

Monitoring of air pollution by total suspended particulates and heavy metals at Frantz Fanon site in Algeria

Surveillance de la pollution de l'air par les particules totales en suspension et les métaux lourds au niveau du site Frantz Fanon, en Algérie

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Abstract

In this paper we applied the technique of Energy dispersive X-ray fluorescence spectroscopy (EDXRF) to study the pollution levels of aerosol particles and heavy metals associated with them (Fe, Mn, Pb, Co, Ni, Cr) in a site influenced by the emissions of the road traffic in Algiers. Aerosols samples were analyzed for different seasons from January 2009 to February 2010. Low volume air sampler was used to collect the samples. The mean aerosols concentrations were 51 $\mu\text{g}/\text{m}^3$ in winter, 67 $\mu\text{g}/\text{m}^3$ in spring, 84 $\mu\text{g}/\text{m}^3$ in summer and 58 $\mu\text{g}/\text{m}^3$ in autumn. The concentration of heavy metals in total suspended particulate matter (TSP) varies with weather conditions. The measured elements showed a strong seasonal variation, with the highest concentration in summer and the lowest in winter. The mean Ni, Co, and Cr levels were higher than the proposed WHO and the European community standards. Significant positive correlation was found between the concentrations of aerosols, Fe and Mn. The enrichment factors with respect to earth crust were investigated to predict the possible sources of heavy metals in air.

Keywords

aerosols, heavy metal, TSP, road traffic, EDXRF, Algiers

Résumé

Dans cet article, nous avons appliqué la technique de la spectroscopie de fluorescence des rayons X (EDXRF) pour étudier les niveaux de pollution par les particules d'aérosol et les métaux lourds qui leur sont associés (Fe, Mn, Pb, Co, Ni, Cr) dans un site influencé par les émissions du trafic routier à Alger. Les échantillons d'aérosols ont été analysés pour différentes saisons, de janvier 2009 à février 2010. Un échantillonneur d'air à faible volume a été utilisé pour collecter les échantillons. Les concentrations moyennes d'aérosols étaient de 51 $\mu\text{g}/\text{m}^3$ en hiver, 67 $\mu\text{g}/\text{m}^3$ au printemps, 84 $\mu\text{g}/\text{m}^3$ en été et 58 $\mu\text{g}/\text{m}^3$ en automne. La concentration des métaux lourds dans les matières particulaires en suspension totale (TSP) varie en fonction des conditions météorologiques. Les éléments mesurés ont montré une forte variation saisonnière, avec la concentration la plus élevée en été et la plus faible en hiver. Les valeurs moyennes de la concentration de Ni, Co et Cr étaient plus élevées que les valeurs proposées par l'OMS et les standards de la communauté européenne. Une corrélation positive significative a été observée entre les concentrations d'aérosols et les éléments Fe, Mn. Les facteurs d'enrichissement de la croûte terrestre ont été étudiés pour prédire les sources possibles de métaux lourds dans l'air.

Mots-clés

aérosols, métaux lourds, TSP, trafic routier, EDXRF, Alger

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1. Introduction

Atmospheric contamination has become a matter of world-wide concern, particularly in the largest cities of the world. In ambient air, heavy metals are present in form of particles and gases. These particulates are one of four categories of air pollutants, and most of them are dangerous to human tissues and organs (e.g. respiratory system, cardiovascular system, nervous system, urinary system) (Xiong, 2015). Their coating of surfaces (soil, water, air...) also led to contamination of the food chain. The World Health Organization (WHO, 2000) has reported that approximately 4 % to 8 % of premature deaths are attributable to exposure to elevated levels of particulate matter in ambient air. The impact of heavy metals varies according to their concentration, toxicity and duration of exposure. As more pollutants accumulate, their risks will be more serious (Mudri, 1993). In the developing countries, the particulate matter forms the major contributor to air pollution and hence the pressure to understand its sources better (Terrouche, 2015).

In urban areas, untimely discharges by the circulation of combustion-engine vehicles from commercial automobiles and heavy-duty vehicles are the main cause of air contamination. The emission factors naturally vary according to the type of vehicles, fuel and vehicle status (Oucher, 2010). Diesel vehicles and, to a lesser extent, gasoline vehicles predominantly emit very fine particles of size less than 0.1 μm (Gertler, 2002).

In addition to the particles emitted from the exhaust, pollution by road traffic also includes particles from road wear tires (a tire loses about 10 % of his weight during his life time), brake pads and motors (Fe, Cu, Cr...) (Oucher, 2010).

Several studies have been carried out on atmospheric pollution by heavy metals in the city of Algiers and show that there is a high level of pollution (Oucher, 2010, 2015; Kerbachi, 1998, 2006; Belamri, 2010; Yahiaoui, 2012; Laïd, 2006; Ali-Khodja, 2008). Road traffic is the most important source of these emissions (Oucher, 2010). We may, from a search for local information by performing a sampling of urban air, show direct or indirect influence of man on the modification of the physicochemical composition of the air. The determination of heavy and toxic elements permits the study of their distributions, the pollution level as well as the risk assessment in the investigated ecosystems.

Undoubtedly the most popular methods for this purpose at present are atomic absorption spectrometry (AAS), and inductively coupled plasma atomic emission spectrometry (ICP-AES). The need for exhaustive

the sample preparation for AAS and ICP-AES has led to increasing interest towards energy dispersive X-ray spectrometry (EDXRF) in environmental investigations (Hou, 2000).

X-ray spectrometry, in its various forms, is now a powerful, well established and mature technique for environmental analysis. It is a non-destructive technique, it offers multi-element capability, economy, high speed and easy operation and can be applied to any kind of samples, with sufficient sensitivity for the determination of many trace elements (Melquiades, 2004).

The main aim of this work is to investigate the seasonal distributions of total suspended particulate matter and their elemental contents (Fe, Mn, Pb, Co, Ni, Cr) in the atmosphere of Frantz Fanon site, where population density is high and is directly exposed to heavier traffic, and to evaluate the contribution of anthropogenic sources on the levels of these elements.

2. Materials and methods

2.1. Study area

The present study was carried out in the center of Algiers, Algeria. Algiers is an urban city located at 36°43' North latitude and 3°15' East longitude. The present city of Algiers has experienced strong extension geographically and demographically. It has a population of over five million inhabitants. It is characterized by industrial activities (refineries oil, mechanical industry...) in addition to waste incineration operations as well as road traffic. More than a million cars come in Algiers over 1.2 million circulating every day (Belamri, 2010).

In order to evaluate the heavy metal content in the atmosphere, the Ministry of Environment, Territorial Planning and Tourism and the Nuclear Research Centre of Algiers (CRNA) conducted since 2005 to date almost a sampling campaign carried out at the sampling stations located in Algiers center (the high Casbah) and the CHU Mustapha Pasha sites.

The sampling site is located at the nuclear research centre (CRNA), on the heights of Algiers (Figure 1) between two roads with high traffic density (100 000 cars per day) (Belamri, 2010), thus the importance of the number of vehicles that pass near the sampling site. Apart from these two major roads in the vicinity of the site, there are no other sources emitting pollution. For this choice, it is important to know the total amount of pollution generated by road traffic in order to take measures to reduce the risks posed by air pollution from automobiles on human health.

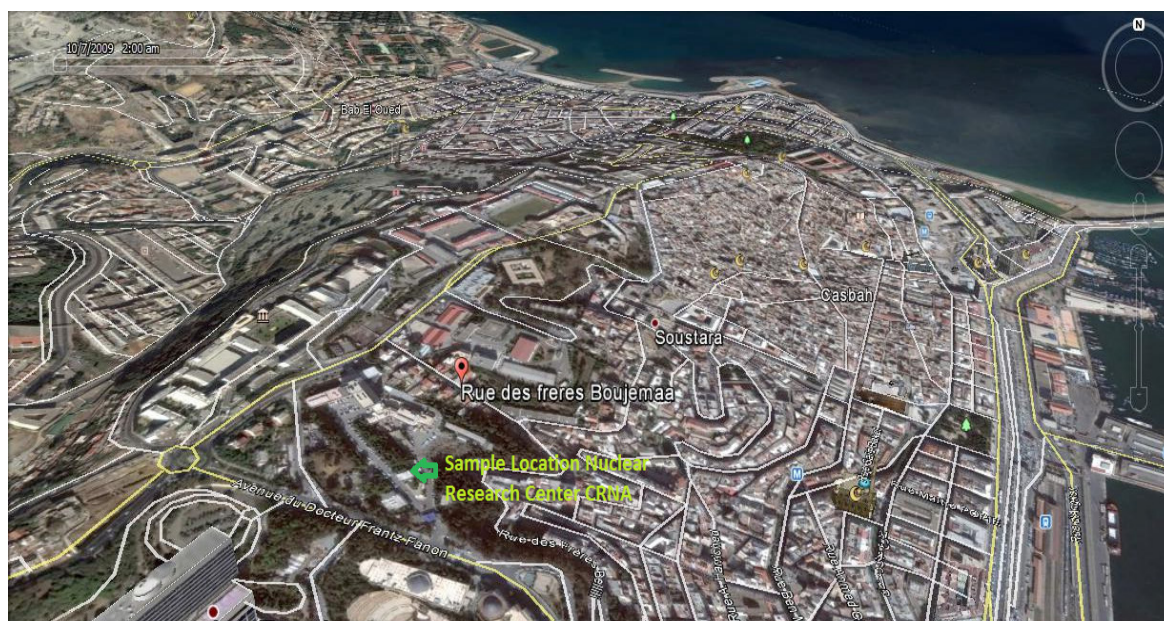


Figure 1. Air sampling area of the urban site of Algiers.
Centre Zone d'échantillonnage de l'air du site urbain d'Alger Centre.

2.2. Sample collection

Air Samples were collected from January 2009 to February 2010 at the same time. The sampling was performed using a Gent Stacked Filter Unit (SFU) sampler with air flow rate of 1.29 m³/h. This type of sampler, recommended by the IAEA in Africa, has the advantage of being easy to use and very cheap, but do not have flow control, it does not maintain a constant particle size cut (Zghaid, 2009). The system is consisting of cellulose ester filters, Millipore type (AA) placed in cascade with pore sizes of 0.8 µm and diameter of 37 mm. Measurements were carried out once per week for 72 hours durations from 8:00-8:00 a.m of the next days. Sampling point is approximately 200 m above sea level.

2.3. Sample preparation and analysis

After sampling and to prevent outside contamination, samples are stored until processing and analysis in sealed petri dishes and kept in desiccators. Knowledge of the deposited weight, the sampling time and the average air flow rate are used to calculate the concentration of particulate matter expressed in µg/m³. For analytical purposes, we have prepared chemical standards from products of known purity, by acid or aqueous dissolution, depending on the sample to dissolve and then we make the necessary dilutions. The concentration of stock solutions is once diluted to make mixtures compatible and non-interfering. We find the desired concentrations in 20 µl. These are taken using a micropipette and deposited on blank filters of the same nature as the filters used for sampling, then, let them dry.

The analysis of trace elements was done using X-ray fluorescence spectrometer (Epsilon 3 Model).

2.4. Meteorological data

The general climate of Algiers city is cold, moist and rainy in winter, high temperature, clear sky and rainless in summer. Meteorology has a significant role in the transformation and transmissions of pollutants. Table 1 summarize meteorological parameters during the period of the present study.

3. Results and discussion

A total of 37 samples were collected from 03/01/2009 to 25/02/2010. During this measurement campaign, the average aerosols concentrations on seasonal basis show that the maximum levels were observed in sunny days (105 µg/m³) and the low levels in the rainy days (28 µg/m³), this is consistent with the results reported by Zghaid, Belamri and Moreno-Grau.

Table 2 shows the range and mean values of concentrations for aerosols and selected heavy metals analyzed based on seasonal variations.

The highest average aerosols concentration in the present study was recorded in summer at 84 µg/m³ since the soil particles become air-borne easily in summer (high temperature) (Al-Momani, 2005) and low in winter (51 µg/m³) (Figure 2).

Table 1. Summary of meteorological parameters during the study period (January 2009 to February 2010).

Récapitulatif des paramètres météorologiques pendant la période d'étude.

Seasons \ Parameters	Rain (mm)	Humidity (%)	Wind speed (Km/h)	Temperature (°C)
Winter				
Min	0	56	14	13
Max	9	84	36	20
Mean	5	75	20	16
Spring				
Min	0	58	12	17
Max	7	87	24	29
Mean	3	74	18	21
Summer				
Min	0	45	16	27
Max	0.6	81	22	36
Mean	0.15	61	19	33
Autumn				
Min	0	58	13	19
Max	12	89	28	34
Mean	3	68	18	25

Table 2. Seasonal variations in atmospheric aerosols ($\mu\text{g}/\text{m}^3$) and their elemental contents levels (ng/m^3).*Variations saisonnières des aérosols atmosphériques ($\mu\text{g}/\text{m}^3$) et de leurs composants (ng/m^3).*

Seasons	Number of samples	TSP	Fe	Mn	Pb	Ni	Co	Cr
Winter								
Min	9	28	1493	47	230	39	2	3
Max		75	4808	158	398	204	20	18
Mean		51	2337	87	328	106	11	8
SD		14	401	20	15	114	7	1
Spring								
Min	9	36	1829	76	96	43	5	51
Max		100	6379	248	365	363	36	16
Mean		65	3803	139	217	169	17	9
SD		2	120	7	46	120	16	7
Summer								
Min	9	44	1566	80	108	89	6	2
Max		105	4848	185	539	764	151	16
Mean		79	3940	152	251	308	40	9
SD		41	1152	23	16	81	3	3
Autumn								
Min	10	36	188	14	17	43	4	4
Max		100	7202	245	675	261	44	15
Mean		54	3446	124	389	158	21	10
SD		13	2250	51	203	50	6	0
Annual								
Min	37	28	188	14	96	39	2	2
Max		105	7202	248	675	764	151	18
Mean		65	3425	126	269	185	23	9

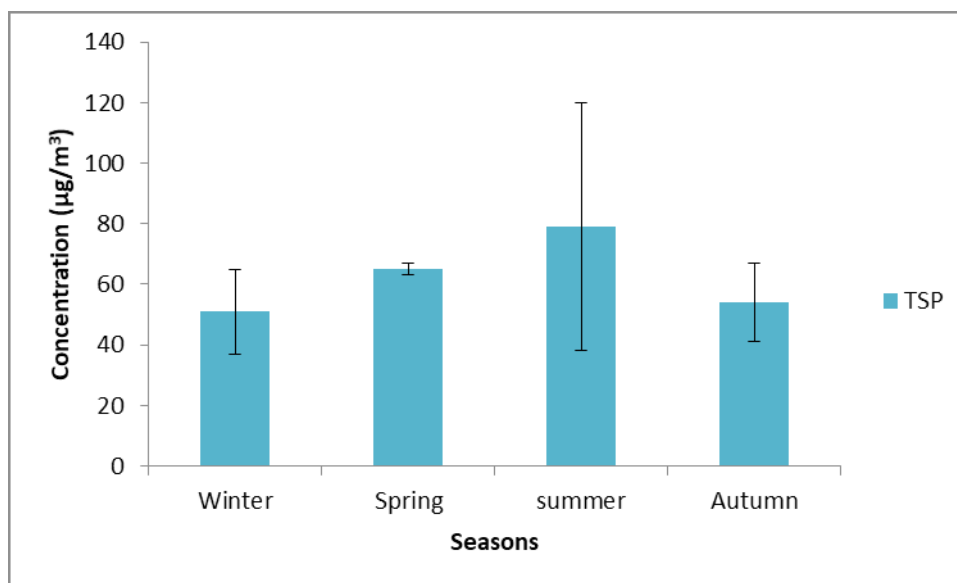


Figure 2. Seasonal variations in the average of total suspended particulates.
Variations saisonnières de la moyenne des particules totales en suspension.

The low concentration in winter can be attributed to wet deposition of TSP caused by precipitation (rainfall), add to this the role of wind in dispersing the particles away from the sampling site. The mean concentration calculated over all samples was 65 µg/m³.

In the present study aerosols annual mean levels is below the Algerian limit value set at 80 µg/m³, it is also much lower than the Moroccan standard (200 µg/m³) and the Tunisian limit value (80 µg/m³). This concentration of TSP in air not exceed the annual average of 60-90 µg/m³ advises by WHO and the European limit value (150 µg/m³) (Busheina, 2017).

Table 2 also shows the summary of mean seasonal variations in elemental levels during the study period. We compared the average concentration of metals in airborne TSP with the Norm suggested by international organizations. The international standards designed for atmospheric Pb, Mn, Cr, and Ni are 0.500, 0.150, 1.10 and 0.00038 µg/m³ (WHO, 2000).

The highest amount of heavy metals was reached in the summer season, except for lead and chromium which are highest in autumn. This is contributed to the raise in human activities over the summer period leading to particulate matter emission (Figure 3). The highest mean concentration was found for Fe at 3 425 ng/m³. This result for iron was predictable, due to its large amount in the earth's crust. Mean concentrations of Pb (269 ng/m³) and Mn (126 ng/m³) were revealed to be inside the WHO and European standards ranges. Based on the results obtained, the level of Pb in air samples in Frantz Fanon site is brought

about by fuel and motor oil combustion since gasoline still contains lead additives in Algeria (Terrouche, 2015). The main source of Mn is earth crust. During the present study, average concentrations of Ni (185 ng/m³) and Cr (9 ng/m³) were higher than those recommended by WHO and the European community. The main emission source of Ni is automobile exhaust fitted with catalytic convertor (Hassan, 2013). Chromium is linked to emissions from the brake lining materials (Moreno-Grau, 2000). The average Co level (23 ng/m³) was 4 times higher than the reference concentration for inhalation of 5 ng/m³ (California Environmental Protection Agency, 1997). The main source for this element is lubricating oils. These results agree with Gharaibeh (2010) who found that the average elemental concentrations in particulate matter were found to be higher in summer season.

3.1. Correlation coefficient analysis results

The correlation coefficient values between aerosols and their elements content are given in Table 3.

Correlation analysis was conducted using the Pearson coefficient method. Inter-element relationships provide interesting information on the sources and pathways of the heavy metals (Lu, 2010). Significant positive correlations were found between the concentrations of aerosols and their components as Fe and Mn.

This may be attributed to the natural input of trace element and aerosols in the atmosphere, as these metals are well known for their crustal abundance.

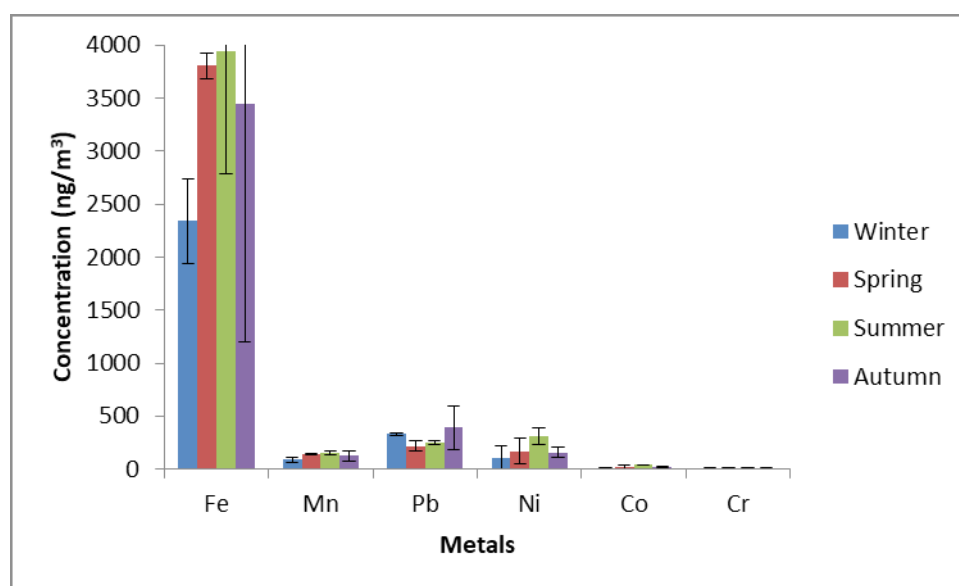


Figure 3. Variation of selected heavy metals concentration according to season.
Variation de la concentration des métaux lourds sélectionnés selon la saison.

A strong significant positive correlation was observed between Fe and Mn ($R = 0.95$), Ni and Co ($R = 0.77$) indicating that both members may be from the same sources (crustal or anthropic) or different sources with the same rate. Relatively average correlation between Mn-Ni, Pb-Fe was observed. No statistically apparent correlation was found between the other elements, their presence may be due to other factors or other sources.

3.2. Enrichment factor

Enrichment factors (EFs) can give an insight into differentiating an anthropogenic source from a natural origin.

The enrichment factor provides the number of times an element is enriched relative to the abundance of

that element in the reference material (earth crust). It was determined depending on the average concentration of elements mentioned by Mason (1966). Enrichment factors are classified into 5 levels of contamination (Sutherland, 2000):

EF<2: anthropogenic enrichment is considered non-existent or very low

2<EF<5: moderate enrichment

5<EF<20: significant enrichment

20<EF<40: very strong enrichment

EF>40: extreme enrichment

Table 3. Correlations matrix for the heavy metal concentrations.
Matrice de corrélation pour les concentrations de métaux lourds.

	TSP	Fe	Mn	Pb	Ni	Cr	Co
TSP	1						
Fe	0,68519201	1					
Mn	0,63297028	0,95595847	1				
Pb	0,04441925	0,45043997	0,36695501	1			
Ni	0,36564793	0,38331484	0,49103878	0,01217079	1		
Cr	0,20952448	0,40585905	0,38997711	0,04778444	0,03626706	1	
Co	0,23491748	0,35389939	0,47644538	0,11773288	0,7704975	0,0916472	1

In this work, crustal Fe levels were utilized as a reference for enrichment factor calculation since soil is considered to be the most important source of Fe in aerosol (Table 4). Results for EFs are presented in the following table.

Table 4. Enrichment Factors (EFs) of selected heavy metals.
Facteurs d'enrichissement (EF) de métaux lourds sélectionnés.

Elements	Enrichment Factors (EFs)
Fe	1
Mn	2
Pb	302
Co	13
Ni	36
Cr	2

In aerosols samples from this study, metals with inferior EFs are Manganese and Chromium, thus representing metals that have its origin from earth crust. The enrichment factor value for Co was 13 (<20), so Cobalt is considered to have a predominantly non crustal source. Nickel has very strong EF with maximum value of 36. The high EF values for Pb indicate that it is present in atmospheric aerosols in concentration too high to be explained in terms of normal crustal weathering process, this element is immensely originating from the anthropogenic sources.

3.3. Comparison to other studies

The comparison will be performed with other countries located at the Mediterranean basin, among them, Morocco (Kenitra), Cairo (Egypt), Libya (Zawiya City), Izmir (Turkey) and Cartagena (Spain) as shown in Table 5.

From this Table, it appears that the mean values of atmospheric aerosols and heavy metal associated with them in the study area were lower, higher or similar to those detected in other countries. This variation was probably due to the difference in the traffic density, industrial activities and the intensity of human activities.

4. Conclusion

The study of air pollution in Algiers center at Frantz Fanon site (CRNA) by total suspended particulates matter and their elemental contents is well defined. This through the results achieved after the different analyzes by XRF spectrometry for all sampled filters.

This study clearly showed that the air breathed at Algiers city is polluted. We found that the average concentration of TSP ($65 \mu\text{g}/\text{m}^3$) in the air not exceeded the WHO and annual average of the European directive.

Seasonal variations for aerosols and heavy metals linked to them were studied. The lowest average of aerosols level ($51 \mu\text{g}/\text{m}^3$) was found in winter, whereas the highest average ($84 \mu\text{g}/\text{m}^3$) was detected in summer. The whole heavy metals have reached their highest concentration for the summer period, except for Pb and Cr. Ambient air concentrations of Pb and Mn were relatively small compared to the WHO guideline values, while those of Ni, Cr and Co were higher than the proposed WHO and the European Community standards.

The values of the enrichment factor (EF) and the correlation coefficient were calculated in order to determine whether the concentrations of heavy metals obtained in aerosols are of anthropogenic or natural origin. Results show that Fe, Mn, Cr come mainly from the soil whereas Co, Ni and Pb are derived from anthropogenic sources.

In Algeria, gasoline still contains lead additives. To minimize the sources of that pollution, it is necessary to put regulations requiring the presence of a catalyst on all new vehicles with better supply of unleaded gasoline and encourage bi-fuel gasoline GPL or GNL.

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Table 5. Particles matter ($\mu\text{g}/\text{m}^3$) and heavy metals contents (ng/m^3) in different countries.
Matière particulaire ($\mu\text{g}/\text{m}^3$) et teneur en métaux lourds (ng/m^3) dans différents pays.

Country	Site		TSP	Fe	Mn	Pb	Ni	Cr	Co	Ref
Frantz Fanon, Algiers (Algeria)	Traffic	Summer	79	3 940	152	251	308	9	40	Present study
		Autumn	54	3 446	124	389	158	10	21	
		Winter	51	2 337	87	328	106	8	11	
		Spring	65	3 803	139	217	169	9	17	
Didouche Mourad (Algeria)	Urbano industrial	Summer	-	-	-	-	-	-	-	Ali-Khodja, 2008
		Autumn	-	-	-	-	-	-	-	
		Winter	117	-	50	46	11	16	-	
		Spring	-	-	-	-	-	-	-	
Kenitra (Morocco)	Traffic	PM10	81	3 079	43	267	449	219	-	Zghaid, 2009
		Coarse	106	2 114	76	217	146	60	-	
		Fine	52	1 936	23	342	166	84	-	
Cairo (Egypt)	Traffic	Summer	250	3 000	140	450	41	25	21	Hassan, 2013
		Autumn	330	3 650	165	570	57	35	26	
		Winter	400	4 400	204	650	68	46	35	
		Spring	290	3 400	153	499	49	28	30	
Izmir (Turkey)	Urban	Summer	-	949	32	115	14	27	-	Yatkin, 2007
		Autumn	-	-	-	-	-	-	-	
		Winter	-	874	25	184	18	25	-	
		Spring	-	-	-	-	-	-	-	
Cartagena (Spain)	Traffic	Summer	97.5	-	-	240	-	-	-	Moreno-Grau, 2000
		Autumn	84.3	-	-	220	-	-	-	
		Winter	86.8	-	-	300	-	-	-	
		Spring	85	-	-	260	-	-	-	
Zawiya City (Lybia)	Traffic	Summer	159	4 432 000	135 000	1 615 000	-	-	-	Busheina, 2017
		Autumn	198	-	-	-	-	-	-	
		Winter	131	-	-	-	-	-	-	
		Spring	239	-	-	-	-	-	-	

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