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Monitoring and statistical evaluation of heavy metals in airborne particulates in Cairo, Egypt

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Abstract

Airborne particulate material was monitored as total suspended particles and particulate matter less than 10 μm at selected sites in Cairo. The selected sites represent heavily industrial and industrial–residential areas of Cairo, Egypt. The filters were further analyzed for lead, cadmium, zinc and nickel using ion chromatography. The chosen method was modified to improve resolution and decrease the retention time. The data obtained were treated statistically using one-way analysis of variance and correlated with the anthropological and industrial activities of the sites. The concentrations of the heavy metals are studied in terms of particle size, time dependence, and safety.

Keywords: Air analysis; Environmental analysis; Statistical analysis; Heavy metals

1. Introduction

Quality of life is a balance between technology progress and environmental risks. This balance can be controlled by analytical chemistry, which is in a central position. Elaboration and rapid determination of anthropogenically toxic elements is urgently required [1–3].

The reliable quantitative assessment of toxic metals in environmental air specimens has led to discoveries of the vital roles of trace metals in human metabolism [4–7]. Human exposure to trace metals occurs primarily through inhalation of air and ingestion of food and water. Concentrations of trace metals in air vary considerably depending upon many factors including proximity to sources of trace metals emissions.

In addition to the obvious environmental effects of airborne particulate matter, it has been shown that particles have a direct effect on climate [8–10]. Particles can scatter and absorb solar radiation, change levels of precipitation, cause the formation of warm and cold clouds and of fog and smog.

It was estimated that more than $15\cdot10^{9}$ kg of particulate matter from anthropogenic sources are emitted into the air each year [11]. Available information suggests that approximately 5% of the anthropogenic emissions, representing over $750\,000\cdot10^{3}$ kg per year are environmentally toxic metals [12].

The primary objective of this work is to put into perspective some of the current concepts with respect to trace metals distribution and emissions in air. This will reflect the potential adverse on health effect due to human occupational exposure to trace metals. In addition, the work was oriented to compare and correlate the obtained data of trace metals concentrations in different locations within Cairo, Egypt,

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which suffer from uncontrolled release of industrial emissions. This would be useful for assessment of the air quality in Cairo compared with the recommended international standards for occupational exposure to several trace metals.

2. Experimental

2.1. Apparatus

A Dionex DX 20001, high-performance ion chromatograph equipped with a UV–Vis detector set at 520 nm was utilized. A 25-μl injection loop was used throughout. An Ion Pac CS5A analytical column containing 55% cross-linked microporous, hydrophobic resin functionalized with fully sulfonated group for cation exchange was used. This column was operated in conjunction with an Ion Pac CG5A guard column. Both CS5A and CG5A are compatible in the pH range 0–14. Oxalic and citric acids mobile phases were prepared in selected buffering solution, 0.1 *M* ammonium hydroxide mixed with 0.05 *M* potassium hydroxide, to optimize the variation of pH value of the mobile phase. Multilevels of different standards of the investigated elements were used to calibrate the instrument. The correlation coefficients ranged from 0.996 to 0.998 for each of the investigated elements. Data acquisition and processing were done using automatic AI-450 computer software.

2.2. Air particulate sampling and sample preparation

Airborne particulate samples were collected from different locations chosen to represent different types of sources of heavy metal pollution in Cairo. Ramsis, Shoubra and Tebbin South represent high traffic areas with densely industrial activities such as alloy manufacturing, lead smelting and lead battery industry, while 10th of Ramadan, 6th of October and Naser cities, represent suburban residential areas (Fig. 1).

A series of air samples were collected during the

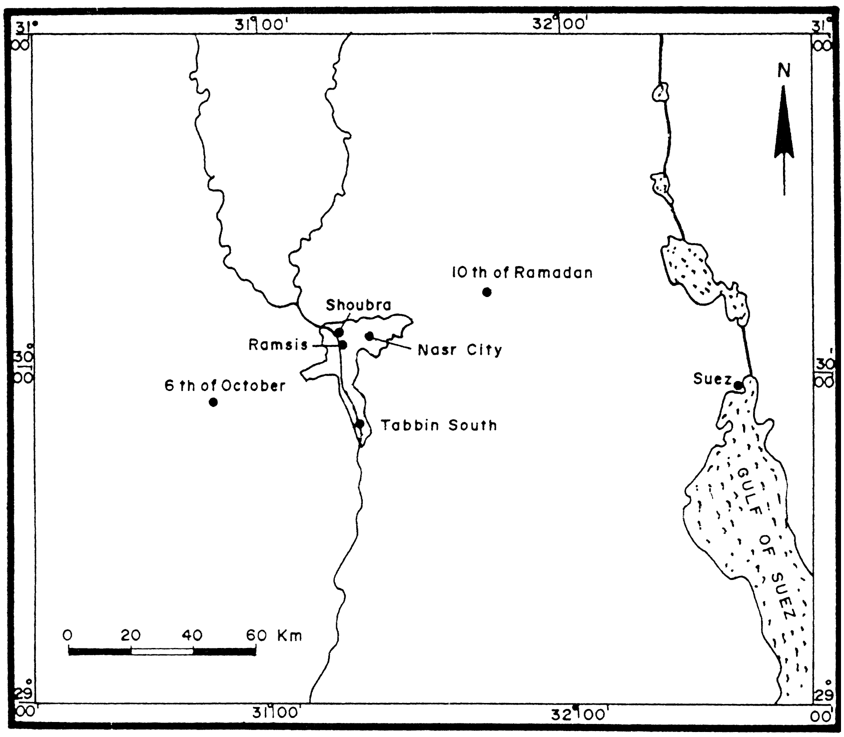

Fig. 1. Selected sites in Great Cairo.

Table 1
Microwave digestion program for air particulate samples

Step	Time	Power
1	00.05.00	250
2	00.05.00	400
3	00.05.00	650
4	00.05.00	250

Vent time: 00.25.00. Total time: 00.29.00.

period from January to October 1999, with a high volume filter sampler (sierra Anderson type) which gives information on the gross loading of the ambient air total suspended particulates (TSP) fraction with a size up to 25–50 μm. In addition to TSP, PM_{10} (particulate matter <10 μm) samples were collected with a size-selective high-volume sampler. Air particulate samples were collected for 24 h or Whatman EPA 2000 filter papers at a flow-rate of 1.0 m/min. The unexposed and exposed filters were preconditioned under controlled temperature and humidity for 24 h before weighing and analysis. PTFE and high-purity polyethylene containers were used for collecting, decomposing and storage of samples before analysis, to avoid the tendency of metals to be adsorbed on the walls of the containers.

An accurate mass of each collected filter was dissolved in the TFM container of a microwave oven with power regulator under temperature and pressure control [13]. The sample was heated in the presence of nitric and hydrofluoric acid according to the program shown in Table 1. After complete digestion, the samples were transferred into volumetric flasks and diluted to known volume with 5% of nitric acid. To test the validity and reliability of the whole investigations including digestion, analysis, quality controls are involved using initial calibration blank (ICB) and initial calibration verification (ICV) at the start and at the end of each run as well as continue calibration blank (CCB) and continuous calibration verification (CCV) were included.

3. Results and discussion

3.1. IC methodology

For an effective and efficient separation to occur, the relative migration rates of the solutes through the column bed of particles must be different (thermodynamic consideration), and their bandwidths must be sufficiently narrow to minimize overlap (kinetic consideration). Resolution (R_s) is a measure of the degree of the separation of two solute bands in terms of relative migration rates and bandwidth and is expressed mathematically as:

$$R_s = \frac{V_r(B) - V_r(A)}{[w(A) + w(B)]/2}$$

where $V_r(A)$ and $V_r(B)$ are the retention volumes for solutes A and B, respectively, and w(A) and w(B) are the peak widths in units of volumes measured at the base for solutes A and B, respectively, where

$$V_r = t_R F$$

t_R and F are the retention time and flow-rate, respectively. Inadequate separations occur when the calculated value for R_s is <0.8, whereas baseline separation is obtained when R_s is >1.25.

The method is first optimized using oxalic acid as eluent at 100 m*M*; pH 4.0 and a flow-rate of 0.85 ml/min. The run time for the four elements was 11.5 min. The resolution was 1.11, 3.24 and 1.75 for Pb^{2+}/Cd^{2+}, Cd^{2+}/Zn^{2+} and Zn^{2+}/Ni^{2+}, respectively. The method was further improved by addition of citric acid (100 m*M*, pH 4.0) to oxalic acid in a gradient elution to study the effect of gradient composition on both the resolution and overall run time (Fig. 2). The resolution data are listed in Table 2 and shown in Fig. 3. It was found that by increasing the citric acid composition, the resolution between Pb^{2+} and Cd^{2+} improved at the expense of decreasing the resolution between Cd^{2+} and Zn^{2+}. On the other hand, the decrease in resolution between Zn^{2+} and Ni^{2+} took place at a slower rate with the net result of decreasing the overall run. The increase of citric acid to 70% led to an overlap between the Cd^{2+} and Zn^{2+} peaks. On using citric acid alone, the run time was found to be very short but at the expense of resolution, which was very poor.

3.2. IC measurement

The detection was carried out using the oxalic–citric (60:40) (48 m*M* oxalic: 40 m*M* citric) gradient

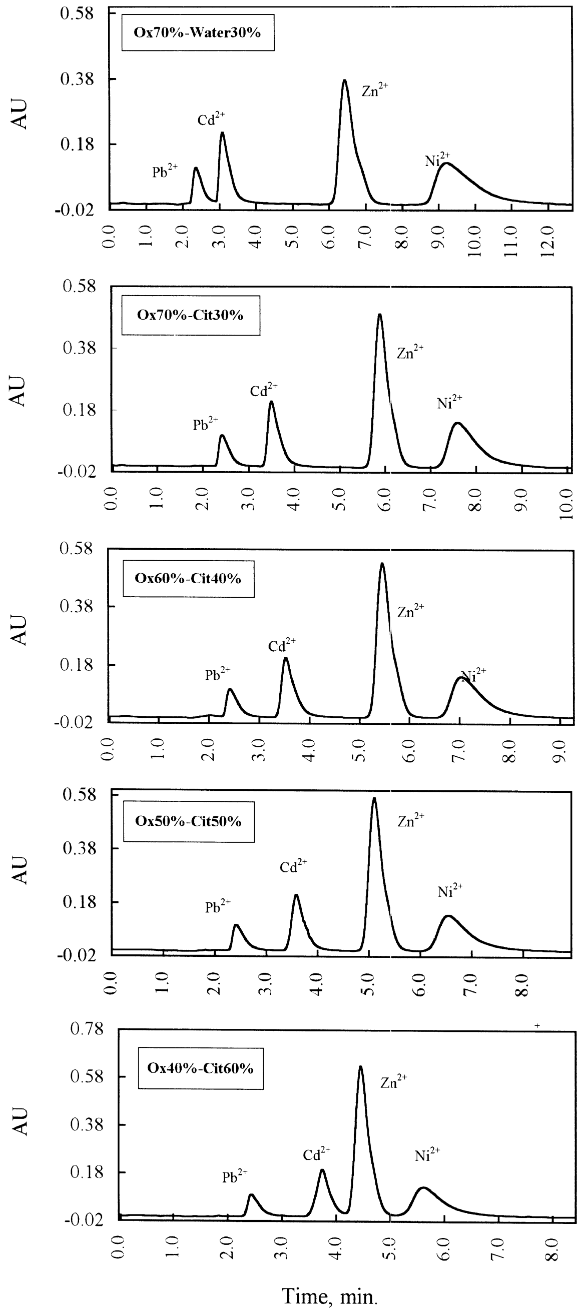

Fig. 2. Ion chromatograms showing the effect of citric acid composition on peak resolution.

composition at pH 4.0 and a flow-rate of 0.85 ml/min. The detection involved derivatization of the column effluent with 4-(2-pyridylazo)resorcinol (PAR) and subsequent photometric determination of the chelate complexes formed at a wavelength of 520 nm. The reagent delivery was achieved using a membrane reactor containing a semipermeable membrane. The measurements of the lead, cadmium, zinc and nickel ions were carried out based on a linear calibration curve constructed by a series of standard solutions ($r=0.9995$). Spiked samples were also analyzed for each metal.

3.3. Particulate matter

Concentrations of particulate matter (PM) across Cairo are extremely high, higher than in any other of the world's largest cities [14]. However, historical trends in Cairo's PM levels are difficult to determine, partly because regular monitoring at fixed sites was not initiated by the Ministry of Health until 1985. A wide monitoring network was initiated by the Ministry of Environment in 1998 (Environmental Information Monitoring Program; EIMP). Table 3 shows the PM distribution across Cairo along with the available average of the previous years. The high RSD values of the PM values may be attributed to the changes in wind direction between months, which is a general trend in Egypt.

Comparing the average PM values obtained with the available PM values from the previous years, it can be concluded that, in general, PM levels have increased consistently with the growth of the polluting activities in Cairo from 1991 to 1999 [15]. This reaches a maximum at Ramsis, which has a PM_{10} average that not only exceeds the reported PM_{10} values but also exceeds the TSP average reported from 1978–1991 which is considered an alarming indicator for adverse pollution of the Ramsis area. The lower value of PM at Nasr City (132.8 $\mu g/m^3$) compared with the average value from previous years (396 $\mu g/m^3$) is attributed to the fact that the average previous-year's value was collected as TSP while that of Nasr City was collected as PM_{10}.

3.4. Concentration of heavy metals and particle size considerations

3.4.1. Heavy metals concentration

Based on the obtained data of the mean concentration at each location, as shown in Tables 4 and

Table 2
Peak resolution as a function of gradient composition

Gradient composition	Run time (min)	Resolution (R_s)		
		Pb^{2+}/ Cd^{2+}	Cd^{2+}/ Zn^{2+}	Zn^{2+}/ Ni^{2+}
Oxalic acid–water (70:30)	11.5	1.11	3.24	1.75
Oxalic acid–citric acid (70:30)	9.0	1.43	3.10	1.60
Oxalic acid–citric acid (60:40)	8.4	1.81	2.78	1.43
Oxalic acid–citric acid (50:50)	7.6	1.91	1.96	1.34
Oxalic acid–citric acid (40:60)	6.5	2.13	1.00	1.22

Oxalic acid (0.08 *M*); citric acid (0.1 *M*); pH (4.0); flow-rate (0.85 ml/min).

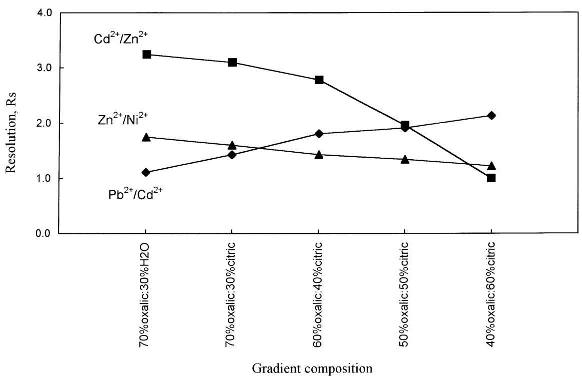

Fig. 3. Effect of gradient composition on peak resolution.

5, it was observed that there is a direct relationship between the trace metal composition of air particulate and the anthropogenic activities. Shoubra, Tabbin South and Ramsis have the highest heavy metal concentrations. Shoubra has over 450 industrial units of various sizes and employs 87 000 people, most of whom are also resident in the area. Industries include ferrous metallurgical work, foundries, lead smelters, ceramics, glass, bricks, textiles and plastics. In Tabbin South, there is a forest of brick factories that holds some lead smelters (which uses old batteries as a source of lead) and some other industrial activities. On the other hand Ramsis is not industrial area but a dense traffic area. The high

Table 3
Particulate matter (μg/m^3) distribution at the selected sites in Cairo

Time	Shoubra (TSP)	Ramsis (PM_{10})	Tabbin South (TSP)	Nasr City (PM_{10})	10th of Ramadan (PM_{10})	6th of October (PM_{10})
Jan.	870	675	531	251	119	121
Feb.	1175	720	728	194	96	108
March	877	712	814	136	78	101
April	651	809	690	96	55	80
May	820	360	843	125	146	101
June	790	801	326	79	61	81
July	596	816	706	89	52	67
Aug.	465	941	1398	96	74	101
Sept.	781	745	813	111	64	94
Oct.	735	733	773	151	87	121
Average	776	731.2	762.2	132.8	83.2	97.5
SD	190.56	150.47	272.73	53.87	30.08	17.55
RSD (%)	24.56	20.58	35.78	40.56	36.16	18.00
Literature average[a]	570	652		396		

[a] Average of annual averages of the period from 1978 to 1991 taken as TSP [14].

Table 4
Concentrations of the heavy metals (μg/m^3) in the selected sites

Month	Sample identity	PM	Pb	Cd	Zn	Ni
Shoubra (TSP)						
Jan.	Sh1	870	0.24	0.71	1.61	3.51
Feb.	Sh2	1175	0.24	1.14	1.75	11.01
March	Sh3	77	ND	0.40	0.24	3.35
April	Sh4	651	0.30	0.84	1.64	1.80
May	Sh5	820	0.26	0.36	2.54	2.58
June	Sh6	790	0.37	0.93	3.16	3.68
July	Sh7	596	2.26	2.38	5.80	2.83
Aug.	Sh8	465	0.70	0.28	0.74	5.99
Sept.	Sh9	781	ND	0.92	2.21	0.01
Oct.	Sh10	735	ND	2.13	0.55	ND
Ramsis (PM$_{10}$)						
Jan.	RS1	675	0.31	0.41	0.77	1.55
Feb.	RS2	720	0.33	0.33	0.83	3.35
March	RS3	712	0.14	0.41	0.61	2.11
April	RS4	809	0.71	0.42	0.65	1.67
May	RS5	360	0.71	0.30	0.51	0.06
June	RS6	801	0.66	0.11	0.83	1.31
July	RS7	816	0.53	0.36	0.84	0.01
Aug.	RS8	941	0.32	0.43	0.98	1.82
Sept.	RS9	745	0.71	0.47	0.88	4.20
Oct.	RS10	733	0.77	0.37	0.83	1.90
Tabbin South (TSP)						
Jan.	TSI	531	044	210	0.55	0.01
Feb.	TS2	728	0A4	3.55	1.01	0.01
March	TS3	814	0.01	1.67	0A6	ND
April	TS4	690	0.41	1.90	0.48	ND
May	TS5	843	0.01	2.08	0.52	ND
June	TS6	326	0.05	0.10	0.15	0.01
July	TS7	706	0.33	4.44	0.65	ND
Aug.	TS8	1398	0.01	6.83	1.87	0.01
Sept.	TS9	813	0.02	2.38	0.61	0.01
Oct.	TS10	773	0.20	1.35	0.71	0.01
Nasr City (PM$_{10}$)						
Jan.	NC1	251	0.88	0.24	0.69	0.04
Feb.	NC2	194	0.15	0.04	0.26	0.01
March	NC3	136	0.24	0.03	0.14	0.15
April	NC4	96	0.03	0.01	0.03	0.01
May	NC5	125	0.10	0.02	0.04	0.03
June	NC6	79	0.08	0.03	0.03	0.02
July	NC7	89	0.05	0.02	0.04	0.02
Aug.	NC8	96	0.24	0.01	0.05	0.04
Sept.	NC9	111	ND	0.03	0.07	0.01
Oct.	NCIO	151	ND	0.02	0.05	1.43

Table 4. Continued

Month	Sample identity	PM	Pb	Cd	Zn	Ni
10th of Ramadan (PM$_{10}$)						
Jan.	RA1	119	ND	ND	0.25	ND
Feb.	RA2	96	0.18	0.05	0.28	ND
March	RA3	78	0.01	0.05	0.27	ND
April	RA4	55	ND	0.01	0.04	ND
May	RA5	146	ND	0.03	0.06	0.02
June	RA6	61	ND	0.01	0.02	ND
July	RA7	52	0.26	0.01	0.03	ND
Aug.	RA8	74	0.01	0.01	0.02	0.01
Sept.	RA9	64	0.02	0.01	0.02	0.01
Oct.	RA10	87	0.03	0.03	0.03	0.01
6th of October (PM$_{10}$)						
Jan.	OC 1	121	0.05	0.15	0.21	0.02
Feb.	OC 2	108	ND	0.05	0.11	0.02
March	OC 3	101	0.05	0.02	0.03	0.01
April	OC 4	80	0.14	0.01	0.02	0.01
May	OC 5	101	0.09	0.01	0.03	0.01
June	OC 6	81	0.03	0.01	0.02	0.01
July	OC 7	67	0.02	0.02	0.02	ND
Aug.	OC 8	101	0.10	0.02	0.07	0.01
Sept.	OC 9	94	0.07	0.03	0.04	0.01
Oct.	OC 10	121	0.05	0.03	0.04	0.01

values of Pb, Cd, Zn, and Ni can be attributed to the fact that Ramsis is located downwind of other industrial areas such as Ghamrah and El Ameriah and also Shoubra. Although Nasr City, 6th of October and 10th of Ramadan contain industrial areas, the filters were collected from the residential areas which are located upwind of the industrial areas. This explain the relatively low values of heavy metals compared with Shoubra, Tabbin south and Ramsis.

3.4.2. Particle size considerations

Major efforts in this area have been initiated by the different groups, such as the US Environmental Protection Agency (EPA) [15], National Institute for Occupational Safety and Health NIOSH [8–10] and

Table 5
Mean concentration (μg/m^3) of different investigated elements at different sites

Location	Pb	Cd	Zn	Ni
Ramsis	0.5208	0.3625	0.7758	1.8014
Shoubra	0.6234	1.0094	2.0345	3.9316
10th of Ramadan	0.0849	0.0234	0.1032	0.0008
Nasr City	0.2239	0.0468	0.1394	0.1647
6th of October	0.0675	0.0337	0.0589	0.0042
Tabbin South	0.1939	2.6482	0.7065	0.0034

California Tri-City project [16]. Such studies provide useful information on the gross size of airborne particulate to differentiate anthropogenic and natural origins. In Cairo, there is a scarcity of data on toxic metals exposure assessment; further studies to evaluate the concentration and composition as a function of particle size are required. Although the majority of the mass of airborne particulate matter is non-metallic [17], this study is concerned with the measurement of metals in particulate matter. In this respect two different size fractionated filters were used namely TSP and PM_{10}.

Comparing the trace metals mean concentration for both types of filters (TSP and PM10); it was found that the larger particles collected on the TSP filter have a higher mean concentration of the different investigated elements Pb, Cd, Co, Zn and Ni than the fine particles collected on the PM_{10}. This is because the larger particles result from mechanical, different community and natural processes while the smaller particles are formed mainly by condensation of gases and vapors. Accordingly, the trace metal composition of air particles close to human activity differs considerably.

Although TSP has higher mean concentration of heavy metals, PM_{10} is expected to have more adverse human effects. Large particles (TSP) which deposit in nasopharyngal system can be removed by mucus and enter the gastrointestinal tract within a short period (1 day). Once in the gastrointestinal tract, particles can have a primary toxic effect. On the other hand, small particles (PM_{10}), that enter the lower respiratory system, not only affect the process of gas exchanges but also cause damage to the lung tissues. The effect becomes adverse when these heavy metals reach the blood; this is due to their ability to replace iron in the hemoglobin and also their catalytic nature which may enhance many biochemical processes.

3.4.3. Testing the significance of differences between several mean concentrations of different locations

Statistical evaluation using one-way analysis of variance (ANOVA) [18] was carried out to correlate the significant differences for the investigated trace metals between the different chosen locations during the period January to October 1999. Source of variance between different locations for each of the investigated elements was calculated based on the sum of squares between different groups (Bet ss), sum of squares within the different groups (with ss) and the total sum of squares (total ss). Different F values for each of the investigated metal are given in Table 6. The highest F value was obtained for cadmium while the lowest F value was obtained for cobalt.

The data also show that very highly significant differences ($P<0.001$) were obtained for Cd, Zn, and Ni while a highly significant difference was obtained for Pb ($P<0.01$).

3.5. Concentration and time dependence

Another area that also warrants interest is the measurement of the time dependence of particulate emissions to identify which segment of the population is exposed during peak emissions. Exposure is a function of dosage and time, and it is possible that exposure to high concentrations for even short periods can present a serious health hazard.

Based on the obtained mean concentrations reported in Table 7, it was found that the highest peak emission for the investigated elements were in July for Pb and Zn, July and August for Cd and February

Table 6
One-way analysis of variance (ANOVA) for the different heavy metals at different locations

Source of variation	Df	SS	MS	F	P
Lead					
Bet ss	5	2.1357	0.4271	4.0103	<0.01
With ss	44	4.6874	0.1065		
Total ss	49	6.8231			
Cadmium					
Bet ss	5	52.2567	10.4510	14.9663	<0.001
With ss	53	37.0100	0.6983		
Total ss	58	89.2610			
Zinc					
Bet ss	5	28.4371	5.6870	11.8750	<0.001
With ss	54	25.8659	0.4789		
Total ss	59	54.3030			
Nickel					
Bet ss	5	108.4550	21.6900	9.6280	<0.001
With ss	42	94.619	2.2528		
Total ss	47	203.074			

Table 7
Mean concentrations (μg/m^3) of the different investigated elements in different months

Month	Pb	Gd	Zn	Ni
Jan.	0.3851	0.7260	0.6988	1.1469
Feb.	0.2706	0.8625	0.7058	2.8808
March	0.0898	0.4329	0.2956	1.4040
April	0.3190	0.5350	0.4785	0.8696
May	0.2344	0.4670	0.6179	0.5345
June	0.2409	0.2018	0.7041	0.9991
July	0.5775	1.2084	1.2299	0.9442
Aug.	0.2291	1.2634	0.6224	1.3055
Sept.	0.2046	0.6408	0.6423	0.7030
Oct.	0.2655	0.6544	0.3682	0.6679

for Ni. The lowest peak emissions for the investigated elements were in March for Pb, June for Cd and June for Zn and Ni. Most elements reached their maximum concentration in July and August. This is consistent with the increased activities leading to particulate matter emission during the summer period.

To correlate the significant differences for the investigated trace metals during the period starting from January to October 1999, ANOVA was used and the results are given in Table 8. Despite the fact that the data demonstrate high mean values in July and August, the statistical analyses of the data revealed non-significant difference during the investigated period. This can be due to high individual variation between months.

Table 8
One-way analysis of variance (ANOVA) of the different heavy metals at different period of times

Source of variation	Df	SS	MS	*F*	*P*
Lead					
Bet ss	9	0.8287	0.0920	0.6141	N.S.
With ss	40	5.9944	0.1499		
Total ss	49	6.8231			
Cadmium					
Bet ss	9	5.4476	0.6052	0.3538	N.S.
With ss	49	83.8124	1.7104		
Total ss	58	89.2600			
Zinc					
Bet ss	9	3.4743	0.3860	0.3797	N.S.
With ss	50	50.8287	1.0165		
Total ss	59	54.3030			
Nickel					
Bet ss	9	20.1873	2.2430	0.4660	N.S.
With ss	38	182.8867	4.8128		
Total ss	47	203.0740			

N.S., not significant.

3.6. Safety levels

The WHO air quality guidelines [19] state that different effects on mortality, respiratory and cardiovascular hospital admissions have been observed at daily average PM_{10} levels well below 100 μg/m^3. In Western Europe, North America and the Western Pacific, except China, annual mean TSP concentrations range between 20 and 80 μg/m^3; PM_{10} levels are between 10 and 55 μg/m^3. High TSP and PM_{10} annual mean concentrations are found in South East Asia ranging between 100 and 400 μg/m^3 for TSP and 100 and 400 μg/m^3 for PM_{10}. However, the highest recorded annual TSP concentrations, 300–500 μg/m^3, are observed in the larger cities of China.

In Cairo it was found that the obtained annual mean concentration for PM_{10} ranges were 55–743 μg/m^3 in 10th of Ramadan, 733–941 μg/m^3 for Ramsis, while the annual mean concentration for TSP ranged between 465 and 1175 μg/m^3 in Shoubra, and between 326 and 1398 μg/m^3 in Tebbin South. These figures demonstrate that both PM_{10} and TSP in certain sites such as Ramsis, Shoubra, 10th of Ramadan and Tebbin South are over the highest recorded values in large cities of China. The Egyptian guideline for TSP is 90 μg/m^3 (annual average) and for PM_{10} is 70 μg/m^3 (24 h average).

According to the WHO the annual mean lead concentration in ambient air is below 0.1 μg/m^3 in most cities of Western Europe and between 0.2 and 0.6 μg/m^3 in Eastern European cities. In North American cities, annual mean lead concentration is below 0.05 μg/m^3. In the studied sites of Cairo, high lead mean concentration was observed in some sites, where it was found to be 0.06–0.62 μg/m^3. These values seem to be lower than the Egyptian annual limit (1.0 μg/m^3) but the observed values represent only the particulate lead and not the total lead emissions which includes other forms such as lead fumes.

However, there are no annual limits for Cd, Zn and Ni in ambient air either in the WHO or the Egyptian guidelines (non-criteria parameters). The values obtained are considered to be high considering the catalytic nature and toxicity of these metals which upon inhalation may lead to dangerous effect on human health. This may be considered a motivation to the international community to define annual limits for these heavy metals in ambient air.

4. Conclusion

This study has confirmed that air burden shows a definite particle size dependence related to chemical composition of the particulate matter of air which reflects human activity.

Focusing and environmental consciousness must be taken into consideration in certain heavy metal polluted sites such as Shoubra, Ramsis and Tebbin South that suffer from different sources of heavy metals pollution.

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