

Indoor and Outdoor Pollution with Heavy Metals in Al-Karak City, Jordan

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Abstract: The concentration of eight potentially toxic heavy metals (Cr, Cu, Ni, V, Ti, Mn, Pb and As) were investigated in indoor and surrounding outdoor dust samples collected from residential and commercial sites in Al-Karak city, Jordan. It was found that at residential areas, the mean concentration of Cr, Cu, Ni, V, Ti, Mn, Pb and As were 72.5, 90.5, 20.8, 37.9, 291.3, 238.8, 51.9 and 2.06 mg/kg, respectively. While, their mean concentrations for outdoor dust were at the 51.7, 57.4, 10.0, 41.0, 351.2, 227.0, 52.7 and 2.7 mg/kg for the same elements, respectively. For the commercial areas, their mean concentrations were 58.5, 122.3, 15.0, 28.6, 195.7, 243.2, 110.4 and 2.4 mg/kg, respectively. Whereas, their mean concentrations for the outdoor dust samples were 57.3, 93.4, 29.3, 27.3, 86.0, 259.4, 75.8 and 2.0 mg/kg, respectively. Furthermore, the relationship between outdoor and indoor dusts was considered and it was clear that the main source of V, Ti, Mn, Pb and As are vehicular emission with the exception of Ni, Cr and Cu which was found at a higher concentration in the indoor dust samples than the outdoor dust samples. The results of this study reveal that heavy metals distribution is different in two investigated areas of the city as it was higher in the commercial area compared to residential area due to higher traffic density. The result indicated that soil or street dust play an important role in the concentration build up in indoor and outdoor dust which originated most probably from automobile emissions.

Keywords: *Indoor dust, outdoor dust, Heavy metals, Al-Karak, ICP-MS.*

Introduction

With the evolution of modern industry, the atmosphere is loaded with various toxic materials such as heavy metals and others. Heavy metals are natural constituents in nature usually occurring in low concentration under normal conditions. Heavy metals are emitted into the atmosphere either from natural origin such as bedrocks erosion or from many different sources in urbanized areas, including vehicle emissions, industrial discharges and other activities (Kurt-Karakus, 2012). Therefore the concentration of heavy metals varies from site to site depending on the anthropogenic activities of the sites.

Air quality was monitored used different methods such as street dust (Al-Momani 2007), tree bark (Ziadat *et al.*, 2015), settled dust (Al Bakaina *et al.*, 2012), street dust (El-Hasan *et al.*, 2006; jiries, 2003) and others. Top soil and roadside dusts in urban areas are indicators of air pollution with the presence of heavy metals from atmospheric deposition (Hadadin *et al.*, 2007; Abed *et al.*, 2009; Howari & Ghrefat, 2011). Indoor air quality is an important part of the environment as people spend more than 88% of their time indoor (Mercier *et al.* 2011). Heavy metal pollutant in urban street dust has become a growing concern in recent years (Shinggu *et al.*, 2007). Recently indoor air quality is gaining greater attention due to its relationship to health as it is a reservoir for chronic exposure to indoor pollutants (Roberts & Ott 2007; Hochstetler *et al.* 2011; Aucott & Caldarelli, 2012) and it is hazardous to animals and vegetation (Tong & Lam, 2000).

The sources of heavy metals in house dust are categorized into two types indoor and outdoor. Indoor sources for house dust are through items used or activities carried out within the houses by occupant's activities which are generally cookers, heaters, consumer products, building and furnishing material, smoking and incense burning in the houses as well as infiltration of outdoor pollutants (Kabata-Pendias, 2011; Jaradat *et al.*, 2004). An outdoor source includes automobile emission, street dust; elements Zn, Cu and Cd are associated with tire wear, corrosion of metallic parts of automobiles (Cui *et al.*, 2004; Lavado *et al.*, 2007; Gharaibeh *et al.*, 2010).

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House dust can be a useful indicator of the amount of metals in indoor and outdoor environments, many of heavy metals such as Pb, Cd, Hg and Sb found in high level in house dust compared to samples collected from garden soil and street dust, (Rasmussen et al. 2001 and Al-Momani, 2007). Lambert and Lane (2004) estimated that there was a direct correlation between the sources and levels of heavy metals in indoor environments with the outdoor. Particulate matter in the indoor environment adsorbs many of the contaminants which are accumulated into the settled dust (Butte *et al.*, 2004). The composition of indoor dust is complex and heterogeneous matrix house dust can be significantly different from house to houserooms, and among houses (Lioy *et al.*, 2002).

The aim of this work is to evaluate indoor air quality in three areas with different anthropogenic activities in Al-Karak city-Jordan in terms of heavy metals by measuring the concentration of heavy metals in dust collected from different anthropogenic activities.

Material and Methods

Study area

Al-Karak city is located in the central part of Jordan with around 300,000 inhabitants. In the study area, there are few small industrial areas and it is expected that their contribution to air pollution is very low; therefore the main source of heavy metals is mainly from traffic sources. Jordan has a Mediterranean climate. Rainfall occurs only during winter seasons extending from November to April. During summer seasons, the northwesterly breeze has a very slight effect on heavy metals redistribution on the investigated area. Sampling was done at the summer season from July to October 2013 to avoid washing away of heavy metals by street runoff therefore the rain do not any effect on the concentrations of heavy metals on dust samples.

Sampling

The sampling area at Al-Karak (Figure 1) city was divided into two areas with different air pollution potentials. **Area A**, characterized with high traffic density and **Area B** was a residential area characterized with medium to low traffic density. Additionally, a remote site with similar geology was selected as a background for monitoring outdoor air pollutants representing locations with very low traffic density.

For both areas A and B, indoor and outdoor dust samples were collected simultaneously using polyethylene brush, tray and containers. At each area the following samples were collected, twelve samples from the city center located directly to the major road, twelve sites from the residential area with a medium to far distance from the major road and three samples from a remote site. The samples were sieved using plastic sieve to remove debris associated with sample collection. The sieved samples were labeled and stored in a PVC vials (15mL) in the refrigerator at 4 °C until the analysis time.

Indoor settled house dust samples were collected from different part of the houses at elevation at least two meters above the ground to avoid soiling; therefore, the indoor dust samples do not consider the different activities inside the houses. The outdoor samples were collected from area surrounded the house.

Heavy Metals Analysis and Instrumentations

Microwave-assisted acid digestion method was used to extract the heavy metals from the dust samples. The digestion of dust samples was carried out according to the EPA method 3051A with slight modification (EPA, 2007). Anton Parr microwave digestion system model Multiwave 3000 (Anton Parr®, Österreich), equipped with 100-mL PFA Teflon vessels (MF 100) and 16-position Rotor was used for sample digestion. For each sample, (0.3 – 0.5) g was accurately weighted (± 0.0001) and transferred to the pre-cleaned Teflon-lined digestion vessels. Then, 12 mL of high purity HNO₃ (70%) and HCl (37-38 %) were added at a ratio of 3:1 v/v. The samples were then digested using the program summarized in Table 1. The sample extract was then quantitatively filtered using glass wool to standard polyethylene volumetric flask and diluted to a final volume of 50 mL using deionized water. The heavy metals analysis were carried-out using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Model: Elan DRC II) (PerkinElmer®, USA), Table 2 shows the Instrumental operation parameter.

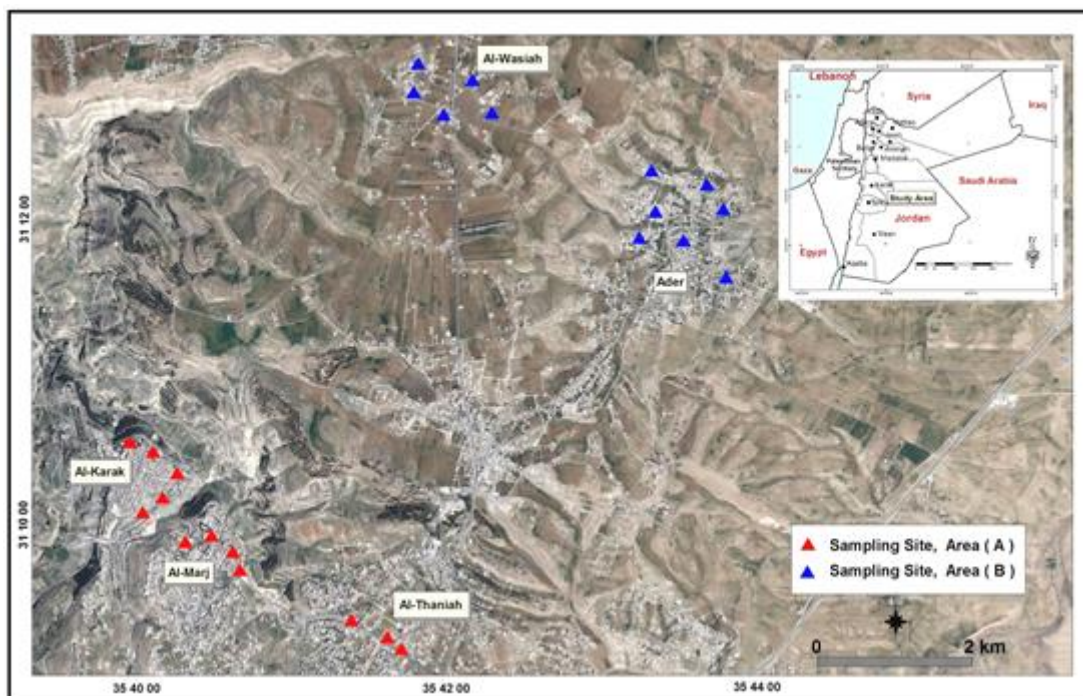


Figure 1: Study Area

Table 1: Operation parameters used for the microwave digestion

Stage	Ramping		Hold (Min)	Temp °C	Pressure (Bar)	Fan Speed (rpm)
	Time (min)	To Power (Watt)				
1	5	800	20	Up to 210	Up to 40	100
2	0	0	45	-	-	200

Table 2: Instrumental operation parameter for ICP-MS (Model: Elan DRC II) (PerkinElmer®, USA)

Operation Parameter	Setting
RF power	1100 W
RF generator Frequency	40 MHz
Analog stage voltage	-1735 V
Pulse stage voltage	840 V
Main water Temperature	19 °C
Interface water Temperature	30 °C
Torch box temperature	44 °C
Lens voltage	6.75 V
Argon flow rate:	-Plasma 15 ml/min
	-Auxiliary 1.2 ml/min
	-Nebulizer 0.84 ml/min
Nebulizer type	Cross-flow
Interface	Pt skimmer cones.
Dwell time	50 ms per amu
Torch	Standard quartz torch

Analytical Method Validation

Eight metals (Zn, Ni, Pb, Cu, Ti, Cr, As and Mn) were analyzed quantitatively based on external calibration curves for series of standard solutions using Inductively coupled plasma Mass spectrometry (ICP-MS) Elan DRC-e (PerkinElmer, Canada-USA). The calibration curves were buildup for each element by the instrument with using the best regression lines with correlation coefficients ($r^2 > 0.999$). The blank solutions showed no appreciable values of heavy metals and their values were ignored. For the degree of accuracy for total heavy metal digestion, 7 samples of laboratory certificated reference material “Calcareous Loam Soil” CRM 141 R (European Commission, 1995), was digested using the same analytical procedure and analyzed subsequently

using ICP-MS. The results were compared with certified CRM values, the percent recovery was computed, and it was found ranging from 82.55 % to 115.20 % with relative standard deviation (RSD %) of less than 10 %, Figure 2

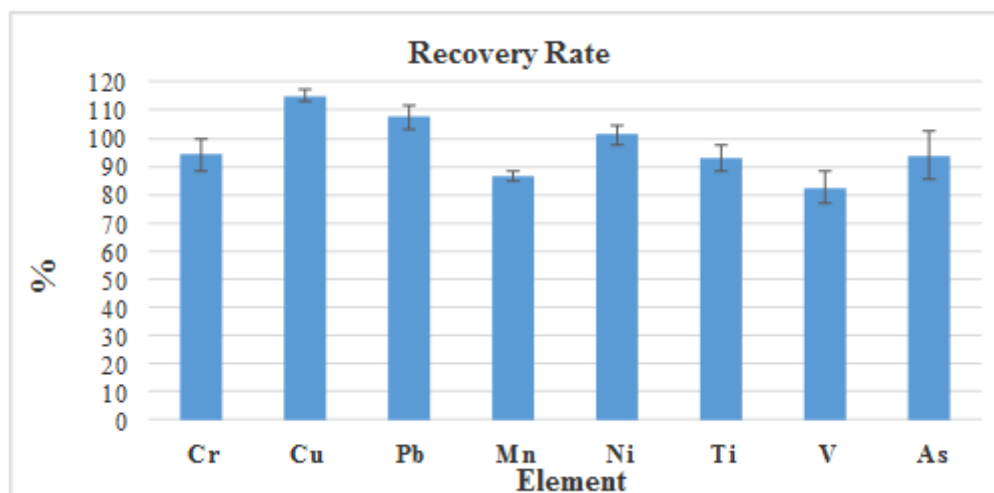


Figure 2. Recovery Rate of the elements using CRM (141 R) by ICP-MS

Quality Control and Method validation

All glassware used for extraction and cleanup method, polyethylene bottle, volumetric flasks and Teflon digestion vessels were washed sequentially twice with tap water, soap, distilled water and soaked in 10% (v/v) nitric acid overnight and then rinsed in deionized water prior to digestion procedures. All dilutions were done using deionized water and A-grade volumetric flasks; all solvents were of Analytical Reagent (AR) grade. The glass wool used for filtration was soaked in 10% (v/v) nitric acid overnight then washed with deionized water before the use.

Results and Discussion

Concentrations of heavy metals

The concentrations of the heavy metals (Cr, Cu, Pb, Mn, Ni, Ti, V and AS) in dust collected samples from eighteen selected sites of Al-Karak city representing residential areas and commercial areas are summarized in Table 3.

Heavy metal content in house dust is influenced by various factors concerning the indoor as well as outdoor activities. The outdoor dust may enter the houses through the windows as they are kept open during the summer period, therefore it is expected to have significant correlation in heavy metals content between indoor and outdoor dust.

Pb is a common metal found in house hold such as leaded paint, pottery glazes, batteries and others. Therefore, most lead found in household dust and road side is the legacy of past production of lead and application of lead in fuel which was banned in Jordan since 2008 in addition to minor source from indoor activities. Similar results obtained in another study of heavy metals contamination in Hong Kong (Li et al. 2004) which attributes high concentrations of Pb due to historical use of Pb.

The lead concentration in the commercial area was higher than residential area. Its concentration in commercial area ranged from 23.7 to 90.5 mg/kg in indoor dust and from 23.9 to 88.6 mg/kg in outdoor dust at the residential area. While, it was varied from 10.7 to 147.9 mg/kg in indoor dust and 30.3-139.5 mg/kg in outdoor dust of the commercial area. The most probable source of Pb can be from tire and brake abrasions contribute significant amounts of Pb into the environment (Thorpe and Harrison 2008).

The concentration of Cr showed different trend, as its concentration was higher in indoor residential area than outdoor dust. Its concentration ranged from 48.9 to 184.5 mg/kg and from 39.0 to 65.3 mg/kg in the indoor and the outdoor dust, respectively. For commercial areas, the concentration of Cr was lower than residential areas as its concentrations in indoor dust ranged from 13.9 to 83.0 mg/kg and from 28.6 to 78.8 mg/kg in outdoor dust. This can be attributed to the use of Cr in many household products such as plating of some home furniture and appliances of the residential area

(Madany et al., 1994; Rashed, 2008)) and to less extent in commercial areas that may be derived from mechanical abrasion of vehicles, as they are used in many parts of the vehicles.

Table 3: Statistical summary of heavy metals concentration in ppb ($\mu\text{g g}^{-1}$) for dust samples collected from residential and commercial area at Al-Karak city

	Heavy Metal	Indoor				Outdoor			
		Min	Max	Mean	\pm SD	Min	Max	Mean	\pm SD
Residential	Cr	48.9	184.5	72.5	42.9	39.0	65.3	51.7	9.3
	Cu	33.2	161.8	90.4	44.5	29.8	102.8	57.4	30.0
	Ni	6.5	463.7	70.0	149.7	0.1	31.7	10.0	10.4
	V	24.9	44.1	37.9	6.3	25.5	51.3	41.0	8.1
	Ti	193.8	407.7	291.3	72.7	117.4	483.3	351.2	131.5
	Mn	156.8	340.0	243.2	52.5	161.0	390.3	259.4	65.7
	Pb	23.7	90.5	51.9	21.3	23.9	88.6	52.7	25.3
	As	1.7	3.1	2.6	0.5	1.5	3.8	2.7	0.8
Commercial	Cr	13.9	83.0	49.5	19.9	28.6	78.8	50.7	15.8
	Cu	14.7	215.1	101.0	59.3	31.8	138.0	73.2	37.1
	Ni	2.0	38.3	10.8	11.9	5.5	105.0	29.2	31.7
	V	4.8	38.9	28.6	10.6	21.3	34.1	27.7	5.1
	Ti	6.3	329.3	160.8	115.5	4.4	125.8	60.9	47.4
	Mn	68.4	311.7	223.2	77.4	119.8	267.2	219.5	48.9
	Pb	10.7	147.9	95.8	39.3	30.3	139.5	75.3	32.7
	As	0.4	2.9	2.3	0.8	1.2	2.9	1.9	0.5

The highest metal concentration at commercial area was Mn as it ranged from 156.8 to 339.9 mg/kg in the indoor dust and from 161.0 to 390.4 mg/kg in the outdoor dust, respectively. However, the concentration of Mn was higher than residential areas, which ranged from 68.4-311.7 mg/kg for indoor dust and from 119.8-267 mg/kg of outdoor dust. The most probable source of Mn in the study area originated from fossil fuel combustion as since 2008 all fuel used in Jordan is unleaded i.e. Mn is substituted Pb in the fuel.

The element Mn and Ti were obtained in significant concentrations which accounts to more than 50% of the analyzed metals at both indoor and outdoor dust samples for commercial area and more than 65% in residential area suggesting that sources other than natural are contributing to their levels.

Ti was found to be in the greatest concentration in residential indoor and outdoor dust samples and it was found at a higher concentration levels than commercial area. Ti concentration in residential area ranged from 193.8 to 407.7 mg/kg for indoor dust and from 117.4 to 483.2 mg/kg for outdoor dust. However, it was found at lower concentration level in commercial areas and ranged from 6.3 to 329.3 mg/kg for indoor dust and from 4.4 to 125.8 mg/kg for outdoor dust. The most probable source of Ti is the use as a white pigment in painting (Titanium White) as more white paint is used at residential areas rather than commercial areas.

The source of Cu, Cr and Ni in dust is supposed to be from car components, tire abrasion, lubricants corrosion of cars, engine wear, brushing, and brake dust respectively (Thorpe and Harrison 2008).

Arsenic (As) levels were low at both residential and commercial sites although it originates from household fuel burning as well as from vehicle exhaust. The concentration of As in residential area ranged from 1.7 to 3.1 mg/kg in indoor dust and from 1.45-3.8 mg/kg in outdoor dust. The concentration of As in commercial area showed lower concentration levels than residential area as it ranged from 0.4 to 2.9 mg/kg in indoor dust and from 1.2-2.9 mg/kg in outdoor dust.

Correlation coefficients

The correlations of heavy metals in indoor dusts are shown in **Table 4**. For residential area, all the metal pairs showed negative relations except for Ni-Cr, Ti-V, Mn-V, As-V, As-Ti and As-Mn pairs which were significant at 95% and/or higher confidence level. For commercial area all metal pairs showed insignificant correlation except Ti-Cr, Mn-Cr, Mn-V, Mn-Ti, Pb-Cu, As-Cr, As-V and As-Mn. Ti correlated strongly with V, which is a product of fuel combustion (Lu et al. 2010), showing

that they may be coming from a common source in addition to additional amount Ti which are produced of brake wear (Atiemo *et al.*, 2011).

Table 4: Correlation matrices between heavy metals concentration in the Indoor dust

		Cu	Ni	V	Ti	Mn	Pb	As
Commercial	Cr	0.24	0.29	0.43	0.83	0.76	0.23	0.73
	Cu		-0.04	0.45	0.26	0.39	0.99	0.56
	Ni			-0.60	0.19	-0.07	-0.04	-0.19
	V				0.49	0.78	0.47	0.86
	Ti					0.70	0.27	0.65
	Mn						0.41	0.91
	Pb							0.59
Residential	Cr	0.67	0.99	0.25	0.49	0.06	-0.06	0.36
	Cu		0.60	0.09	0.44	-0.18	0.67	0.12
	Ni			0.24	0.41	0.08	-0.14	0.35
	V				0.79	0.87	0.00	0.92
	Ti					0.49	0.22	0.87
	Mn						-0.29	0.73
	Pb							-0.07

Although Ni showed low concentrations in both investigated areas, and the correlation analysis showed that the Ni and Cr correlated strongly with each other in dust samples collected from residential area but poorly at other sites. Therefore, their origin cannot be attributed to corrosion of motor vehicle parts as it was found in other sites (Lu *et al.* 2009; Al-Shayeb & Seaward 2001). The source of Ni and Cr in indoor dust of the residential area is assumed to be from corrosion of appliance used in the houses rather than corrosion of motor vehicle parts.

Copper is present as one of the most abundant element in both indoor dust and outdoor dust as it present in many parts of indoor and outdoor equipments. The concentration of Cu in the residential area ranged from 33.2 to 161.8 mg/kg for indoor dust and from 29.8 to 102.8 mg/kg in outdoor dust. However, for commercial area it showed a wider range as it ranged from 14.7 to 292.2 mg/kg in indoor dust and from 31.8 to 255.1 mg/kg for outdoor dust. Therefore results of correlation analysis may indicate a common sources origin for both sites, such as automobile emission.

Indoor-Outdoor Metals Concentration Relationship

Although heavy metals in dust could be derived from indoor and outdoor sources, the result of this study indicated that for residential area Indoor-Outdoor Metals Concentration Relationship for V, Mn, Pb and As concentrations are similar which is related to exterior dust particulates mainly from vehicle emission, while for Cr, Cu and Ni the concentrations are higher in indoor dust samples compared with the outdoor dust samples indicating that they were generated interior activities (Figure 3). However for commercial areas the concentrations of Cu, Ti, As and Pb were higher in indoor samples than outdoor samples and Cr, V, Mn were very similar indicating external source while Ni showed higher concentrations in outdoor dust than indoor dust indicating internal source (Figure 4).

From the review of results and their comparison with the values given in the literature (Table 5), we can conclude that the concentrations of metals appear relatively low and lower than in many developed countries, especially referring to Cu, Pb, Ni, Cr and Mn concentration levels. The relatively low percentage of lead compared to some countries including Amman city, can be explained by the limited use of leaded gasoline as the primary source of this metal in the urban air particles. On the other hand, the mean concentrations of lead, Mn in indoor dust were higher than other developed countries such as Australia and Hong Kong which might be due to the use of only unleaded fuel in Jordan since 2008 and the other studies were done long time ago.

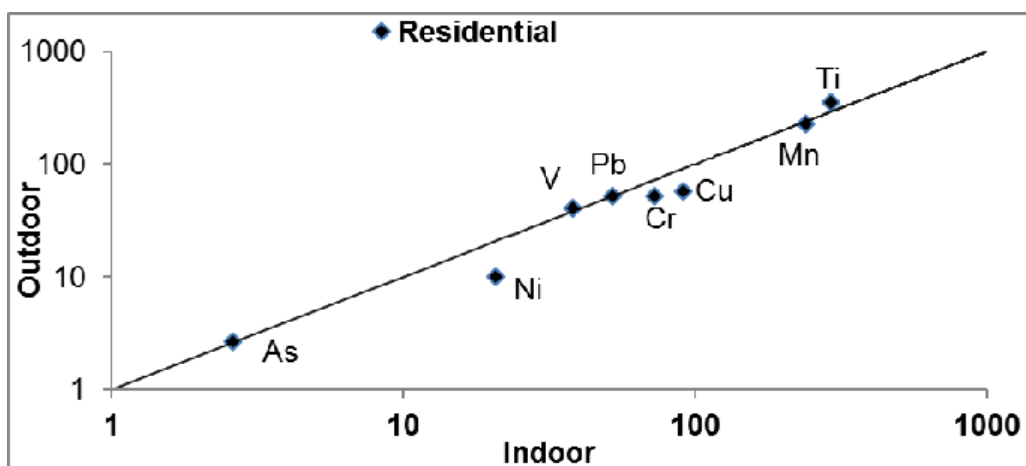


Figure 3: Residential Area Indoor-Outdoor Metals Concentration Relationship.

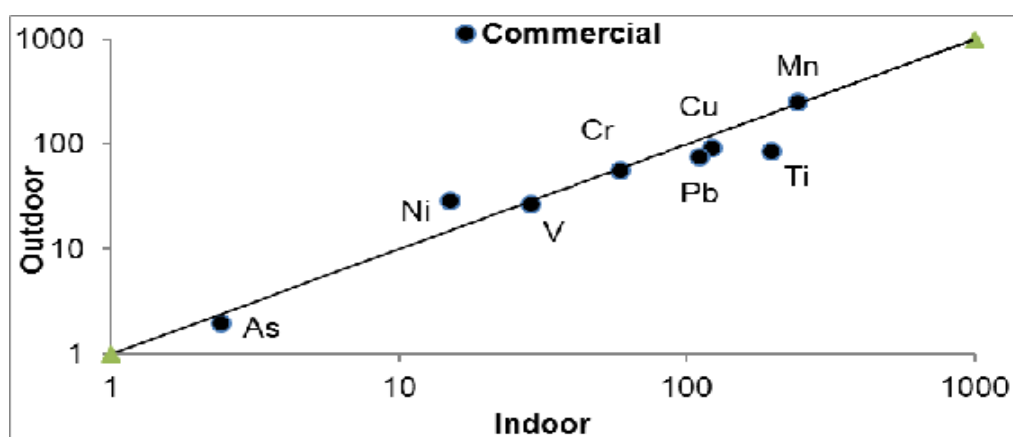


Figure 4: Commercial Area Indoor-Outdoor Metals Concentration Relationship.

Table 5. Global distribution of heavy metals in indoor dust in $\mu\text{g/g}$.

City	Cu	Pb	Ni	Cr	Mn	Ref.
Al-Karak-Jordan	73.9	52.3	40.0	62.1	251.3	This study
Amman-Jordan	160	206	47	77	304	Al-Momani, I 2007
Ottawa, Canada	157	222	52	69	267	Rasmussen et al., (2001)
Sydney, Australia	93	76	15	65	48	Chattopadhyay et al. (2003)
Warsaw, Poland	109	124	30	90	NA	Lisiewicz et al. (2000)
Kwun Tong, China	806	308	NA	NA	283	Tong and Lam, (2000)
Hong Kong	167	164	NA	NA	193	Tong and Lam (1998)
Selangor, Malaysia	NA	850	830	NA	NA	Latif et al., (2009)

NA: not available.

Conclusions

This study was carried out to evaluate the indoor and outdoor air quality in terms of heavy metals in Al-Karak province, Jordan. The high metal concentration of the sampling area indicated that the major source of heavy metals were from traffic emission in addition to minor quantities from anthropogenic sources. The results showed that the concentration of heavy metals Cu, Cr, Ni in residential areas and Ti, Cu and Pb in commercial areas of Al-Karak province were generally higher in indoor dust compared with outdoor dust. Meanwhile, there were no significant differences for other metals between indoor and outdoor dust. The results of heavy metal concentration in investigated areas were in order $\text{Mn} > \text{Ti} > \text{Cu} > \text{Pb} > \text{Cr} > \text{V} > \text{Ni} > \text{As}$ in commercial area and $\text{Ti} > \text{Mn} > \text{Cu} > \text{Cr} > \text{Pb} > \text{V} > \text{Ni} > \text{As}$ in residential area.

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