.1	
	El Paso		31.5	
	Houston		17.6	
	Las Vegas		18.1	
	Long Beach		46.8	
	Westbury		11.5	
Bastarreche/Spain	Bastarreche Place	25/2/2004–15/3/2005	47	[6]
Rome/Italy	Episode of sea spray formation	11/10–17/10–19/11–27/12 (2004)	24.3	[7]
	Dust storm	31/10–1/11–2/12–3/12–4/12 (2004)	73.9	
	Stable atmosphere	2–6/11–11–17/12	50.6	
England	Network of 14 stations distributed throughout the country	January 1993-December 1995	24.8	[8]

The mean annual PM10 level reported by Kerbachi et al. [9] in Algiers at a distance of 8 m away from a roadway with a fleet of 25,000 vehicles/day was equal to 80.9 μg/m^3. The Algiers problem was not very different from the problem in Constantine, where the car fleet supported by the roadway under study averaged 1500 vehicles/hour during the daytime. Heavy diesel trucks and buses represented 15% of the traffic volume.

Table 2 shows typical ranges of average PM10 concentrations in various regions of the world [10].

Table 2. Annual average PM10 concentrations in various regions of the world (μg/m^3).

Asia	35–220
Latin America	30–129
Africa	40–125
Australia/New Zealand	28–127
Europe	20–70
United States	20–60

Methods

Sampling and measurements

PM10 measurement

PM10 sampling was carried out at Daksi which is an urban site in Constantine with significant traffic to the centre of the city to the south and to major residential areas to the north ($X = 287{,}948$; $Y = 4{,}026{,}439$). PM10 samples were collected from 25 March 2010 to 24 May 2010 every two days. This site is mostly affected by road traffic PM emissions (vehicle exhausts and road dust) and also by PM sources of industrial origin located in the metropolitan area (industrial area of Hamma Bouziane located 7 km north of the city of Constantine). The average daily traffic flow was about 25,000 vehicles a day. Thanks to credit policies and facilities provided by Algerian banks until September 2009, the motor vehicle population has grown substantially during the last 10 years in Algeria. The Algerian vehicle fleet is the second most important in Africa after South Africa. It amounts to approximately 8 million at present. In 2000, it was estimated at 2.9 million units.

PM10 sampling used fibre filters and an Airmetrics MiniVol portable air sampler operating 5 L per minute for pre-programmed 24 h sampling periods. Filters were changed at midday (12 h). A rechargeable battery pack powers the unit. It was situated 7 m above the ground level on the top of a polyclinic. The sampler was placed at about 8 m from the roadside (figure 1).

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 Semi-Micro balance (Shimadzu balance, model AUW-D). The initial weights were determined after a similar period of desiccation. The mass of the sample on each filter was calculated by subtracting the initial weight from the final weight.

Samples were subjected to mass measurements using a gravimetric method and atomic absorption spectrometry (AAS) for Fe, Zn and Cu metals. Proper care in handling the fibre glass filters was observed in order to avoid contamination problems. Lab personnel must wear vinyl gloves when filters are being prepared for lab conditioning and weighing. After the gravimetric analysis, the filters were stored until the AAS elemental analysis could be performed. A Analyst UMR 6264 AAS supporting an acetylene flame was used to analyse the composition of heavy metal elements in PM10. Method IO-3.2 was used for the determination of metals in ambient particulate matter (US Environmental Protection Agency 1999) [11]. Three blanks were subjected to the same procedure to prepare the blank samples. The results of the analysis of blank samples were used to estimate the element concentration produced by the filter and sample preparation. The extraction and digestion procedure followed that of Kuvarega and Taru [12].

Detection of volcanic ashes

The present study is an attempt to identify the potential emission sources, which fall in the track of the HYSPLIT back trajectories under the given meteorological conditions [13]. The back trajectories provide the Lagrangian path of the air parcels in the chosen timescale; the source locations of the pollutants can then be identified.

Back trajectories were computed with the HYSPLIT 4.4 model [14], which provides reliable descriptions of the path followed by the air masses on a synoptic scale [15].

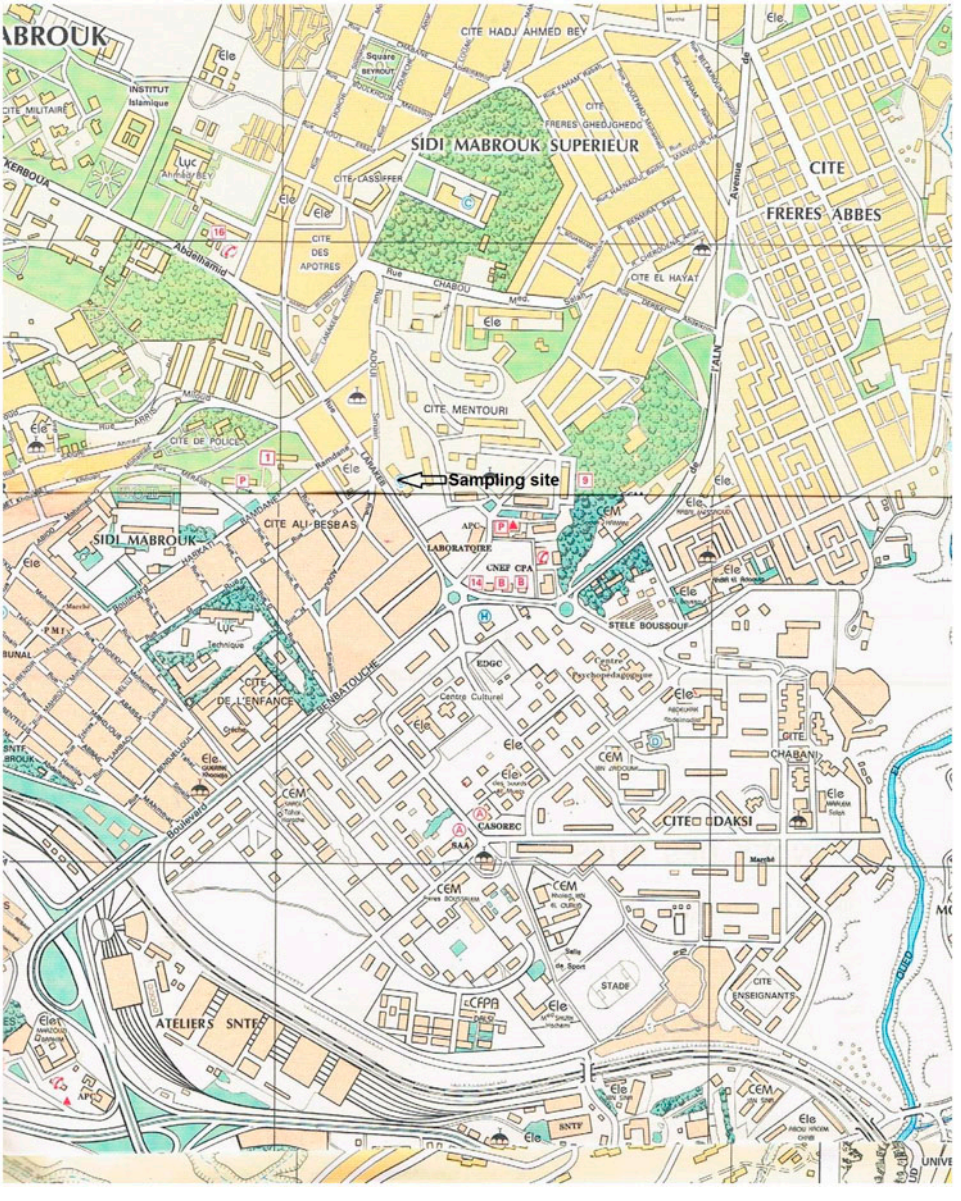

Figure 1. PM10 sampling site. Reproduced with permission of INCT (Institut National de Cartographie et de Télédétection, Constantine, Algeria).

Kinematical three-dimensional back trajectories are calculated using the vertical wind component given by the meteorological model [16].

In general, the transport of air masses over a regional scale (~1000 km) takes 2–3 days [17]. As back trajectories in our study are reaching 3500 km within 315 h (where the volcanic activity was taking place), we have run the HYSPLIT for 315 h which is the maximum modelling time.

The detection of African dust incursions

The possible day-to-day influence of external PM contributions from African dust outbreaks over this region of Algeria was investigated by means of the information from the aerosol maps: Marine Meteorology Division of the Naval Research Laboratory, USA (NRL) [18] and BSC-/DREAM dust maps [19].

The NAAPS (Navy Aerosol Analysis and Prediction System Global) Aerosol Model operates dust simulations in near real time. It provides 120-h forecasts. Regional plots of NAAPS results are presented in a 4-panel format as individual images or as two-day loops containing nine images (six-hourly plots).

DREAM delivers reliable operational dust forecasts to predict all the major dust events. The model predicts the atmospheric life cycle of the eroded desert dust in the Earth Sciences Division of the Barcelona Supercomputing Centre-Centro Nacional de Supercomputación (BSC-CNS) by solving the Euler-type partial differential non-linear equation for dust mass continuity [20].

Results

PM10 concentrations

Figures 2 and 3 show temporal variations of mass and chemical species of PM10. The average PM10 concentration was 49 μg/m^3. This may be regarded as relatively high when compared with those from European urban areas owing to the important contribution of mineral matter [21–23]. It is not very different from the annual mean PM10 level of 40 μg/m^3 that was reported at an urban Mediterranean location in Barcelona. It greatly exceeds the annual average recommended by the WHO of 20 μg/m^3 [24]. The lowest and extreme mass concentration values of PM10 were 7.59 and 159.4 μg/m^3, respectively, during the measurement period.

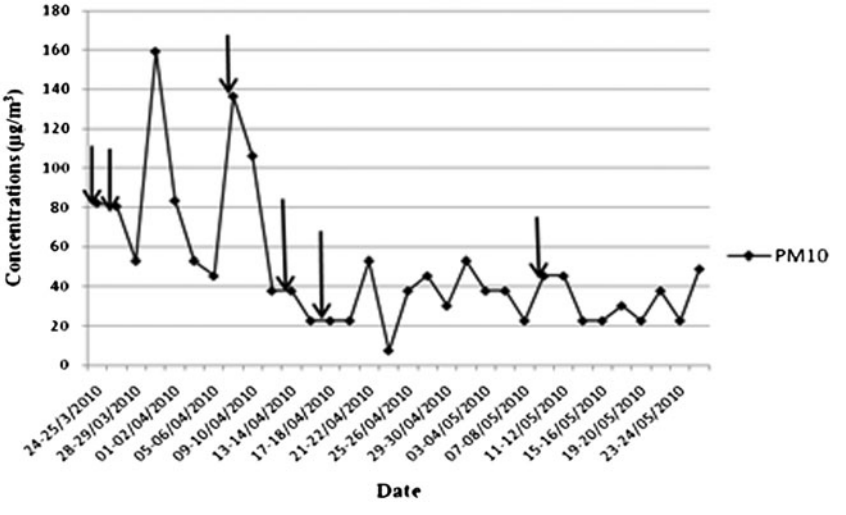

Figure 2. Time series of PM10 concentrations at Daksi. The arrows indicate days with African dust outbreak influence (contributing more than 40 μg/m^3).

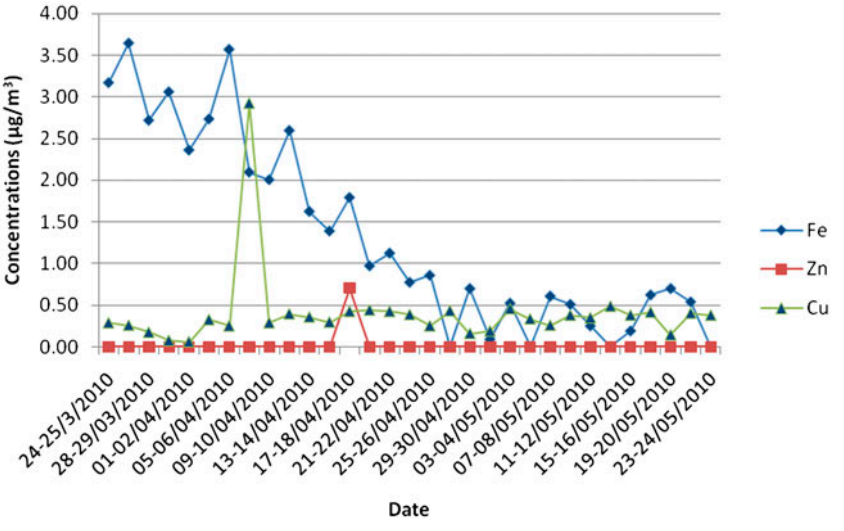

Figure 3. Time series of metallic elements concentrations at Daksi.

Table 3 shows some descriptive statistics of the measured concentrations. Figure 2 shows that the highest PM10 concentrations were observed with filters n° 4, 8 and 9 with levels reaching 159.4, 136.6 and 106.2 µg/m^3, respectively. The latter can be linked to the absence or scarcity of precipitation and/or the occurrence of sandstorms. The second concentration can be linked to the sandstorm which occurred on the 7–8 April 2010 (figures 4 and 5). The contribution of African dust was in the range 80–160 µg/m^3. The other two concentrations (30–31 March 2010 and 11–12 April 2010) are not caused by sandstorms, but are correlated with north-west winds which can bring resuspended dust easily to the sampling site from an elevated upwind area.

In addition, there are two notable trends in the study time interval. The period extending from 24 March 2010 to 9 April 2010 was characterized by relatively high concentrations ranging from 45.9 to 159 µg/m^3. The average concentration recorded during this period was 88.7 µg/m^3. A sharp drop in average concentration was then observed during the period extending between 11 April 2010 and 24 May 2010. The average for this period was 32.8 µg/m^3. The decrease in concentrations between samples 9 (9–10 April 2010) and 10 (11–12 April 2010) was 64%. This sudden PM10 drop appears to be explained by rainfall. This was only 3 mm on the morning of 12 April 2010 (sample No. 10) and 33 mm on 17 April 2010 (sample No. 13). Nevertheless, precipitation scavenging of atmospheric aerosols, soil wetness and the subsequent interruption of dust resuspension may explain the sharp decrease in levels of PM10.

The lower concentrations are generally either aligned with weekends or attributable to meteorological conditions such as rain, and the absence of sandstorms.

It is anticipated that mineral matter includes both an important anthropogenic contribution and significant inputs from natural sources, such as African dust [25].

For the period considered (March 2010–May 2010), mean PM levels for days with African dust outbreaks reached 68 µg/m^3 for PM10 and for the rest of the days 45 µg/m^3.

Figure 3 shows temporal variations of chemical species of PM10 (Fe, Zn and Cu).

Table 3 summarizes the results of the analysis of PM10, metallic elements and precipitation data.

Table 3. Meteorological data, mass and metal concentrations.

N°	Date	Prec. (mm)	(μg/m^3)			
			PM10	Fe	Zn	Cu
01	24–25/03/10	0	82.30	3.17	0.00	0.29
02	26–27/03/10	0	80.59	3.64	0.00	0.26
03	28–29/03/0	0	53.13	2.72	0.00	0.17
04	30–31/03/10	0	159.40	3.06	0.00	0.08
05	01–02/04/20	0	83.49	2.36	0.00	0.06
06	03–04/04/20	0	53.13	2.73	0.00	0.33
07	05–06/04/20	0	45.54	3.57	0.00	0.25
08	07–08/04/20	0	136.63	2.09	0.00	2.92
09	09–10/04/20	1	106.27	2.00	0.00	0.29
10	11–12/04/20	3	37.95	2.60	0.00	0.39
11	13–14/04/20	0	37.95	1.62	0.00	0.36
12	15–16/04/10	0	22.77	1.39	0.00	0.30
13	17–18/04/10	33	22.77	1.79	0.71	0.43
14	19–20/04/10	0	22.77	0.97	0.00	0.44
15	21–22/04/10	0	53.13	1.12	0.00	0.43
16	23–24/04/10	0	7.59	0.77	0.00	0.39
17	25–26/04/10	0	37.95	0.86	0.00	0.25
18	27–28/04/10	0	45.54	0.00	0.00	0.43
19	29–30/04/10	0	30.36	0.70	0.00	0.16
20	01–02/05/10	2	53.13	0.09	0.00	0.19
21	03–04/05/10	7	37.95	0.52	0.00	0.46
22	05–06/05/10	0	37.95	0.00	0.00	0.33
23	07–08/05/10	0	22.77	0.61	0.00	0.26
24	09–10/05/10	0	45.54	0.51	0.00	0.38
25	11–12/05/10	0	45.54	0.25	0.00	0.35
26	13–14/05/10	0	22.77	0.00	0.00	0.49
27	15–16/05/10	0	22.77	0.19	0.00	0.38
28	17–18/05/10	0	30.36	0.62	0.00	0.42
29	19–20/05/10	0	22.77	0.70	0.00	0.14
30	21–22/05/10	0	37.95	0.54	0.00	0.40
31	23–24/05/10	6	22.77	0.00	0.00	0.38

Iron

The temporal variation of dust laden iron in ambient air is well correlated with that of PM10 (0.56) (figure 3). The average iron concentration is 3.06 μg/m^3. Iron particles originate mainly from the wear of rocks and soil [26]. By way of comparison, Fe concentrations measured at different commercial, industrial, residential and rural sites from 1991 to 1995 in Won Ju in South Korea varied between 1.02 and 3.37 μg/m^3 on a monthly basis [27]. Fe concentrations measured in Cairo were reported to vary between 1.2 and 6 μg/m^3 in six polluted areas [2]. The mean Fe concentration measured at a heavy traffic street intersection in central Madrid was equal to 1.93 μg/m^3 [28]. The average Fe concentration measured at our site compares well with these values.

Zinc

No zinc was present in all samples with the exception of filter n° 13 (17–18 April 2010) which revealed a concentration of 0.71 μg/m^3. This sample coincided with heavy rain which amounted to 33 mm in 24 h.

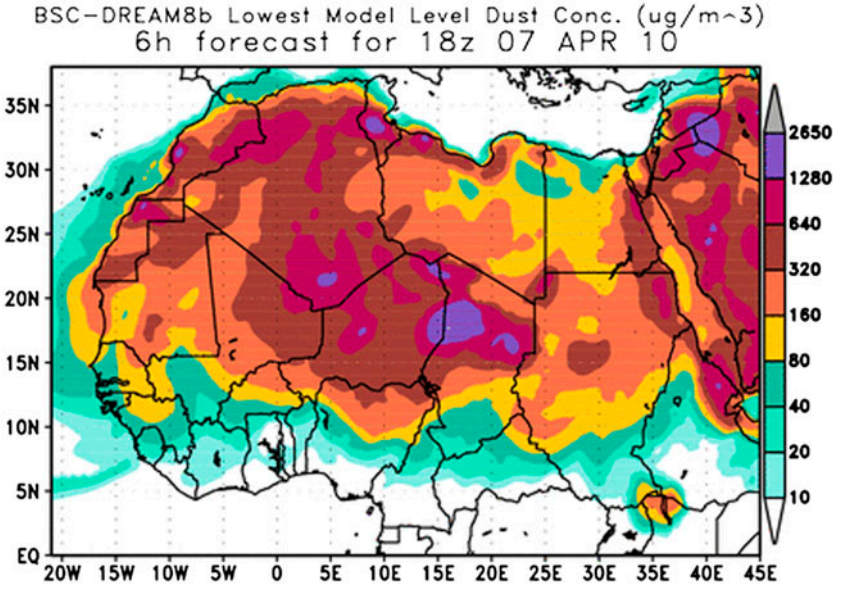

Figure 4. Saharan dust concentration modelled for the date 07/04/2010 at 18 h (BSC/Dream forecast: Barcelona Super Computing Centre: Dust REgional Atmospheric Model).

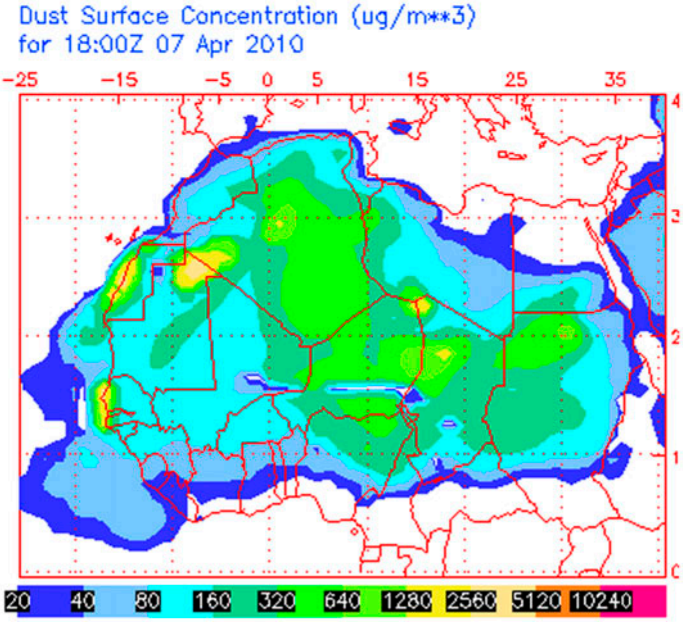

Figure 5. Contribution of sandstorm to PM10 on 07/04/2010 at 18 h.

An analysis of the geographical origin was performed for remote sources. The movement of masses can be depicted by back-trajectories. The HY-SPLIT trajectory model (hybrid single-particle Lagrangian integrated trajectories) performed an air back-trajectory

analysis. A 10-day model vertical velocity back trajectory was obtained at 385 m AGL at 24 h steps. Meteorological data fields supplied by the National Climatic Data Center (NCDC, USA) have been used as the input. The trajectory to the measurement site was developed on 17 April 2010 at 19 h (figure 6). This trajectory passed through the geographical region of Iceland's Eyjafjöll volcano.

We selected 385 m as the receptor height for the following reasons. Our meteorological station installed next to the TAS sampler recorded an hourly volume of 14 mm of rain at 19 h UTC. Historical cloud top height data are available at every hour for over one million geographical sites in the world [29]. Rainy clouds were shown to be located at 400 m in Constantine for that rain vent.

A possible explanation would be that Zn originating from Iceland's volcanic activity which started on 20 March 2010 was subject to rain scavenging over the city of Constantine on 17 April 2010 at 19 h. The consecutive evaporation of rainwater would have left Zn particles on the soil surface. Traffic movement would have generated the resuspension of road dust containing high levels of Zn.

Several studies indicate that Zn levels are among the highest in volcanic ash. The yearly total fluxes to the atmosphere of Zn from degassing volcanoes worldwide are among the most important [30]. Zn was among the elements with the highest concentrations ranging

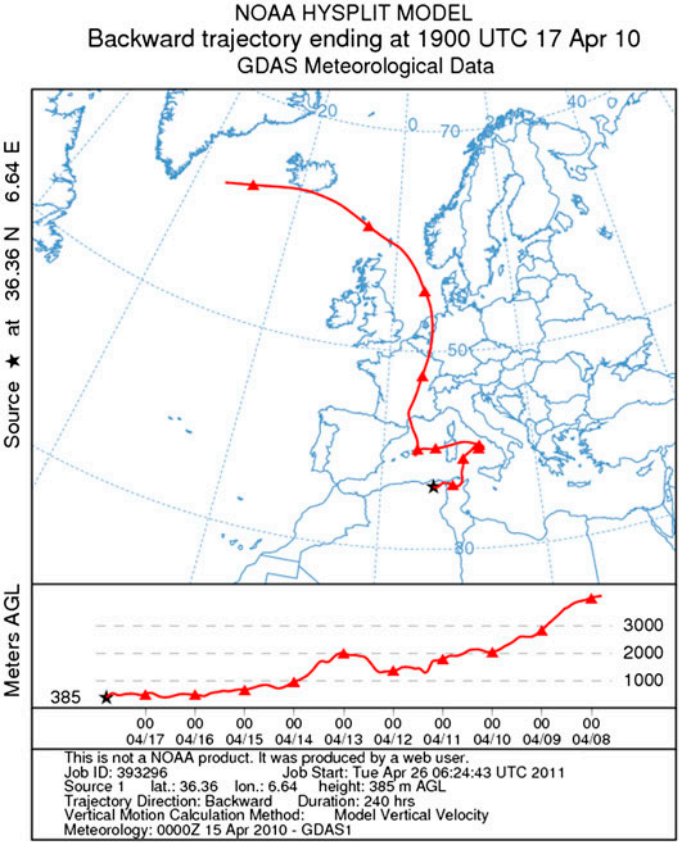

Figure 6. 10-day back trajectory ending at 19 h on 17/04/2010.

from 55 to 85 µg/g in volcanic ashes ejected from Copahue volcano in Argentina [31]. Ancient volcanic ash deposits in northern Argentina are very well preserved and their analysis revealed that Zn was the fourth most ubiquitous element in eight samples [32].

According to WHO [33], zinc adsorbed on particles of small diameter and low density can travel in the atmosphere as far as Norway even if their sources are located in Central Europe.

Copper

The average Cu concentration was 0.40 µg/m^3. Such a value does not exceed the annual Massachusetts Department of Environmental Protection allowable ambient limit of 0.54 µg/m^3 [34]. Cu levels varied between 60 and 2920 ng/m^3. The mean Cu concentration measured from June 1999 to May 2000 twice per week on working days at a heavy traffic street intersection in central Madrid was equal to 112 ng/m^3 [28]. Cu concentrations observed in PM10 in Barcelona, Spain between 2003 and 2006 were equal to 80 ng/m^3 [22]. Cu levels measured at the study area may be considered as relatively high when compared with the mean levels observed at urban sites in Spain. They are higher than measured Cu concentrations from 25/11/2004 to 07/10/2005 at the Directorate of Health

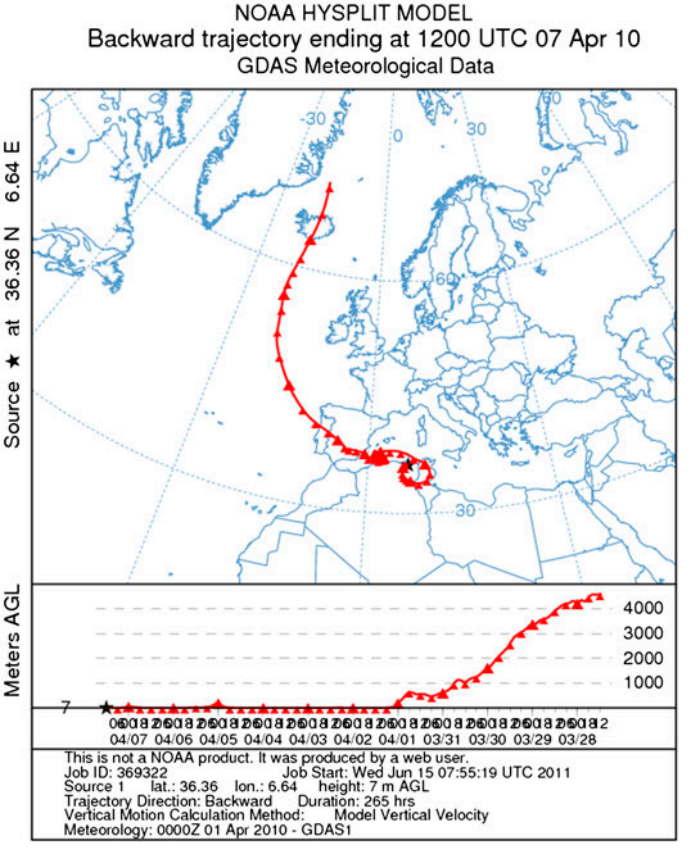

Figure 7. 11-day back trajectory ending at 12 h on 07/04/2010.

located in Avenue Belouizdad in Constantine. These values ranged from 2.66 to 341.48 ng/m^3 with a mean value of 50 ng/m^3 [35]. They are, however, comparable to reported concentrations by WHO for the cities of Chicago and St. Louis ($0.1 < Cu < 1610$ ng/m^3 in the particles from 1 to 2.5 microns and $0.1 < Cu < 2240$ ng/m^3 in particles 2.5–10 μm) [36].

Analysis of 11-day back trajectories for the sampling 24-h period coinciding with the peak Cu concentration (7–8 April 2010) shows that air masses originating from the volcanic eruption of Eyjaföll reached our study site four times (12, 13, 16 and 04 h) (figure 7). The absence of Zn in this phenomenon reflects the variability in the composition of the volcanic ash over time. In addition, the maximum level of copper (2920 ng/m^3) could be attributed to emissions from the metallurgical industry located NNW of the sampling site, from brake wear in motor vehicles, and to road dust emanating from the intersection overlooking the clinic by prevailing NW and NNW winds. Copper is also emitted by catalytic converters. According to Magdalene [37], cars emit mainly copper and iron.

Conclusions

This work allowed measurements for the first time of PM10 and some associated metallic elements in an urban site within the city of Constantine. This site reveals the presence of PM10 and metal elements at concentrations far above internationally acceptable levels. This study showed that iron is well correlated with PM10 indicating that their origin is, at least in part, common. The presence of zinc, detected on one occasion on 17–18 April 2010, is related to the resuspension of dust initially brought to the ground by rains, which were heavily loaded with zinc transported by volcanic ash originating from Iceland's Eyjaföll volcano. Subsequent evaporation of rainwater and resuspension by wind of dust-laden Zn led to the detection of Zn at an abnormally high concentration of 0.78 μg/m^3. Copper is linked to volcanic ash, wind sand, brake-wear emissions as well as wind-blown particles. Through analysis of back trajectories starting from the measurement site, we highlighted the variability in time of found materials originating from the eruption.

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