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Evaluation of air quality in a suburban area of Algiers: A study using INAA and EDXRF technique

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Abstract

This study shows the levels of air pollution concerning the Total atmospheric Suspended Particles (TSP) and the mineral fraction which is conveyed by the latter in a suburban site, west of Algiers. The particulate sampling is performed by a low-flow sampler. The study shows that this suburban site, the air pollution by the TSP reached relatively low levels. We note that 76% of the levels found do not exceed the quality objective set by the Algerian regulations and the WHO (50µg/m3), therefore, the TSP pollution is tolerable, with an average of (40µg/m3). The analysis of metals associated with TSP by nuclear methods: Instrumental Neutron Activation Analysis (INAA) and the Energy Dispersive X-Ray Fluorescence (EDXRF) technique. INAA shows the presence of Cr, Pb, As, Ni, Fe, V, Zn, Mn, Se, Co, Sb, Ba, Br, Sc, Hf, Ce , Ca and Cu accusing relatively low values compared with limit values that regulate this form of pollution and the values published in the same line of research, but the inhalation of certain metals such as lead, chromium, nickel, arsenic, vanadium, manganese, cobalt and antimony even in small amounts may lead to toxic concentrations by accumulation effect in the body.

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Introduction

Several studies consider that air pollution and its effects are recognized as important environmental problems, among the authors we cite Rattapon and Darryl (2019), Brunekreef and Holgate (2002), Pope and Dockery (2012) and they consider that urban particulate air pollution has a major impact on the quality of life and human health. Indeed too this, Van Wijnen and Van der Zee (1998), Ma *et al.* (2017) confess that more and more people, not only those who live in big cities and along highways, but also people living on the outskirts and next to highways, chronic respiratory symptoms, decreased lung function and asthma.

However, determining the chemical composition of a particular material is essential to know its emitting sources and its potential effects on the environment and human health. According to Fang *et al.* (2006) and Liu *et al.* (2008), the air quality of suburban and industrial environments can be strongly affected by atmospheric emissions pollutants. In addition, Hsu *et al.* (2005) and Uematsu (2010) confirm that the characterization of TSP can provide information for the study of the atmospheric contribution of many essential biogeochemical trace metals.

Currently, the studies of air pollution have reached a new level due to the possibility of detecting new classes of toxicants: nano- and micro-sized particles. The microparticles have a higher toxicity and possess the ability to penetrate cell membranes and then to circulate and accumulate in organs and tissues of the human body. TSP and the metals associated with it can cause health problems, including burning eyes and nose, itchy sore throat and respiratory problems.

On a global scale, great progress in understanding the effects of air pollution on health has been made. In this subject, the principal concepts on the problem are introduced in many studies. The primary contributions to atmospheric pollution in the form of solid aerosols come from two sources; the first is natural sources such as dispersion of crustal material, volcanic eruptions, aerosols of marine origin and pollens. The second is anthropogenic sources such as combustion of fuel oil and coal, combustion of fuels related to vehicular traffic, and various emissions related to industrial and human activities.

According to David M *et al.* (2016), the knowledge of the chemical composition of particulate matter helps to estimate the impact of various pollution sources on the ambient air quality. So, traffic-related air pollution has also increasingly been identified as an important contributor to adverse health effects of air pollution. Swietlicki *et al.* (1996), affirm that a Long term exposure to toxic trace metals such as As, Cd, Cr, Cu, Ni, Zn, Pb, and Hg even at low concentrations can be deleterious to the human health as well as the ecosystem, and according to Gamble *et al.* (1996) and Hongyan *et al.* (2017), these can lead to cardiovascular diseases and respiratory disorders (Gamble *et al.*, 1996). Air quality today is an essential environmental concern. For example, in China premature deaths due to air pollution have exceeded 1 million people a year in recent years. In Algeria, Only a few studies have been conducted to study the air pollution. So the shortage on the data to characterize air quality makes the impact of this form of pollution on the environment very badly known. This work has been devoted to characterize the Total atmospheric Suspended Particles (TSP) considering a study site in the west of the Algerian capital (Algiers). The elemental composition in these particles was determined using Instrumental Neutron Activation Analysis (INAA) and the Energy Dispersive X-Ray Fluorescence (ED-XRF) technique.

Materials and methods

Study site

The measurement site presented in Fig.1 is situated in city of Draria. The region of Draria is located in the west of Algiers at 36°43'47" N latitude and 3°03'28" E longitude, and 200m altitude above sea level. It has approximately 40 000 inhabitants, sampling campaign was carried out from May 2010 to September 2010,the sampling point was placed at a height of 13m above ground level and about 10km southwest from the center of the heavy-traffic intersection as shown in Fig. 1.

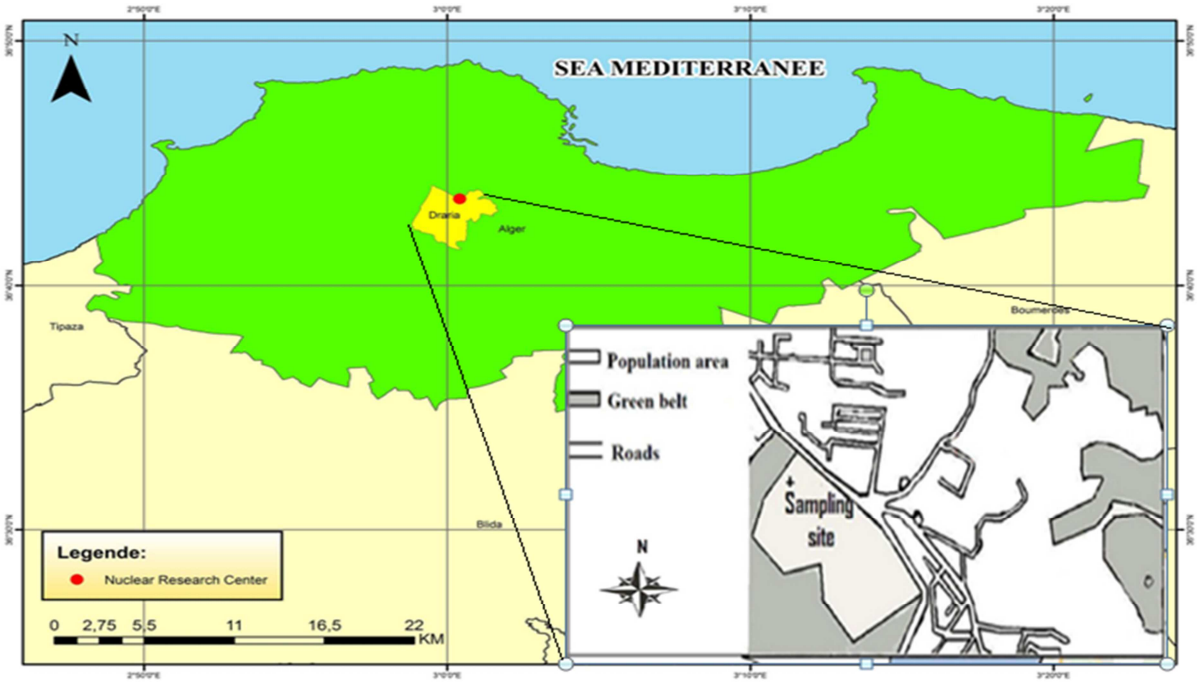


Fig. 1. ARCGIS10.2 and map image showing the location of the sampling site.

Air particulate samples were collected on 37mm diameter 0.8 µm pore size, cellulose ester filter papers, by pump aspiration, with a collection rate of 1m³ for 1 h.

The method of collection on a filter was developed specifically for measuring the mass concentration and chemical composition of the aerosol. Its principle is to collect on a filter media a representative sample of the aerosol aspiration of a known volume of air. The sample usually consists of a sampling head, a filter holder containing the filter medium, a device for measuring the air flow and a pumping system (Delmas, 2005).

Analytical instruments and analysis

We determined the levels of TSP by the gravimetric method and a total of 53 aerosols samples were weighed before and after sampling with an analytical balance. The statement of the filters is manual, dusty filters must be weighed in the same conditions of temperature and humidity as blank filters without loss of material during handling. After collection and weighing, we introduced the sample in a sealed Petri dish which is stored in desiccators at room temperature. Knowledge of weight deposited, the duration of sampling and the air average volume, we calculated the concentration of particulate matter expressed in µg/m³.

All the procedures were strictly quality controlled to avoid any possible contamination.

To analyze the metals that are carried by the TSP, we used two nuclear techniques: Instrumental Neutron Activation Analysis and the Energy Dispersive X-ray Fluorescence. The big advantage of these techniques lies in the analysis of the sample directly in its solid form. They avoid a step of solution which is long, costly and may distort the sample.

The very high sensitivity for many elements, the multielement capability and the possibility of performing instrumental analyses of very small samples are the main requirements of the instrumental neutron activation analysis.

In the case of trace elements determination in aerosols, this nuclear analytical technique offers its best performance and is widely used in this type of studies (Alian and Sansoni, 1990, Gallorini, 1995; Ondov and al, 1995; Landsberger and al, 1997).

In this study, collected samples were irradiated with thermal neutrons at the NUR research reactor at the Nuclear Research Center of Draria. The measurements were carried out using a high purity Ge detector, aerosols samples were performed in the reactor at a neutron flux of $2*10^{13}$ n /cm^2/s during 6 h for long and medium half-life radioisotope determination (Almeida and al, 2006), which operates at a power of 1 MW.

A counting system is used for the gamma spectrometry then, the raw spectral data with other required experimental parameters are processed with the computer program GENIE 2000.

But, in this study, with INAA technique, we cannot quantify certain elements such as Pb, Ni, Mn and V that are of particular interest as atmospheric pollutants because of unavailability of a pneumatic sample transfer system which used for short irradiations, so to determine these elements; we used the energy dispersive X-ray fluorescence.

It is a technique well established in qualitative and quantitative elemental analysis for the scientific researches. The X-ray fluorescence (XRF) technique is a non-destructive quick measurement, rapid sample preparation and the preparation can be automated. ED-XRF analysis has unique advantages, allowing for non-destructive elemental analysis at ambient air pressure (Cesareo and al, 1998; Bernasconi and al, 2000; Ferrero and al, 2002; Misra NL and Mudher KDS, 2002; Al-Merey and al 2005; Padilla and al, 2006; Tsuji and al, 2006).

Collected samples were irradiated with radioactive sources and re-emit a photoluminescence characteristic of elements present in the sample in the field of X-ray.

Two radioactive sources were used for the atomic excitation of different samples: Cadmium 109 and Iron 55. Each spectrum from the ED-XRF was analyzed by the QXAS/ AXIL software package (Zhang *et al.*, 2010).

Results and discussions

Time variations of mass concentration of TSP are shown in Fig. 2 moderate TSP concentration was observed, they were ranging from 15 to 85 (mg/m^3). The mean value for the period of this study was 40±15. The measurements were conducted during the period which had summer weather conditions with high temperature and infrequent rain. Precipitation would reduce resuspension of particles, for example road dust. The temporal evolution of the levels of total suspended particulates shows that 76% of the values obtained are below the target value Algerian (50µg/m^3) and were all below the limit defined by United States standard of TSP (250µg/m^3).

Based on these standards, we see that TSP pollution of the sampling period which runs from mid May to end September 2010 is tolerable and not worrisome with an average of (40µg/m^3).

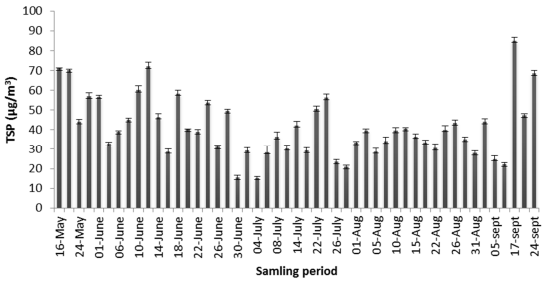


Fig. 2. Time variations of mass concentration of TSP at the Draria sampling site.

According to several scientific studies, the TSP content varies significantly between different world regions. For example, in China TSP concentrations in the air reach annual averages of the order of 402 (µm/m^3) (Lucy T *et al*, 2017). In Mexico, at the Santa Catarina and San Nicolas stations, the TSP concentrations that were revealed were of the order of 84.73 ± 12.85 (µg/m^3) and 83.63 ± 15.26 (µg/m^3) respectively (El Desouky M, 1988).

If we take another example which concerns the regions whose climate is Saharan: the countries of golf, one found that the concentrations in TSP are very important, case of Kuwait 1 572 000 and 9 160 000 (ng/m^3) (Beijer and Jernelöv, 1986).

Wind speed plays an important role in dispersal conditions. We note that low concentrations of particles most often coincide with high wind speeds (better dispersion of atmospheric particles), but certain concentrations seem to increase as the wind speed increases. The reduction is easy to understand by a greater turbulence, therefore a better dilution of the pollutant. The increase in concentrations with the wind speed can be explained by a contribution of pollutant, mainly from wind erosion and the local suspension of fine particles in the atmosphere.

The period is characterized by high average temperatures, exceeding 20°C on most days of sampling. The high temperatures favor the photochemical formation of secondary particles. Most of these days there is zero precipitation, which does not result in the leaching of the atmosphere. We note that despite this combined effect of these two factors, the temporal evolution of the contents of total suspended particles, shows that 76% of the values obtained remain below the Algerian target value and WHO standards (50µg/m³ and 100µg/m³) and remain far from the daily limit value recommended by WHO and USA for TSP (250µg/m³). On the basis of these standards, we note that in Draria, pollution by TSP over the sampling period which extends from mid May to the end of September 2010 is tolerable and is not worrying with an average of (40µg/m³). Box and whisker plots of mass concentrations range in (ng/m³) of 18 elements, namely: As, Ni, Pb, Mn,Cr, Ca, , V, Fe, , Cu, Zn, Se, Br, Ce, Co, Sb, Ba, Sc, and Hf quantified in TSP samples are given in Fig. 4.

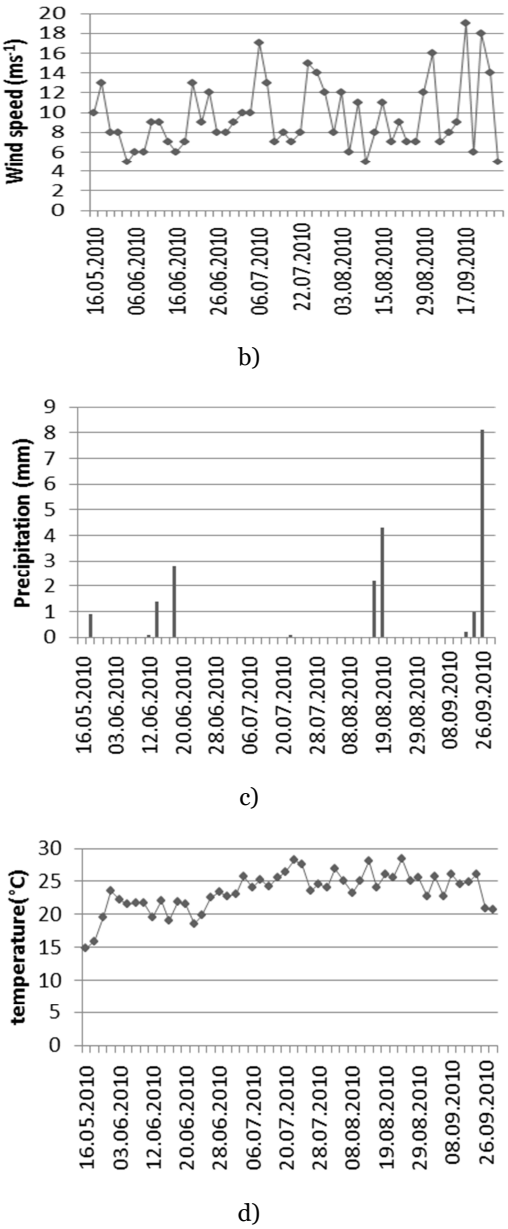
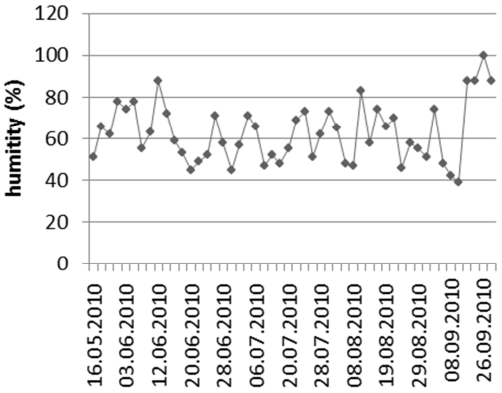


Fig. 3. Meteological data during the sampling period: (a) relative humidity (%), (b) wind speed (m/s), (c) cumulative precipitation (mm), and (d) temperature (°C).



a)

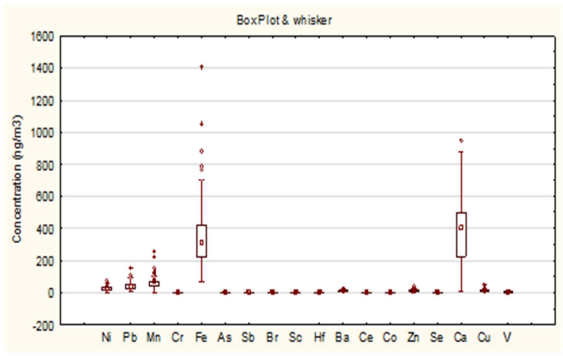


Fig. 4. Box and whisker plots of mass concentrations at Draria city nearby Algiers (n = 53 samples).

The major part of the elements have determinate by INAA while all lead, nickel, vanadium, manganese, calcium and copper measurements were carried out by EDXRF. Among the various species present in the particulate matter, a great attention has been devoted since many years to the study of the elements with higher toxicity and great diffusion in the environment (As, Cd, Cr, Hg, Ni, Pb, etc.) because of anthropogenic (road traffic, domestic heating, industrial emission) and natural (air mass transport, volcanic eruption, desert, etc.) pollutant emissions. In particular, the metals differ from other toxic substances because they are neither created nor destroyed by humans (Shakya *et al.*, 2017).

From this table, it can be seen that the mean measured concentrations of Pb, Mn and As are below both the World Health Organization (WHO) and European Directive (ED) guideline limits (250ng/m^3; 1000ng/m^3 and 6ng/m^3 respectively) hence are not likely to have any adverse effects on human health, but the mean measured concentrations of Ni exceeded ED guideline limit (20ng/m^3) which was probably due to the direct traffic emissions.

Among the elements detected in TSP, calcium has the highest average, which is of the order of 395.26 ± 211.78 (ng/m^3) followed by iron with an average concentration of 372. 56 ± 253.17 (ng/m^3). In the literature, there are many works that are interested in studying the probable emetic sources of the metallic elements carried by the TSP. the probable sources of these elements are the resuspension of terrestrial dust, building materials and road traffic (Shakya *et al.*, 2018). Also, we have noticed that the Mn content is relatively high with an average of around 70.45 (ng /m^3). The probable emitting source of this element is the surrounding soil (Hueglin *et al.*, 2005).

As we have noticed the presence of Zn, Ba, Br and Cu in the atmospheric particles. According to previous studies, this can be attributed mainly to emissions from car exhausts, as well as to abrasion from brakes and tires (Guoa *et al.*, 2003; Hagler *et al.*, 2007; Wu *et al.*, 2007; Viana *et al.*, 2008). Followed by the Pb which is known as an anthropogenic metal (Janssen and al., 1997). The presence of Ni, Sb, As, Cr and V in TSP in a site like Draria will only be accepted as markers for burning heavy fuel oil, as long as there is no local industrial activity (Manoli and al., 2002; Almeida *et al.*, 2006; Moreno *et al.*, 2006; Koc-ak *et al.*, 2007; Bouhila *et al.*, 2015; Chaiwat *et al.*, 2020). The Draria sampling and surveillance site is surrounded by construction sites generating high dust pollution, which probably explains the presence of Ce and Se in TSP (Birmili *et al.*, 2006). Among the metals that we detected with the two methods namely INAA and EDXRF, we have iron and zinc (see Fig. 5 and 6).

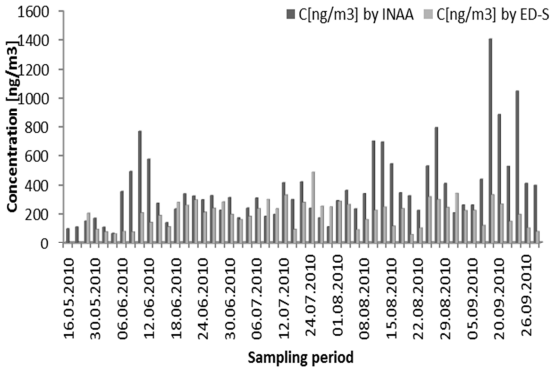


Fig. 5. comparative representation of the iron concentration detected by INAA and ED-XRF during the sampling period.

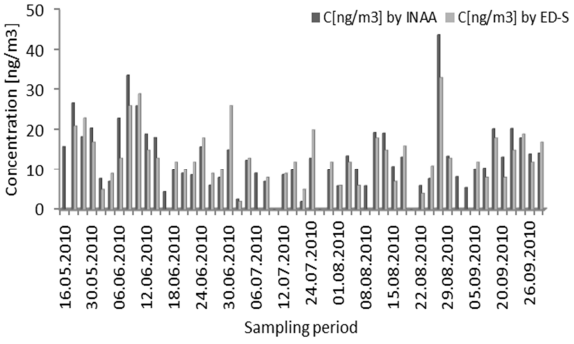


Fig. 6. comparative representation of the zinc concentration detected by INAA and ED-XRF during the sampling period.

From the results obtained, we find that iron is present in all samples at relatively high concentrations, this observation is valid whatever the analysis technique used, but we note that the neutron activation reveals

higher concentrations that ED-XRF with an average of 372 (ng/m3), in parallel, 206 (ng/m3) is the average recorded by ED-XRF, this means that neutron activation is the most sensitive technique to detect this element. Even in the absence of national or international guide value, these levels stay very high and undoubtedly pose a risk to human health during chronic exposure.

The probable source of this element is natural terrigenous, therefore, the contamination of this site could come from the phenomenon of wind erosion and the suspension of soil particles in the atmosphere during drought, but according to Birmili (2006), iron can also come from road traffic (brake wear) (Sandrine *et al.*, 2005).

The means of zinc concentrations obtained by the two analysis techniques, namely neutron activation and ED-XRF are 12.66 and 11.51 (ng/m3) respectively.

The absence of national and / or international standards makes it difficult to assess the level of air pollution by this element, but in comparison with the results of some work published internationally these values remain low and tolerable.

According to several studies and in the same line of research (Pakkanen *et al.*, 2001) zinc is among the elements that characterize an anthropogenic origin. Indeed, in a region with little industrialization like Draria, the presence of zinc in the air may be directly associated with road traffic and According to Pakkanen *et al.*, 2001, it would appear that Zn is emitted significantly during combustion phenomena in addition to being contained in brake and tire linings (Belamri *et al.*, 2009).

From the results obtained, we note that the level of detection of zinc by ED-XRF in TSP is on average of the same order of size as that observed by neutron activation. Generally, the detection sensitivity of zinc in TSP for the two analysis techniques is good. Due to the absence of guide and limit values for certain metallic elements, we have compared our results with those found by other studies which are also interested in studying the average pollution levels conveyed by TSP in other regions of certain large cities of Europe, China and India at different sampling years, results from the same site and in central Algiers (table 2).

In our case, the average total metal concentrations for Cr, Cu, Zn, Fe, V, Se, Co, Sb, Ba, Ca, Br, Sc, Hf and Ce were generally lower than the values previously measured at different sites around the world.

The results are presented in Table 1. Overall, Mn, Fe, Pb and Ni are the most dominant trace elements, but the ranks in concentration of other trace elements are different from one country to the next.

It is clear from the results in Table 2 that the concentration levels in the city of Draria during this study are lower than those measured in 2011 and they are still lower than those obtained in Algiers center.

It is clear from the results in Table 2 that the concentration levels in the town of Draria during this study are lower than those measured in 2011 for the elements Fr, Br, Hf, Co, Mn and V. however, the concentrations of Cr, As, Sb, Sc, Ce, Zn, Se and Ca are higher than those obtained in 2011.

On the other hand, the metals Ba, Cu, Pb and Ni are not detectable in 2011 for this same site. If we compare the concentrations revealed in Algiers-center are high compared to the results found in Draria namely Fe, Cr, Sb, Hf, Co and Se. It shows that the concentration levels of Fe, Br, Zn, Ca and Cu revealed in Goteborg (Sweden) are higher than our results, exceptionally for Mn.

We note that the pollution levels recorded in Copenhagen (Denmark) in As, Sb, Ba, Ce, Co, Se, Cu and V are very high compared to those recorded in Draria which are on a tiny scale. These elements characterize the industrial zones and Draria is a poor region in terms of industry. This is also the case in Chandigarth (india) where we notice extremely high levels of pollution compared to Draria.

Table 1. Trace elemental concentrations (ng/m³) in air particulate matter in the study area (n = 53 samples) and other world regions.

	Present study 2010	Draria city 2011[a]	Algiers city, 2008[b]	Goteborg *2002*[c].	Copenhagen, *2007*[d]		Chandigarth, *2000* [e].	China, 2016 [f]	
								Dalian	Xiamen
Cr	2.06	1.43	35.4					10.4	7.6
Fe	372	540	1380	1190	18.5	8.69	20277	2376.2	302.0
As	0.29	0.19			27.5	38		28.9	16.7
Sb	0.80	0.71	2.17		123	40			
Br	0.31	2.39		9					
Sc	0.20	0.15							
Hf	0.03	0.04	0.067						
Ba	11.35	/			703	540		116.4	38.5
Ce	1.41	1.30			8.5	4.5			
Co	0.23	0.71	0.64		13	20.8			
Zn	13.41	10.0		49	2.88	4.5	1209	362.5	263.7
Se	0.25	0.15	0.74		32.9	36.8			
Ca	395	270		740	13.37	8.6	13695	4437.7	348.4
Mn	70.47	425		20	295	198	718	53.0	14.9
Cu	14.12	/		16	168	1.56	30	147.0	35.6
Pb	44.07	/					1497	124.4	45.0
Ni	27.06	/			8.6	19.87	229	43.7	6.8
V	2.95	4.36			316	428		19.6	8.5
	Suburban site (TSP). (2010).	Suburban site (TSP). 2011	Urban site (TSP). (2008)	Urban site (TSP). 1999.	Site of proximi-ty (TSP). 2005	Urban site (TSP). 2005.	Urban site (TSP). 1996.	Urban site (TSP). 2004	Urban site (TSP). 2004

[a] (Bouhila *et al.*, 2015) ; [b] (Pakkanen *et al* 2001) ; [c] (Belamri *et al.*, 2009) ; [d] (Anoop *et al.*, 2007) ; [e] (Bandhu *et al.*, 2000) ; [f] (Ma *et al.*, 2017).

In China, the metal concentrations conveyed by TSP are too high by comparing them to those revealed in Draria, certainly because of the different industries which characterize China compared to Algeria which remains far for this type of investments, exceptionally for Mn where its concentration is lower than that obtained at Draria.

So, in a mediterranean environment, suburban and little industry as Draria, the main aerosol particle sources are suspended soil dust which was indicated by the presence of major elements as Fe and direct traffic emissions which were indicated especially by the presence of Pb and Ni in the measured elements, therefore, the population living in proximity to the urban area is exposed to relatively low levels of air pollution and generally, air mass transport, traffic and badly maintained automobiles contribute most to urban pollution problems in Algiers.

Consequently, in a suburban site and an area with little industry as Draria, metal components can be derived from within and / or outside of the town, as these elements move over long distances and from Sandrine (2005), the journey ends metals according to the kinetic processes in different weather conditions, by filing in the vicinity of emission sources[0-10 Km] for the metals associated with coarse particles, and a deposit that is far from the emission sources [from 10 to 1000 km] to the elements associated with small particles (Sandrine *et al.*, 2005).

Conclusion

In this study, we assessed the levels of ambient air pollution by total suspended particulates and certain metals that are transported by them.

The study shows that this suburban site, the air pollution by the TSP reached relatively low levels, we note that 76% of the levels are bellow both the Algerian and WHO guideline limits (50µg/m³, 250µg/m³ respectively). The average grade of the sampling period (continuous monitoring) is 40 (µg/m³), the maximum is about 85 (µg/m³) and compared with levels measured in urban sites of Algiers, pollution levels recorded on Draria are not worrying, this means that the air quality improved by moving away from large cities. The average total metal concentrations for Pb, Mn, As, Ni, Cr, Cu, Zn, Fe, V, Se, Co, Sb, Ba, Ca, Br, Sc, Hf and Ce were

generally lower than the values previously measured at different sites around the world and do not exceed the values that regulate this form of pollution internationally, but inhalation of certain metals such as Arsenic, Cobalt, Lead, Nickel, Chromium, Manganese and Vanadium in spite of small amounts, may lead to toxic concentrations by accumulation effect in the human body.

Acknowledgments

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Author Contributions

L.H. (Lecturer) provided assistance on the nuclear techniques used.

Z.B (researcher) carried out the experiments of INAA.

S.L. (Lecturer) wrote and revised the manuscript.

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