AUSTRALIAN JOURNAL OF BASIC AND APPLIED SCIENCES

ISSN:1991-8178 EISSN: 2309-8414
Journal home page: www.ajbasweb.com

Assessment of air pollution by heavy metals in the urban center of Algiers

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ARTICLE INFO

Article history:
Received 18 January 2017
Accepted 28 March 2017
Available online 15 April 2017

Keywords:
Air pollution, Total Suspended Particulate, Heavy metals, PIXE, INAA, XRF

ABSTRACT

Background: Growth of population number and industrial units in parallel of increase of terrestrial, maritime and air transports are indicators of a pressure on the environment in particular on the air quality. The intense human activity added to the natural activity leads to the generation of aerosols which are composed of particles of very varied sizes. In particular, fine dust which constitutes a significant factor of pollution involving a deterioration of the air quality. The urban zone needs clean air to assure a public health. To achieve this goal, we need to enrich our knowledge on the sources and the impacts of the atmospheric pollutants. Understanding the sources, the composition and the mode of transformation of the atmospheric pollutants is the first step in air quality monitoring. **Methods:** The study and control can be achieved by using adequate nuclear techniques analyses. The aim of this work is to study air pollution in urban zone of Algiers for the evaluation of the contents of heavy metals in various selected sites. A sampling campaign was carried out during period 2005/2007 in three sites in the Algiers city: Bab El-Oued the High Casbah and Mustapha Bacha**.** In order to identify and quantify their concentrations, nuclear techniques such PIXE (Proton Induced X-ray Emission), ED-XRF (Energy Dispersive X-ray Fluorescence) and the INAA (Instrumental Neutron Activation Analysis) are used in this study. These techniques are powerful tools used in air pollution monitoring for the determination of trace heavy metals**.** We identified at the same time 18 elements (Na, Sc, Cr, Fe, Co, Zn, Se, Ag, Sb, Ce, Hf, Ta, Hg, Cu, Pb, Ni, Cd). **Results**: the concentrations of the elements Hg, Co, Ni, Cr and Ag vary around 2 ng/m^3. The concentrations of element Zn vary over a wide range from 3 ng / m^3 to more than 500 ng / m^3. A large values of Pb were measured (5000 ng/m^3, 8000 ng/m^3) and 200 ng/m^3 for Cu. The element Fe appears in all analyzed filters. The concentrations range from low level to high level (8000 ng/m^3)**. Conclusion:** the high level measured concentrations show the significant air pollution in urban areas. The major emission sources identified were classified as sea spray (SS), suspended soil and vehicle emissions (vehicle emission VE, diesel and petrol). This analysis allowed us to deduce some significant sources affecting the receptor sites. Among these pollutants one finds heavy metals contained in the suspended matter.

INTRODUCTION

Algiers capital of Algeria knows a terrible growth of its automobile park. Each year approximately 200,000 to 300,000 units comes to be added to the 2,500,000 vehicles already existing, creating a situation for which neither the state nor the population can manage its immediate and short-term consequences. One will not have forget the other sectors of agriculture and industries which know a significant growth rates and paradoxically accentuate the environmental pollution and contribute to degradation of the air quality. Some air pollution studies in Algiers city have been done (Markowicz and Haselberger, 1992, Kerbachi *et al*., 2004, Belamri and Benrachedi, 2010, Yahiaoui *et al*., 2012). These studies showed the evidence existence of air pollution which can cause harm to the environment. It may present a threat to Algiers's ecosystems and the

ToCite ThisArticle: Mr. Belamri, L. Bounemia, A. Azbouche, K. Boukeffoussa, C. Lakhdar Chaouch., Assessment of air pollution by heavy metals in the urban center of Algiers. ***Aust. J. Basic & Appl. Sci.,*** *11(5): 35-44, 2017*

health of the citizens. Therefore, it is important to proceed collecting intense data in order to monitor more precisely the air quality.

Troposphere, low part of the atmosphere, is characterized by a strong socio-economic activity accompanied by a serious deterioration of the air quality of this layer due mainly to the rejection of the pollutants. Clean air is a prerequisite to human health and welfare. In spite of the introduction of less polluting technologies in industry, energy production and transport during the past decades, air pollution remains a major health risk (HTAP, 2010). Air pollution causes evident poor air quality. This pollution can harm the environment.

Air pollution studies done in Algiers city (Gondala *et al*., 2009, Elichegaray, 2008, Paul and Pillai, 1978) have shown existence of air pollution contributing to the degradation of life quality in the city. It has become urgently to take care of the problem of pollution by heavy metals in the city of Algiers. This work fits in this context to quantify air pollutants by nuclear techniques available in our laboratory in order to measure the concentrations of heavy metals in the suspended particulate.

For the evaluation of the contents of heavy metals in various selected sites in Algiers a sampling campaign was carried out during period 2005/2007 in three sites in the Algiers city: Bab El-Oued, the High Casbah and Mustapha Bacha Fig.1. In order to identify and quantify their concentrations, nuclear techniques such PIXE (Belamri and Benrachedi, 2009), EDXRF (Avino *et al*., 2008) and the INAA are used in this study. These techniques constitute powerful tools to achieve this goal Compared to SAA of air pollution analysis (Techato and Van Beem, 2014) A total of 53 samples have been collected from three sites, 25 at Mustapha Bacha, 16 at Haute Casbah and 12 at Bab El–Oued). The samples were analyzed by three techniques as follow:

- 25 samples by PIXE technique;
- 12 samples by EDXRF technique;
- 16 samples by INAA technique.

Fig. 1: Localization of sampling sites

MATERIALS AND METHODS

2.1. Sampling area description:

In order to assess the heavy metal content in the atmosphere at the capital Algiers, the Ministry of the Environment and the Nuclear Research Center of Algiers (CRNA) conducted since 2005 a sampling campaign at different stations located in Frantz Fanon, CHU Mustapha Bacha and the CHU Bab El Oued.

The sampling site located in Frantz Fanon at the high Casbah of Algiers is at the heights of 200 m above the sea level surrounded by two heavy traffic roads.

The site of the CHU Bab El Oued is in the West of Algiers about 250m far from the Mediterranean Sea with an area of 1.8 km^2 characterized by high density population (1000,000 inhabitants). More than 200,000 vehicles pass each day near the site. The hospital is equipped with an incinerator at a distance of 200m from the sampler.

The third site Mustapha Bacha hospital is in Sidi M'Hamed in the Est part of Algiers city with a high population density estimated at 220 000 inhabitants in an area of only 7.8 Km^2. The sampler is placed in the hospital ground at a distance of 20 m far from the incinerator. In the West side at 60 m Ghermoul Street and in the south at 150 m the Hassiba Ben Bouali Street. About 20000 vehicles per day enter the hospital of Mustapha Bacha.

2.2. Sampling methods:

The air collection system (Fig.2) used is an LVS pump aspiration (Low Volume Sampler), with a rate of 17 L/mn. We use ester cellulose air filter with 8 µm porosity and 45 mm diameter to collect the samples (Masclet, 2005, White, 1987). The mass of the TSP (total suspended particulate) is determined by weighing the filter before and after sampling (Fig.3).

Fig. 2: Low volume sampler LVS

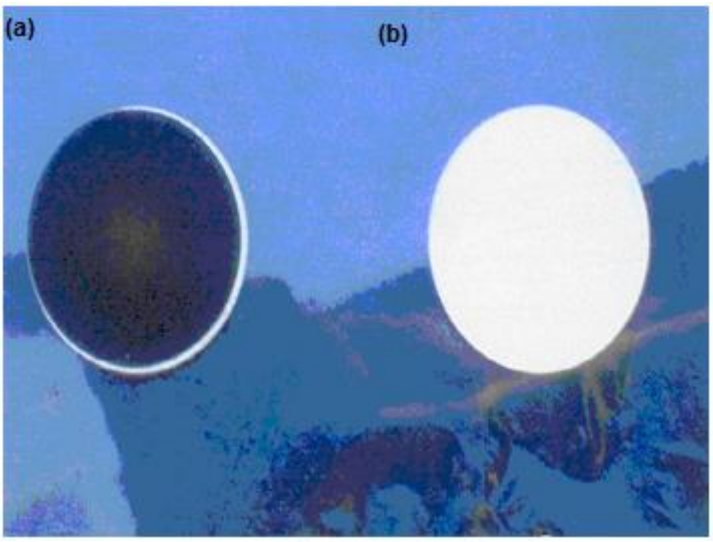

Fig. 3: Filter before (b) and after (a) sampling

2.3. Trace element determination by PIXE (Particle Induced X-ray Emission) in air filter samples:

This technique is based on the measurement of X-rays characteristic of the element produced after the atomic transition of the internal (K, L or M) shells following ionization of the target atom by charged particles (Chpolski, 1977)**.** The relation between the element of atomic number Z, and X- ray Energy, is given by the following equation:

$$E = \frac{2\pi m_0 e^4}{h^2}(Z-\sigma)^2\left(\frac{1}{n_1^2}-\frac{1}{n_2^2}\right) \quad (1)$$

Where n_1, n_2 are principal quantum numbers of the initial and final states, Z is atomic number, h is Planck's constant, m_0 and e are respectively, mass and electron charge and σ is the screen constant.

The protons beam is delivered by a Van De Graaf accelerator. On the outlet side of the accelerating tube, the beam of protons is deviated at 90° by analysis magnet. The beam passes then through quadripolar lenses before crossing two graphite collimators. X-rays emitted after the bombardment of the sample are detected at

135° angle from the direction of the incidental beam. A semiconductor Si(Li) detector with resolution of 155eV at energy 5.9 keV of ^{55}Fe radioactive source, manufactured by CANBERRA (Canberra Industries), was used in this work for sample measurement. The impulse delivered by the detector is then amplified and recorded in the form of a spectrum by an electronic chain.

The treatment of the spectra is carried out by AXIL software. It allows the identification of the elements and the calculation of the area which is used for the quantification of the concentration.

To quantify the concentration of the elements identified in the sample, we use the comparative method given by equation (2), which requires the irradiation of the sample and the external standard under the same conditions:

$$C_e = \frac{I_e}{I_s}.C_s \quad (2)$$

Where:

C_e is concentration of the element in the sample,

Cs is concentration of the element in standard,

Ie, Is are respectively the intensity of the peak of the element in the sample and in the standard. The standards used are pellets IAEA standards and deposits of standard solution on ester cellulose filters.

2.4. Trace element determination by XRF in air filter samples:

The spectrometry of x-ray fluorescence is a technique which use X-rays characteristic emitted by the elements targeted in the sample (Chpolski, 1977). The intensity of the peaks related the identified elements indicate the quantity of each element in the collected sample. This technique was applied for the quantification of the heavy metals contained in the air filter samples collected in Bab El-Oued site. The samples and the standards are irradiated by a source of Cadmium 109 for lower energies and by a source of Americium 241 for higher energies. The radiations emitted by the samples are collected using a semiconductor detector Si (Li) during 2000 s in order to obtain a sufficient level counting for statistical consideration.

2.5. Trace element determination by INAA in air filter samples:

The trace element concentrations were also determined by INAA according to the procedure fully detailed in Revel and Ayrault (2000). Instrumental Neutron Activation Analysis is a sensitive technique and nondestructive in the majority of the cases **(**Djingova and Kuleff, 2000) The sample is irradiated by a neutron source in a reactor, during this process the stable elements become radioactive then decrease by emitting gamma rays throughout a half-life varying from the second to years. Their analysis allows us to know elements present in the samples and measure their concentrations**.** The concentrations are always calculated by the relative method by using external standards irradiated with the sample under the same conditions:

$$\frac{I_x}{I_s} = \frac{C_x.\left(e^{-\lambda t_d}\right)_x}{C_s.\left(e^{-\lambda t_d}\right)_s} \quad (3)$$

Where the x and s indicate respectively the sample and the standard, the C_X and C_S are indicate respectively concentrations of the element in the sample and in the standard, the I is counting under the photoelectric peak and λ is the decay constant of the element.

Two procedures were performed to measure the concentrations of the heavy metal elements. The first one was applied for determining elements producing short half-life radioisotopes like: Al, Cl, I, Mg, Mn, Ti and V and the second one for determining the elements producing long half-life radioisotopes like: Ag, As, Au, Ba, Br, Ca, Cd, Ce, Co, Cr, Cs, Dy, Fe, Hg, K, La, Mo, Na, Rb, Sb, Sc, Se, Sm, Sr, Th, U, W, Zn and Zr.

For the procedure one, the filter was folded with polyethylene twisters, sealed in an ultra-pure polyethylene bag, and irradiated individually for five minute in a thermal neutron flux ($1.3*10^{13} n.cm^{-2}s^{-1}$) at the experimental nuclear reactor of Algiers.

For the second procedure, the filter parts were folded into ultra-pure aluminum thin foil. Thirty filters were irradiated simultaneously for 6 h at a thermal flux of 2.310^{13} $n.cm^{-2}.s^{-1}$.Successive gamma radioactivity measurements were then performed.

To quantify pollutants in the collected filters, standard filters were prepared by deposits targeted elements in the solution.

Results:

Average TSP concentrations of urban site from year 2007 comparison with the limits are shown in Fig.4. In the evaluation of the air quality of urban center of Algiers, it was concluded that most of the loaded filters exceed the Algerian pollution limits for total suspended particles (Official Gazette, No. 1, 2006).

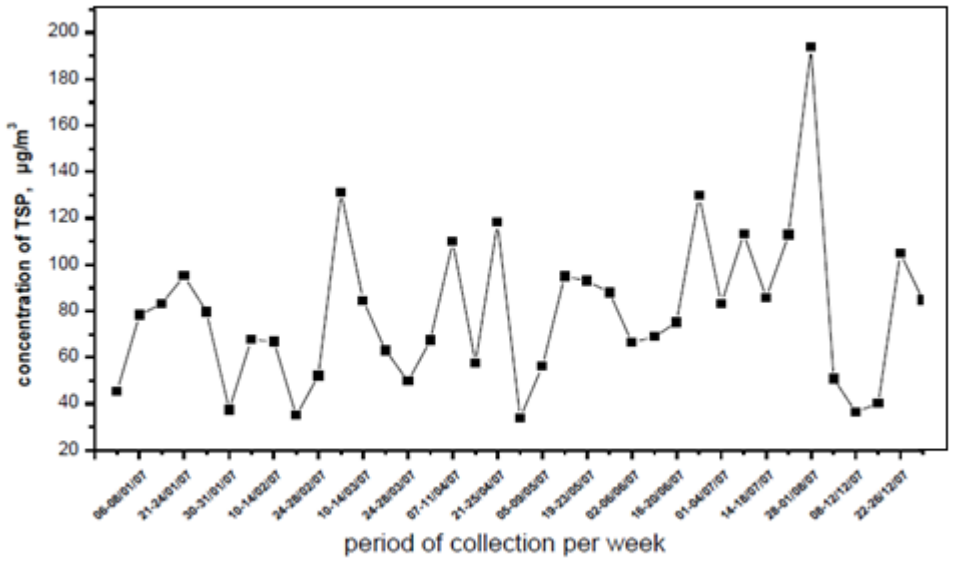

Fig. 4: TSP averages from year 2007

A spectrum PIXE (Fig.5) collected after irradiation of the sample shows elements identified. The concentrations of the quantified elements are represented in Fig.6.

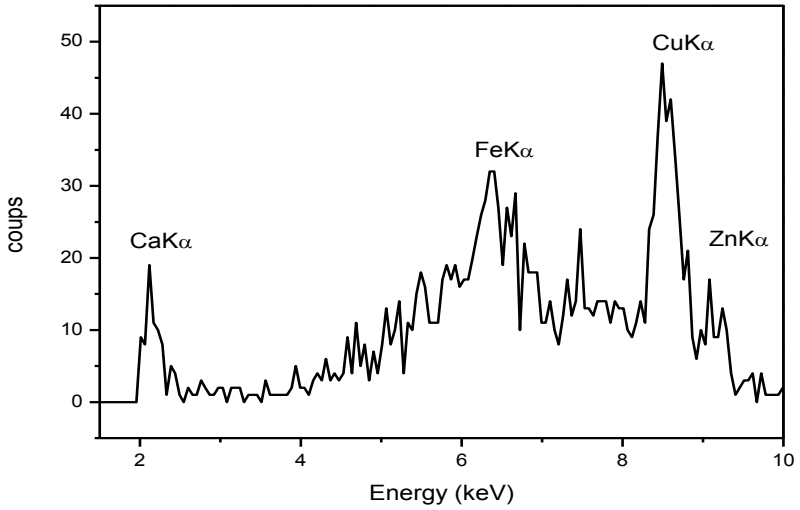

Fig. 5: Spectrum PIXE of air filter sample at 2 MeV protons beam energy.

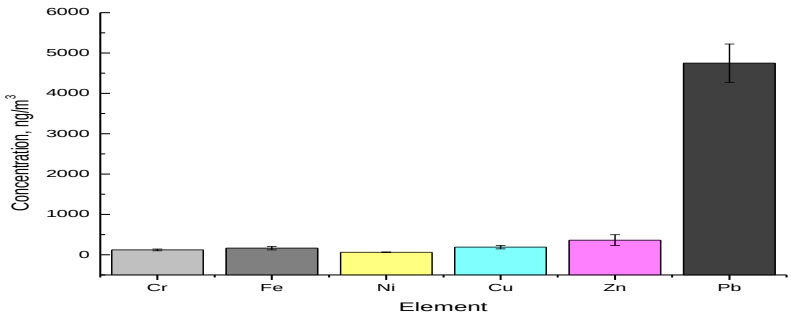

Fig. 6: Concentration of the elements analyzed by PIXE in air sample filters collected at the Haute Casbah.

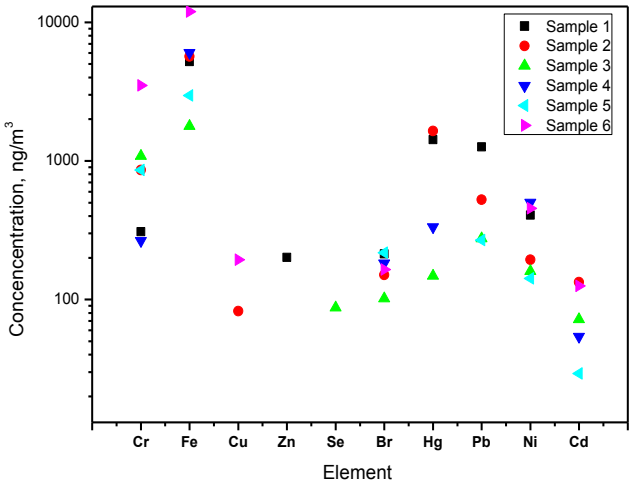

Fig. 7: Concentration of elements analyzed by EDXRF in air filter samples collectedat CHU Bab El Oued.

Elements such as: Na, Mg, Cl, Sc, Cr, Ti, V, Fe, Co, Cu, Zn, Br, Ag, Sb, and Hf, Ta and Hg were measured on air filter obtained using the INAA technique Fig.8,9. The collection of the gamma spectrum was carried out using a gamma spectrometry chain associated with a high purity germanium detector HP(Ge)**.**

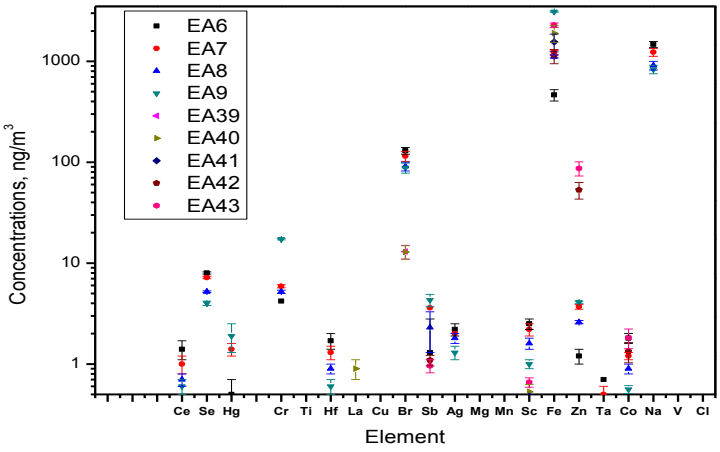

Fig. 8: Concentration of elements in air filter sample analyzed by INAA collected at the Haute Casbah.

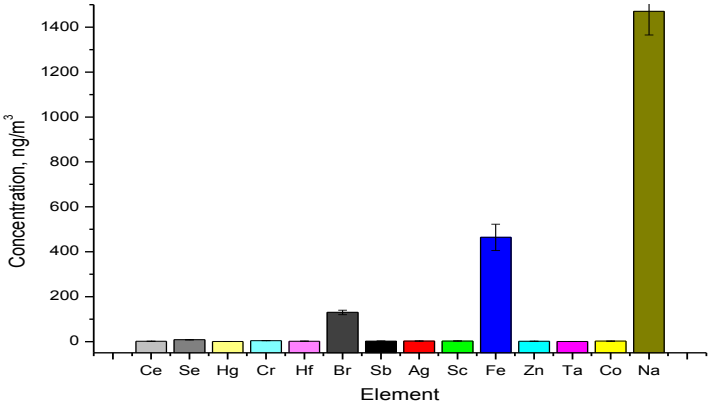

Fig. 9: Concentration of elements analyzed by INAA in air filter sample of air (Irradiation time 6h) at the Haute Casbah.

INAA, PIXE and EDXRF allowed us to identify several elements in the air filter samples collected in the studied sites in Algiers city. More than 20 elements were measured. These elements are considered as heavy metals contribute to the degradation of the air quality **(**Kayasth and Swain, 2004; Song-Miao F, 2004**).**

Antimony:

Antimony is an element relatively not very abundant in the earth's crust 0.2 ppm (Avino *et al*, 2008**).** It enters in the manufacture of the batteries (alloy Lead-Antimony).With the capacity detection of INAA one could record traces in the irradiated samples. We noted that in the irradiated samples during 6h relatively higher concentration contents (4.3 ng/m^3) are measured.

Cadmium:

Relatively rare element in the earth's crust (0.16 ppm), cadmium in nature is found generally associated with zinc (in the form of sulphide) or lead. In our case, we measure a very weak concentration in the air filters analyzed by the PIXE, a maximum of 189ng/m^3 in the case of the XRF, with an average of 61ng/m^3**.**

Iron:

Iron is quite present in high quantity in the samples of the site of Bab El-Oued, with a maximum of 1228 ng/m^3, an average of 657ng/m^3. We can note high concentrations found we use INAA technique.

Magnesium:

Is the sixth element by order of abundance in the earth's crust (2.76%). We have obtained a maximum about level of 253 ng/m^3 in a sample irradiated during 5mn by INAA technique.

Mercury:

Small quantity has been obtained on the site of the High Casbah; an abnormaly amount of Hg is noted here in the site of Bab El-Oued, where we recorded a maximum of 2391 ng/m^3 with an average of 792ng/m^3. The sources of emissions are probably the cement factories and the incinerator.

Lead:

In the case of the PIXE, we measure an average of 190ng/m^3, and467ng/m^3 in the case of XRF. The incinerator of the CHU hospital of Bab El Oued is one of the probable source of pollution.

Statistical studies:

The statistical studies by correlation factors between the identified elements indicate the existence of dependence of a linear type (Table.1).

Table 1: Correlation between concentrations of analyzed elements.

	Cr	Fe	Cu	Zn	Se	Br	Hg	Pb	Ni	Cd
Cr	1.0000									
Fe	-0.0709	1.0000								
Cu	0.5975	0.3454	1.0000							
Zn	-0.0916	0.1670	0.0290	1.0000						
Se	0.0363	0.4153	0.2372	-0.3123	1.0000					
Br	0.2625	0.5301	0.0744	0.2679	-0.1452	1.0000				
Hg	-0.3236	0.3004	-0.2332	0.6237	-0.0857	-0.0233	1.0000			
Pb	-0.0931	0.4551	-0.1658	0.6912	-0.2388	0.6780	0.5843	1.0000		
Ni	0.1319	0.3148	-0.0913	0.5126	-0.3965	0.3920	0.4976	0.5290	1.0000	
Cd	0.4356	0.2139	-0.0744	-0.4218	0.1179	0.3604	0.0422	0.1465	0.2205	1.0000

The positive coefficients of correlation found between elements indicate the tendency of variation of the elements in the same way i.e when the concentrations of an element increase the other concentrations increase. Hence it is possible to demonstrate the existence of one by the other element. If the correlation coefficient is important, one might infer that these elements have a high probability of coming from the same source of pollution. Factors in our correlations are not significant in most cases; we can conclude that this is a multitude of sources of pollution.

Positive correlations factors calculated (Table 1) shows strong correlations between the following elements: (Ni, Zn), (Pb, Zn), (Cu, Cr), (Fe, Br), (Zn, Hg), (Br, Pb), (Hg, Pb) in excess of 50%. This shows the coexistence of these elements and can help us to identify pollution sources.

For negative correlations values observed for example of (Cd, Zn) and (Cr, Hg) are indicators showing the no coexistence of these elements. Increasing the concentration of one element predicts the decrease of the other. This means, that the two elements have not the same pollution sources.

Correlation with the weather parameters:

In order to check the influence of the weather parameters such as: the temperature (T), the water content (Hu), the pressure (p) and speed wind (V) on the concentrations of the heavy metals in the air, we establish the matrix of the following coefficients of correlationTable.2:

Table 2: Correlation between some elements and the meteorological parameters

Elements	T	Hu	P	V
Ti	0.4323	-0.8563	-0.8676	0.5366
Cu	0.3487	0.052 4	0.1035	0.074
Mg	0.2646	-0.2350	-0.8532	0.9397
Mn	-0.0296	-0.5984	-0.8532	0.6081
Na	0.3636	0.3032	-0.4463	0.8968
V	-0.4756	-0.6133	-0.5964	0.1399
Cl	0.3080	0.3284	-0.4302	0.8887

The informative contents of this matrix are significant. It is noticed that the presence or the absence of certain elements in the air is dependent on the weather parameters.

Factors Correlations calculated (Table 2) show several tendency in relation with meteorological parameters. We can see that the increase in the rain causes a reduction of the quantities of pollutants collected for the negative correlation factor. This shows evidence of leaching phenomena due to precipitation.

All values of correlations between pollutant concentrations and wind speed are positive. This shows that the load concentrations and the measured pollutants increase with the wind speed. The correlation value of (Cl and wind speed) R = 0.8887 and that of (Na and wind speed) R = 0.8968 indicate that the source is probably the sea spread regarding the sampling site located a few hundred meters far from the coast.

The concentration of Magnesium, is perfectly correlated (R = 0.94) with the speed of the wind. The careers containing dolomite ($MgCO_3$) are the principal transmitting sources, where wind is the means of transport of the particles Fig.10.

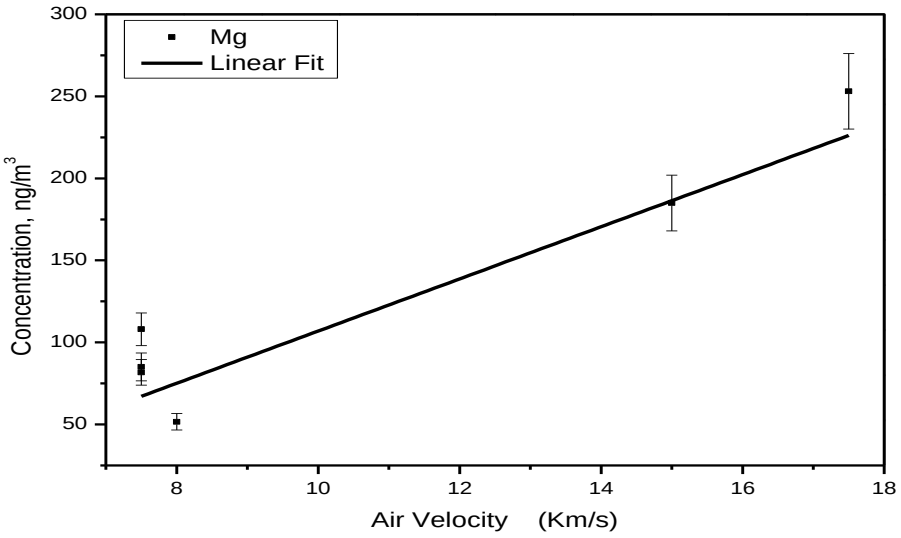

Fig. 10: Linear regression between Mg and the wind

Conclusion:

In this study we have quantified the air pollution by three nuclear techniques PIXE, EDXRF and INAA. We identified at the same time a broad pallet of elements which are due to sources of natural origin (Mg, Ca, Na) and the sources of anthropic origin (Zn, Pb, Hg, Cr). Source emissions of precursor gaseous and primary particles are highly variable dueto differences in fuel use, operating conditions, and sampling methods.

The concentrations of the elements Hg, Co, Ni, Cr and Ag vary around 2ng/m^3. The concentrations of element Zn vary over a wide range from 3 ng / m^3 to more than 500 ng / m^3. We recorded peaks of Pb with concentrations of 5000 n.g/m^3 and 8000 ng/$m^3$and for Cu 200 ng/m^3.

The Fe element appears in all collected and analyzed air filters. Iron constitutes a major pollutant in the samples. We recorded a peak of 1228 ng/m^3at the site of Bab El-Oued which is also polluted by toxic elements of anthropic origin such as Mercury, Zinc and Lead. The major emission sources identified were classified as sea spray (SS), suspended soil and vehicle emissions (vehicle emissions VE, diesel and petrol). We also try to

see the influence of the weather parameters in correlation with the concentration of certain elements in the atmosphere.

The weather parameters are regarded as an important factor which controls the concentration of elements in the atmosphere. Source and ambient measurements must be monitored in continuous time to establish source/receptor relationships. Factors Correlations calculated show several tendency in relation with meteorological parameters.

This analysis allowed us to deduce some significant sources affecting the receptor sites due mainly to a heavy traffic an industrial fabric in full expansion (oil refineries, mechanical engineering industry and chemical industry).

The other goal of this study is to use existing nuclear technique. The results demonstrate the powerful tools of nuclear analyses techniques (NAA, PIXEand ED-XRF) in the field of air pollution monitoring.

These results of urban air quality in order can be used to compare the existing national standard to international standard to make proposal guidelines to serve as a basis for protecting public health and in order to reduce air pollutant. Urban pollution results are strongly dependent on the traffic intensity, industrial areas density of inhabitants. All these factors have a harmful impact on the human bod that is very difficult to trace and therefore it is necessary to use analytic bioindicators.

Lichens have the capacity to accumulate elements and airborne particles from rain and dry deposition. This means, they can be used as biomonitors of air pollution. We expect to use in our future studies in the field of air pollution the lichens bionindicator. This species of lichens can provide a lot of information on the quality of the environment and are very useful in evaluating long-term effects of biomonitoring.

The results of the future study will be used to increase knowledge of the transfer of the pollutants in biological species. By using lichens as bioindicator we can follow the track and the dispersion of the elements in the air. Indeed, we should identify the chemical composition in order to monitor their impact.

ACKNOWLEDGMENTS

Thank to Nuclear Research Center of Algiers (CRNA) for supporting this works.

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