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Lead and Cadmium Contamination in Roadside Soils in Irbid City, Jordan: A Case Study

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This study evaluates the amount and distribution of Pb and Cd in roadside soils adjacent to two main roads in Irbid, Jordan, from October 2001 to July 2002. A total of 260 samples were collected from Irbid-Howara Street and Yarmouk University Street. Lead and Cd content were measured using Graphite Furnace Atomic Absorption Spectrometry. The environmental significance of this study is discussed in terms of the influence of traffic density on Pb and Cd concentrations in roadside soils, and enrichment factors were calculated to evaluate the degree of pollution. The accuracy of the results obtained has been examined and two standard reference materials, CRM 142 R (soil) and SRM

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2709 (soil), were analyzed to confirm the accuracy of the results. The precision of the measurements was assessed in terms of relative standard deviation (RSD) using five replicate analyses of samples collected from the roadside sites. RSD values for Pb and Cd were found to be less than 6%. The overall Pb concentration in roadside soil samples was 325 and 431 $\mu g\ g^{-1}$ for Pb and 1.142 and 1.135 $\mu g\ g^{-1}$ for Cd in Yarmouk University Street and Irbid-Houwa Street, respectively. Results indicated that levels of Pb and Cd were decreasing as the distance from the road increases. Enrichment factor results were 655 and 826 for Pb, and 252 and 237 for Cd in Yarmouk University Street and Irbid-Houwa Street, respectively.

Keywords Graphite furnace atomic absorption spectrometry, lead, cadmium, roadside soils, Jordan.

Introduction

Contamination of roadside soils with heavy metals arises from a number of sources, such as vehicles, road wear, slipperiness control, buildings (heating and corrosion), local building activities, and pollution from local industries. Trace metal concentrations, such as Cd, Cu, Zn, and particularly Pb, in surface soils have been the focus of investigations (Sutherland, 2000). The accumulation of these metals in topsoil is greatly influenced by traffic volume and motor vehicles, which introduce a number of toxic metals into the atmosphere (Wixon and Davies, 1994). Pb, in particular, is an environmental pollutant in soils because of the use of alkyl lead compounds as antiknock and freezing additives in fuel (Ward, 1990; Choudhary *et al*., 1998; Carlosena *et al.*, 1998). The main source of Cd in roadways is tire erosion (Ferguson and Kim, 1991). Pb is particularly toxic to the brain, kidneys, reproductive system, and cardiovascular system as a result of chronic and acute exposure and the repeated exposure of Pb buildup in the body (Ratccliffe, 1981). It was reported that children absorb Pb more readily in their gastrointestinal tracts than adults, and the developing nervous system is very susceptible to the deleterious effect of lead (WHO, 1995).

Cadmium has an extremely long biological half-life in humans and is accumulated in body tissues, particularly in the liver and kidneys. It is known to accumulate in the human kidney for a relatively long time, from 20 to 30 years, and at high doses is also known to produce health effects on the respiratory system and has been associated with bone disease (WHO, 1993). Moreover, carcinogenic effects of Cd have been reported (Dertmar *et al*., 1994). Thus, the soil analysis of Pb and Cd in environmental samples, including surface soils, has become increasingly important in urban environmental assessment because of their long residence time and their potential toxicity (Sutherland, 2000).

Soil Pb levels vary, depending mainly on the location and proximity to lead–based activities and vehicular density (Culbard *et al*., 1988). A concentration of 200–500 $\mu g\ g^{-1}$ of lead is commonly found in urban soil. The United States Centers for Disease Control and the U.S. Environmental Protection Agency (US-EPA) considered levels greater than 500 $\mu g\ g^{-1}$ to be hazardous (CDC, 1985); whereas Hafen *et al*. consider a 1000 $\mu g\ g^{-1}$ soil concentration of Pb in soil as hazardous waste (Hafen and Brinmann, 1996). Other researchers calculated "acceptable" levels of Pb in soil or dust between <100 and 1000 $\mu g\ g^{-1}$ (Reagan and Slibergled, 1990).

During the last two decades, measures have been taken to reduce Pb emissions from motor vehicles. For example, in the UK the Pb content of leaded petrol was reduced from 0.3g Pb /L to 0.134 g Pb/L in January 1986 (Wang *et al*., 1998), and the complete removal of Pb from petrol in the UK was mandated by legislation that came into force in January 2000 (Massadeh and Snook, 2002).

In Jordan, the Pb content in gasoline did not exceed 0.15 g/L (from 1983 until 2001) according to data obtained from The Jordanian Refinery of Petrol Company (JRPC, 2001), taking into account the international standard levels set by human health authorities, such as the World Health Organization (WHO). The potential health risks associated with Pb have therefore led to reduced Pb levels in Jordanian petrol. Thus, sales of unleaded petrol in Jordan have increased since March 1995. The Jordanian Refinery of Petrol Company has produced a large quantity of unleaded petrol to cover domestic needs. By the end of 2006, all gasoline produced by the refinery in Jordan will be unleaded. On the other hand, most vehicles in operation in Jordan have engines designed for leaded gasoline (JRPC, 2001). Thus, leaded petrol is still used in these vehicles.

The major objectives of this study were: (1) to evaluate levels of Pb and Cd contamination of selected roadside soils in two representative major roads in Irbid City; (2) to study the impact of vehicular traffic and location on the Pb and Cd content of roadside soils; and (3) to calculate the enrichment factors for Pb and Cd in roadside soil samples in order to evaluate the degree of enrichment above the normal "background" concentration.

Experimental

Reagents

All chemicals were of high purity analytical reagent grade. Lead nitrate (Scharlau) and cadmium nitrate (Scharlau) were used to prepare standards; 70% HNO_3 (Scharlau) and 35–38% HCl (Scharlau) were used for both extraction and acid digestion procedures. 1000 ppm stock solutions of the elements analyzed were prepared by acid dissolution of the appropriate masses of either lead nitrate and cadmium nitrate (López-García *et al*., 1996).

Collection of Samples

In this study, 260 samples were taken from roadside soil (surface scrapings) during the period of October 2001 to July 2002. At each location, three spots were taken within an area of 1 m^2 and 1–3 cm depth, and then pooled together before being transported to the laboratory. Samples were kept in a thoroughly pre-cleaned polyethylene bottles (glassware and plasticware used were soaked in 10% (v/v) HNO_3 for 24 hours and rinsed thoroughly with de-ionized water prior to use). The sampling points are designated as roadside soils from three different sites along the street with subscripts ($YURSD_1$), ($YURSD_2$), ($YURSD_3$) for Yarmouk University Street and ($IHRSD_1$), ($IHRSD_2$), ($IHRSD_3$) for Irbid-Howara Street, which were carrying about 18,000 and 21,000 vehicles per day for Yarmouk University and Irbid-Howara streets in Irbid city, respectively (Figure 1). Roadside samples were collected in polyethylene containers at regular weekly intervals, transported to the laboratory and dried in the oven at 105°C for 20 hours.

Methods and Instrumentation

The recommended extraction procedure used in this work was carried out as follows: (i) After first rejecting extraneous large objects and material above a size of ca. 2 mm the collected samples of roadside soil were dried for 20 hours in an oven (model D4C Genlab, Widnes, England) at 105°C to obtain constant weight. (ii) A quantitative weighed sample of about 0.5 g (using a sartorious analytical balance; model A 120 S), was extracted in a 25 ml mixture of 4M HNO_3 and 4M HCl with a ratio of (15:10) in a polyethylene

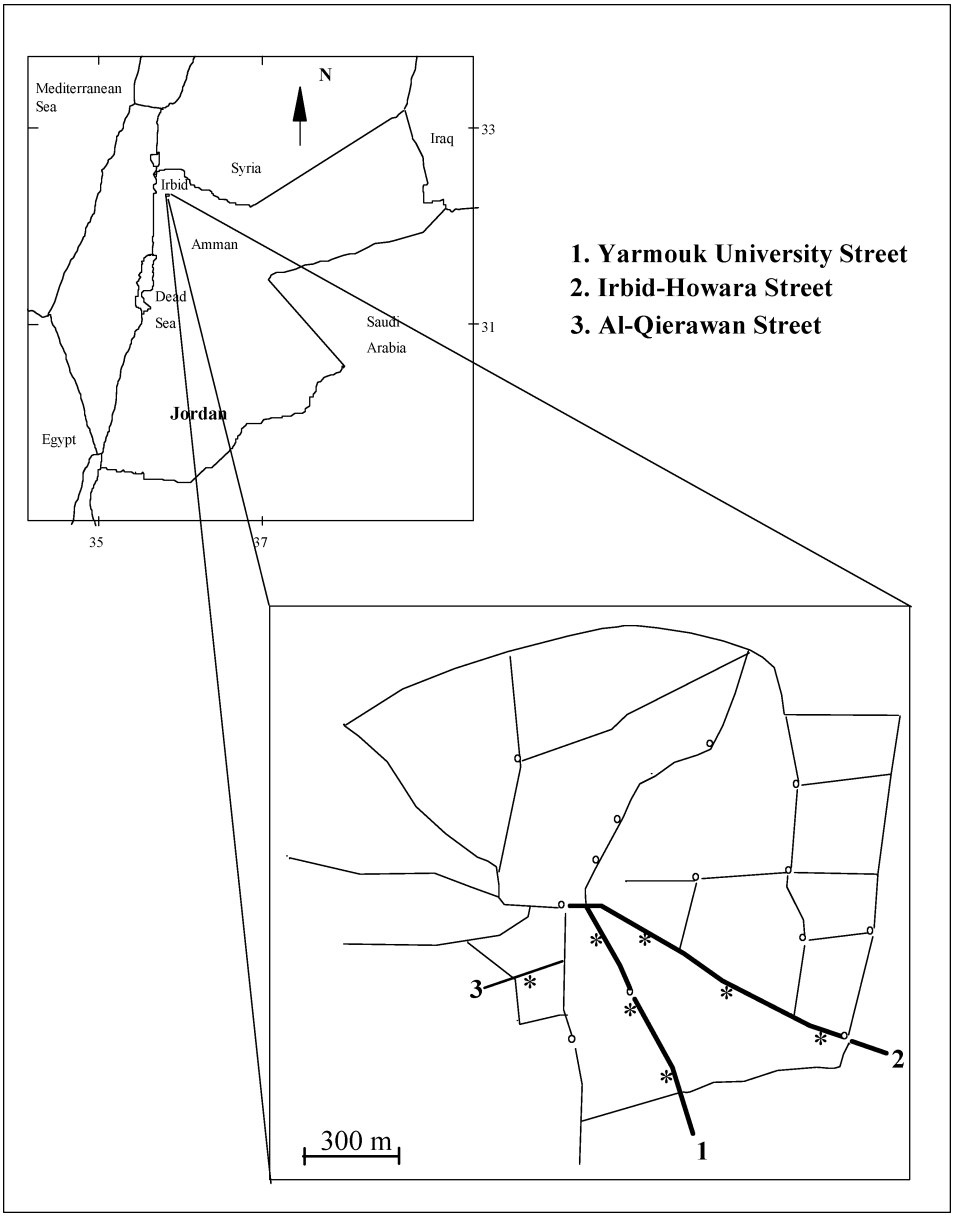

Figure 1. Jordan map shows the site of Irbid City; the area of study (* represents the sampling points).

container. (iii) These suspensions were stirred for 4 hours in a shaker-water bath (model D-3162 Kottermann labortechnik, type 3047, West Germany) at 105°C to achieve an efficient extraction. (iv) The suspension was filtered using a Whatman filter paper (No. 41) into a 25 ml volumetric flask; the volume was completed with de-ionized water up to the mark. (v) Aliquots of these samples were injected into the GFAAS tube. (vi) Calibrations for Pb and Cd were performed using the aqueous standards. (vii) Blanks were prepared in the same way but omitting the sample. Graphite furnace atomic absorption spectrometry (HGA-800, auto sampler AS-71, Perkin Elmer) was used for the determination of Pb and Cd. The statistical

Table 1
Pb and Cd levels (μg g^{-1}) in two reference materials, CRM 142 R and SRM2709, using the extraction

	Certified value (μg g^{-1})	Concentration found (μg g^{-1})
CRM 142 R		
Pb	40.2	38.9 ± 2.1
Cd	0.34	0.33 ± 0.018
SRM 2709		
Pb	18.9	18.4 ± 1.08
Cd	0.38	0.37 ± 0.021

analysis of the results for analysis of variance (ANOVA) and t-test were applied at 95% confidence level.

Analysis of Reference Materials

Two reference materials, CRM 142R (soil) and SRM 2709 (soil), were analyzed to assess the accuracy and reliability of the extraction method used throughout. The results obtained for five replicate injections are in agreement with certified values for Pb and Cd (Table 1).

Results and Discussion

Lead

Data were averaged based on a monthly basis to simplify presentation. Graphical presentation show the mean monthly Pb levels from three different sites along Yarmouk University Street (YURSD) (Figure 2a and Irbid-Howara Street (IHRSD); Figure 2b). Both of these roads are heavy traffic commuter routes. However, in Figure 2a there was no marked decrease in the overall level of Pb. Lead levels lie between 356 (344–369), 313 (304–322) and 305 (296–316) μg g^{-1} in YURSD1, YURSD2 and YURSD3, respectively. As shown in Figure 2b the overall Pb levels are 450 (436–464), 421 (412–431) and 423 (411–436) μg g^{-1} in IHRSD1, IHRSD2 and IHRSD3, respectively.

Lead levels in IHRSD are greater than those in YURSD. This may be attributed to the different compass direction for IHRSD (East-West) and YURSD (North-South). Secondly, traffic conditions are different. When the relationship between roadside soil Pb concentration and the distance from IHRSD was investigated, it was observed that the roadside soil Pb levels fall off with distance from Irbid Howara Street (Figure 3a). That is, the highest Pb concentrations for all samples were found in soil samples from the highly trafficked site, reflecting the long-term accumulation of vehicle related pollutants.

Samples collected at 5 meters before the traffic lights in one–way streets of IHRSD were compared with those samples collected at 5 meters after the traffic light. Results represented in Figure 4 show that Pb levels after the traffic light were slightly higher than Pb levels just before the traffic light throughout the period of study, a condition attributed to the effect of vehicle acceleration after stopping at the traffic lights.

For the roadside soil samples collected from the residential street (Al-Kierwan Street) and the busy road (Irbid-Howara Street), the results confirmed that greater traffic densities produce higher Pb levels (Figure 5).

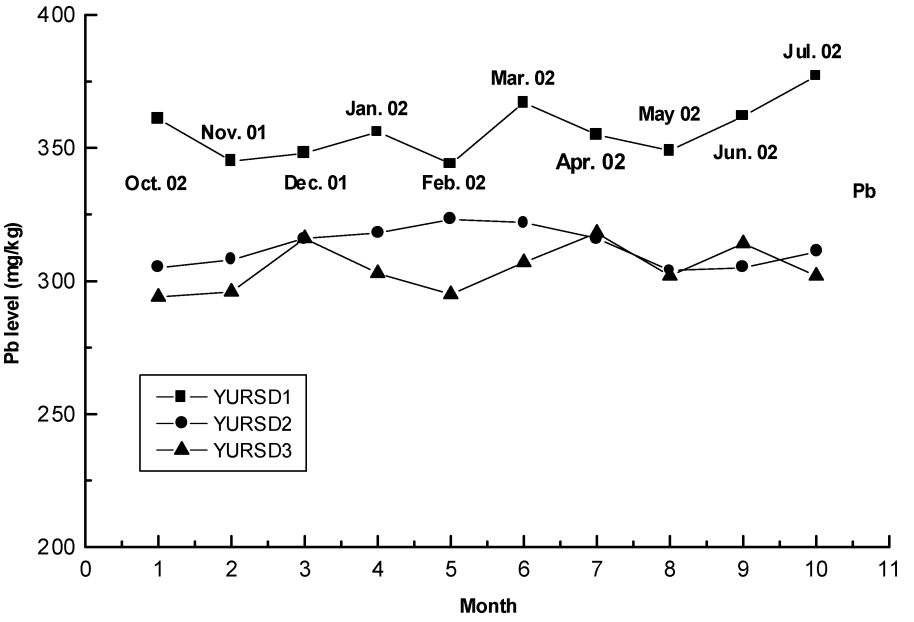

Figure 2a. The mean Pb levels (μg g^{-1}) from three different sites in Yarmouk University Street at monthly intervals from October 2001 to July 2002.

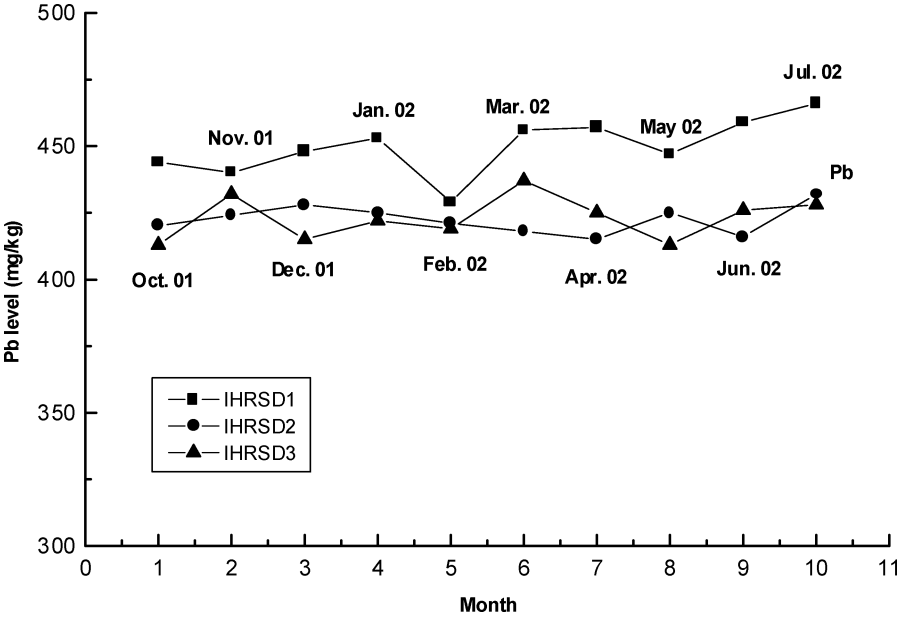

Figure 2b. The mean Pb levels (μg g^{-1}) from three different sites in Irbid-Howara Street at monthly intervals from October 2001 to July 2002.

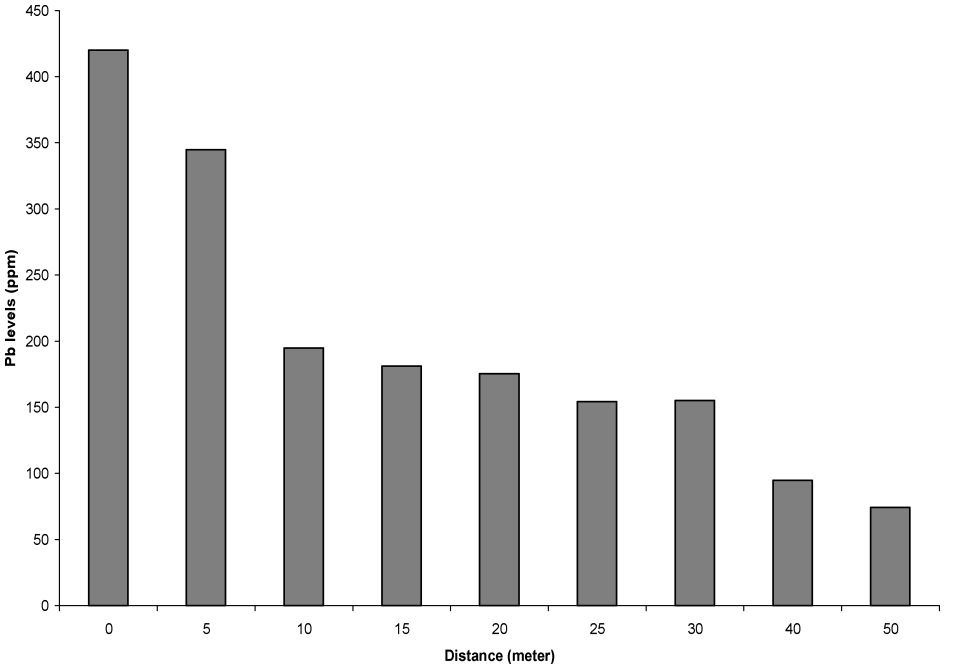

Figure 3a. The relation between Pb level (μg g^{-1}) and distance from the Irbid-Howara Street.

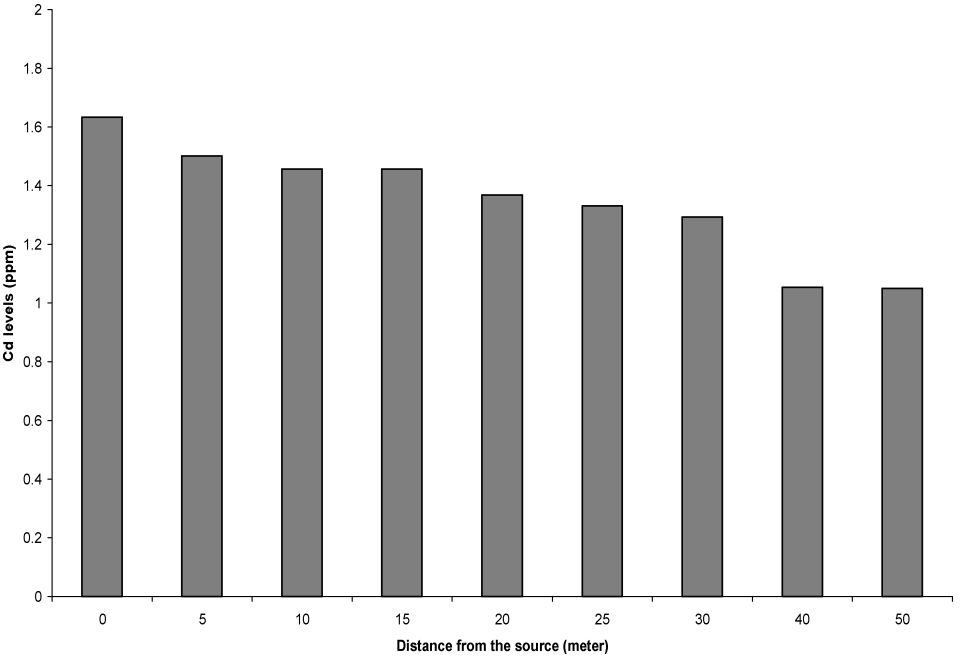

Figure 3b. The relation between Cd level (μg g^{-1}) and distance from the Irbid-Howara Street.

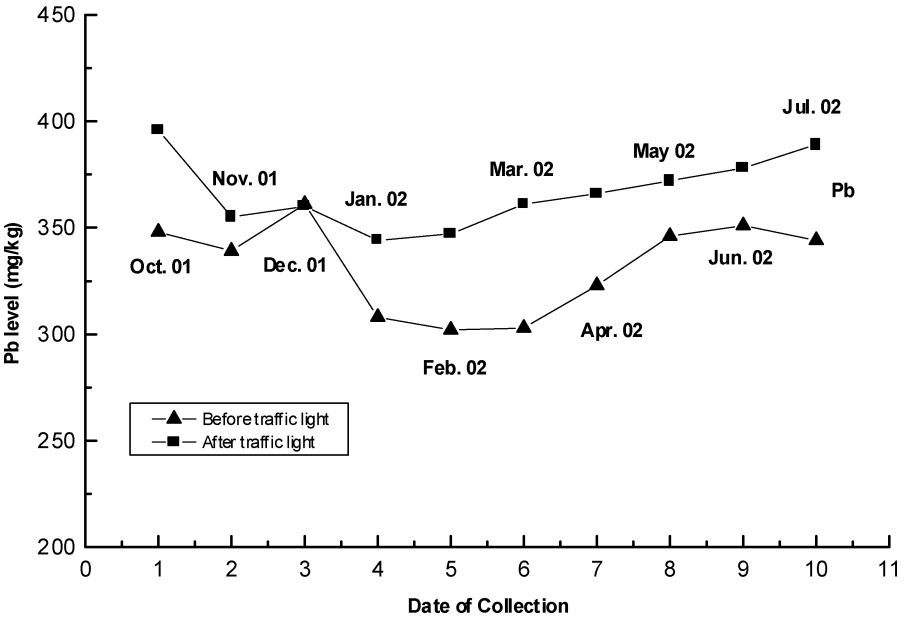

Figure 4. A comparison between Pb level (μg g^{-1}) before and after the traffic light in a representative street in Irbid city.

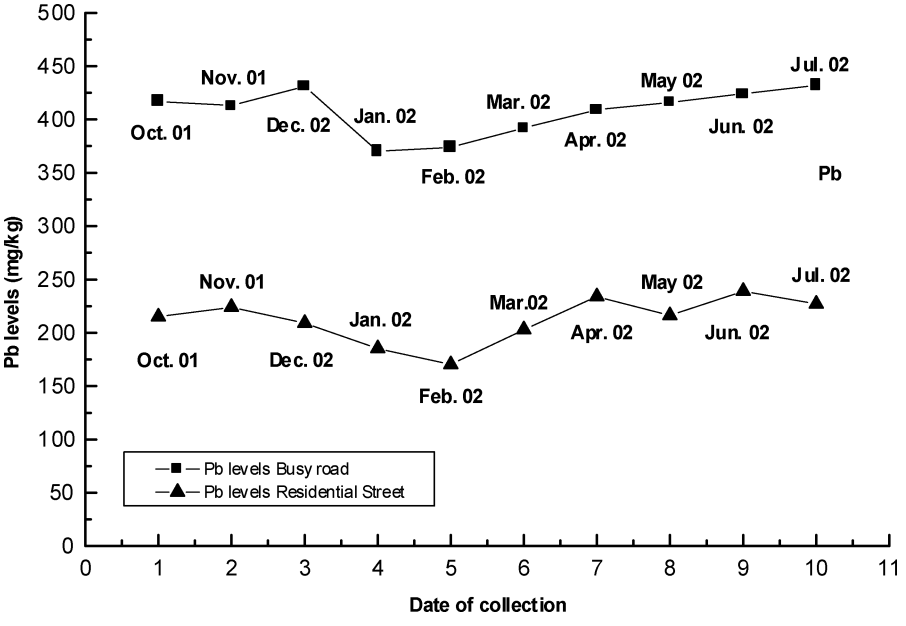

Figure 5. A comparison between Pb level (μg g^{-1}) in a representative busy road and residential street in Irbid City.

Table 2
A comparison between Pb (mean ± SD) μg g^{-1} at the entrance of petrol station and 50 m away from the petrol station

Collection date	Pb (mean ± SD) μg g^{-1} at petrol station	Pb (mean ± SD) μg g^{-1} at 50 meters away from the petrol station
26-10-2001	489 ± 12	401 ± 9
25-11-2001	467 ± 9	421 ± 13
12-12-2001	456 ± 11	395 ± 12
17-1-2002	441 ± 12	380 ± 13
15-2-2002	423 ± 10	392 ± 14
3-3-2002	451 ± 9	425 ± 12
11-4-2002	481 ± 8	440 ± 14
11-5-2002	479 ± 13	433 ± 24
18-6-2002	498 ± 7	430 ± 23
15-7-2002	509 ± 9	435 ± 15

Results for samples collected from the Irbid-Howara petrol station show that the mean Pb levels were much higher and significantly different compared with those samples collected at 50 meters away from the petrol station, as shown in Table 2.

Cadmium

The results of average Cd levels in roadside soil samples from three different sites collected in Yarmouk University Street and Irbid-Howara Street during the 10–month period from October 2001 to July 2002 are represented in Figures 6a and 6b, respectively. For Yarmouk University Street, it is observed that the overall level of Cd lies between 1.108–1.180 μg g^{-1} for YURSD1, 1.112–1.180 μg g^{-1} for YURSD2 and 1.107–1.163 μg g^{-1} for YURSD3. In Irbid-Howara, the overall Cd level lies between 1.105–1.160 μg g^{-1} for IHRSD1, 1.104–1.177 μg g^{-1} for IHRSD2 and 1.083–1.182 μg g^{-1} for IHRSD3. These results indicate that Cd levels among the sites of each road are not significantly different. This indicates that the existence of Cd in roadside soils may be due to the tire erosion.

Cadmium levels in roadside soils decrease with distance from the main road (Figure 3b). Also, as previously observed for Pb, the Cd levels in the heavy traffic areas (Irbid-Howara) were greater than Cd levels along the residential street (Al-Kierwan Street). This feature is attributed to the wear and tear of tires, and the greater traffic density on the busy road compared to the residential street.

Enrichment Factor

The enrichment factor (EF) for heavy metals in roadside soils gives an indication of the degree of pollution in contaminated sites. The EF may be calculated according to Ogunsola *et al*. (1994), and expressed as:

$$(\mathrm{EF})_{\mathrm{crust}} = \frac{(C_i/C_{Al})_{\mathrm{sample}}}{(C_i/C_{Al})_{\mathrm{Crust}}}$$

where C_i is the concentration of the element considered in the sample or the crust and C_{Al} is the concentration of the reference element. Orenella reported that the levels of Al,

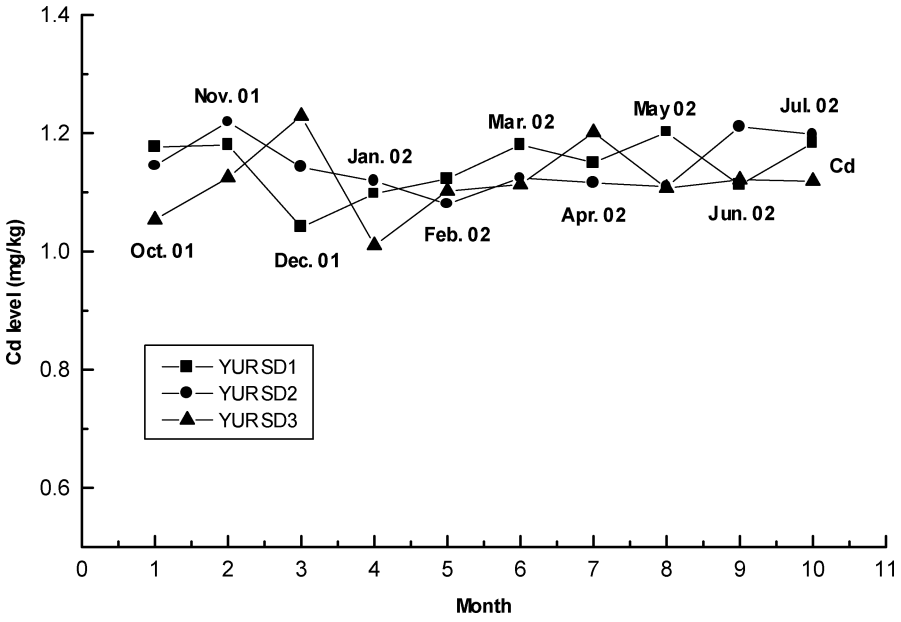

Figure 6a. The mean Cd levels (μg g^{-1}) from three different sites in Yarmouk University Street at monthly intervals from October 2001 to July 2002.

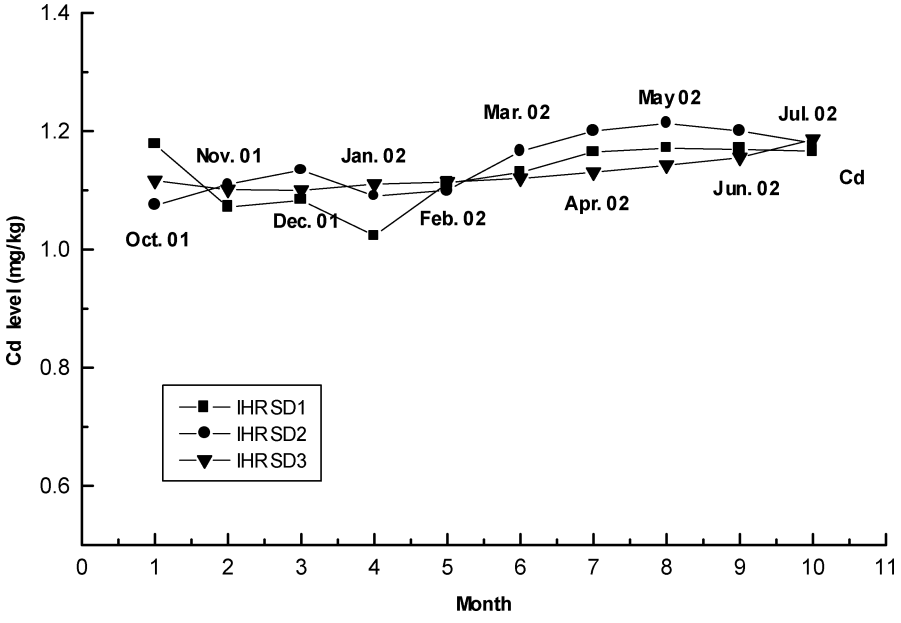

Figure 6b. The mean Cd levels (μg g^{-1}) from three different sites in Irbid-Howara Street at monthly intervals from October 2001 to July 2002.

Pb and Cd in the earth crust are 81300, 16 and 0.15 ppm, respectively (Orenella *et al*., 2002). Other researchers used Al as the normalizing element, and proposed a mass per area enrichment ratio (MAER) in order to characterize anthropogenic pollution (Sutherland, 2000). However, our results show that the average enrichment factors for Pb were 655 and 826 in YURSD and IHRSD, and the average enrichment factors for Cd were 252 and 237 in YURSD and IHRSD, respectively. This infers that sites on both streets are impacted with Pb and Cd. Moreover, there was a significant difference in the enrichment factor for Pb between YURSD and IHRSD ($t_{cal.}$ (2.54) is greater than the $t_{crit.}$ (2.086). For Cd, there was no significant difference in the enrichment factor between YURSD and IHRSD ($t_{cal.}$ (0.7) is less than the t critical value). The enrichment factor for Pb and Cd decreased with distance from the source of contamination (Figure 7), because the concentration of Pb and Cd decreased in the more distant sampling positions. (Figures 3a and 3b).

Statistical Treatment of Data

The Minitab statistical package and Microsoft Excel were used for ANOVA and t-test calculations. Analysis of variance (ANOVA) demonstrated that there were significant differences in the average Pb concentration in the three different sites over the period of study. Based on ANOVA, the $F_{crit.}$ value (4.0068) is less than the $F_{cal.(1,58)}$ value (165) for the entire sample pools between sites YURS1 and YURS2. Results show that there were significant differences in Pb levels in these three sites. For IHRSD, ANOVA shows that the $F_{cal.}$ value (4.0068) is less than the $F_{cal.(1,58)}$ (77.74) for the entire sample pools between sites IHRS1 and IHRS2.

Moreover, there was a significant difference in the average Pb levels between IHRSD and YURSD over the period of 10 months (t_{cal} value was 47.11 and the $t_{crit.}$ is 2.13. Also, t-tests for samples collected before and after traffic light shows that there was a significant difference in Pb levels, $t_{cal}(3.68) > t_{crit}$ (2.08).

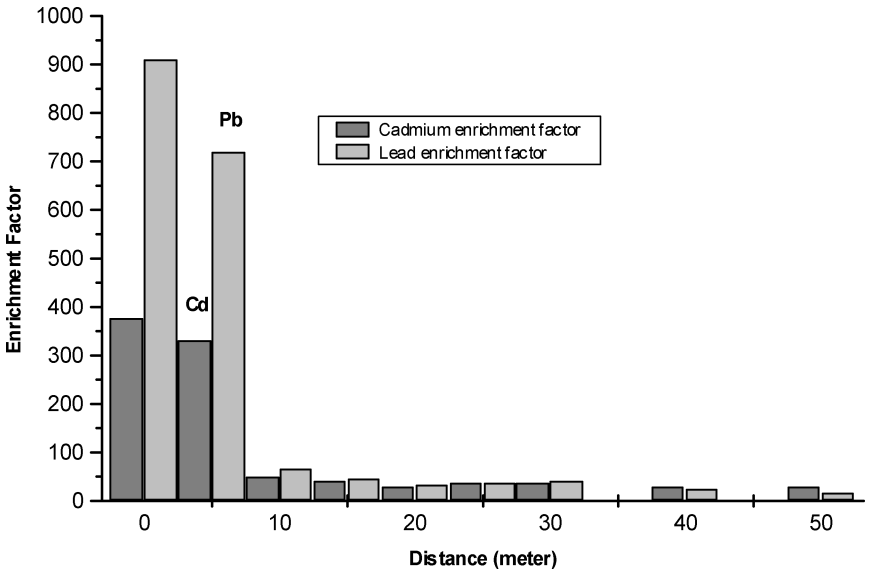

Figure 7. The enrichment factor for Cd and Pb in roadside soil samples in IHRSD as a function of distance with respect to crust composition.

For samples collected from the residential street (Al-Qirwan Street) and the busy road (IHRSD), the results show that the $t_{cal.}(23) > t_{crit}$ (2.08). This implies that there was a significant difference between the roadside soil samples collected from a residential street (Alqierwan Street) and the busy road (IHRSD). Also, for samples collected from the Irbid-Howara petrol station, the results show that the mean Pb levels were much higher and significantly different compared with those samples collected at 50 meters away from petrol station, the absolute $t_{cal.}$ (5.01) being more than the t_{crit} (2.08) (Table 2).

Results for Cd show that there was no significant difference in Cd levels between IHRSD and YURSD over the time period that samples were collected, (t_{cal} (0.97) is less than $t_{crit.}$ (2.09)). Also, ANOVA confirmed that there was no significant difference in Cd levels over the period of sample collection, ($F_{(1,18)}$ calculated (0.9386) is $<F_{tab}$ (4.413)).

To test the accuracy and feasibility of the Pb and Cd determination using GFAAS, standard reference materials (CRM 142R and SRM 2709) were extracted with the same procedure mentioned before, and analyzed together with samples (Table 1). Results of five replicate analyses are consistent with the certified values.

Conclusions

Lead and Cd have been confirmed as major metals causing road traffic pollution in Jordan. This form of pollution is attributed to emissions from motor vehicles (Jardat and Momani, 1999; Akhteer, 1993; Martin *et al*., 1988). The amount of Pb deposited along the roads was variable, being dependent on factors such as traffic volume and distance from the road. Results of this study show that the traffic volume caused a significant increase in Pb content in roadside soils. Higher Pb concentrations were found in sites with a higher traffic volume on main roads and in the entrance of petrol station. It seems reasonable to conclude that Pb and Cd in roadside soil levels are significantly higher on busy roads compared to residential roads. Pb and Cd concentration levels in roadside soil decline with distance from the main roads. We therefore conclude that there is a correlation between the roadside soil concentration of heavy metal and the distance from the road.

It can also be observed that the frequencies with which motor vehicles stop, start, and accelerate, especially at traffic lights, may help to explain differences in the Pb levels in roadside soil. It is clear that Pb levels vary from time to time and depend on the volume of traffic. The Pb level at 30 meters from the busy road was around 155 μg g^{-1}, which suggests that this is the base-line level of lead in Irbid City due to sources of Pb other than Pb from combusted petrol.

The enrichment factors for Pb and Cd were high, indicating that both main roads of this study are significantly contaminated with Pb and Cd. The high enrichment coefficient for Pb and Cd (non-crustal) indicates anthropogenic sources of contamination for these metals, i.e. automobile emission products. The degree of such pollution is apparently dependent on the type, intensity, and duration of the activity.

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