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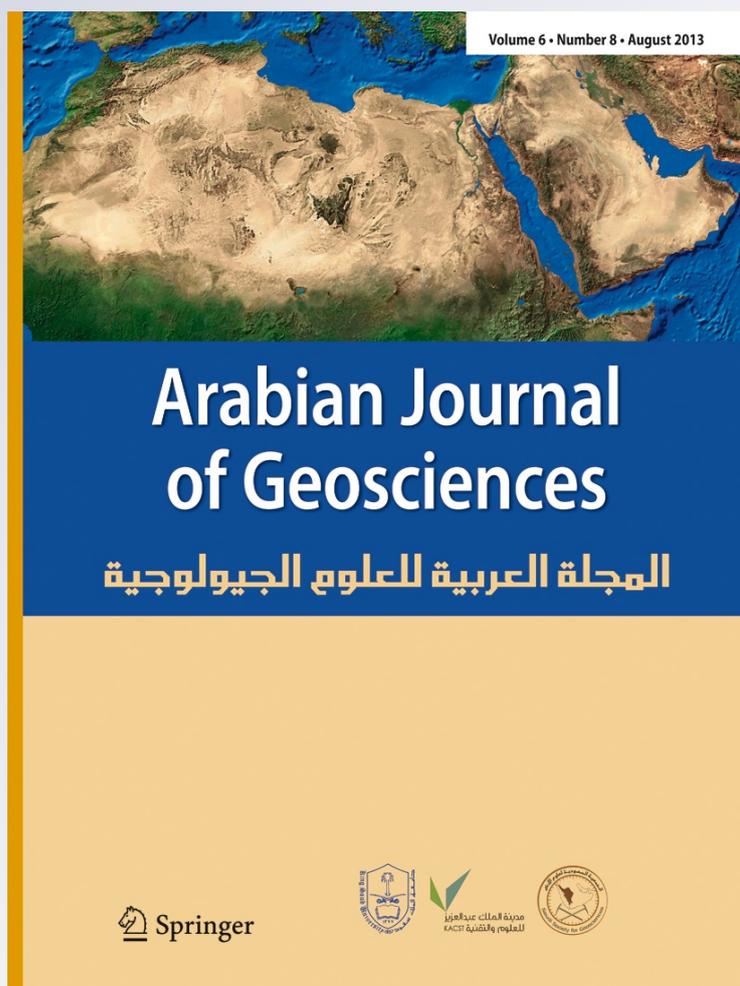
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Mohamed A. Habila · John P. Giesy

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Abstract Concentrations of lead (Pb) in domestic water and blood plasma in the Olya and Al-Batha regions of Riyadh City, Saudi Arabia were correlated ($r^2=0.03$, $p<0.0072$ and $r^2=0.37$, $p<0.00092$, respectively). Greater concentrations of Pb in domestic water of Olya and Al-Batha (0.0119 and 0.03 mg/l, respectively) were greater than concentrations of Pb in bottled water and was also greater than the concentration of 0.01 mg Pb/l recommended by both the World Health Organization US Environmental Protection Agency (USEPA). In Al-Batha, 52.2 % of the population had concentrations of Pb in blood that exceeded 10 $\mu\text{g Pb/dl}$, which

is the concentration used by USEPA to classify people as being at risk from effects of Pb. In Al-Batha, 17.5 and 22.5 % of the population exceeded 20–40 and $>40 \mu\text{g Pb/dl}$, respectively. In Olya, 37 and 10 % of the population had concentrations of Pb in blood that exceeded 10 and 20–40 $\mu\text{g Pb/dl}$, respectively, while none of the concentrations of PB exceeded 40 $\mu\text{g Pb/dl}$.

Keywords Pollution · Water quality · Human health · Middle East · Lead

A. M. Al-Othman
Department of Community Health Sciences,
College of Applied Medical Science, King Saud University,
Riyadh 11433, Saudi Arabia

Z. A. Al-Othman · G. E. El-Desoky · M. A. Habila
Chemistry Department, College of Science, King Saud University,
Riyadh 11451, Saudi Arabia

G. E. El-Desoky · M. A. M. Aboul-Soud
The Biochemistry Department, Faculty of Agriculture,
Cairo University,
Giza 12613, Egypt

M. A. M. Aboul-Soud (✉) · J. P. Giesy
King Saud University,
P.O. BOX 2245, Riyadh 11451, Kingdom of Saudi Arabia
e-mail: maboulsoud@ksu.edu.sa

J. P. Giesy
Department of Veterinary Biomedical Sciences and Toxicology
Centre, University of Saskatchewan,
Saskatoon, SK, Canada

J. P. Giesy
Department of Zoology, Center for Integrative Toxicology,
Michigan State University,
East Lansing, MI, USA

Introduction

Concentrations of Pb in humans are approximately 500-fold greater than those of pre-industrial times (Sridhar et al. 2001). Humans can be exposed to lead (Pb) from a variety of sources, including air, water, food, and housing (Seux and Dab 1994; Fischbein 1998). Exposure through any pathway including ingestion, inhalation, or dermal contact can cause significant toxicity (Lochitch 1993; Fischbein 1998; United State Department of Health and Human Services 1999). Lead can be absorbed through the digestive tract, the lungs, and the skin (USEPA 2000; Devenci 2006). One possible source of exposure to lead in the general population is drinking water, which can become contaminated by contact with the distribution network (Shannon and Graef 1989; Meyer et al. 1992; Tricard and Guillemot 1994; Watt et al. 1996; Conio et al. 1996; Gharaibeh et al. 1998; Adonaylo and Oteiza 1999; Guidotti 2004; Wasserman et al. 2004). The primary source of lead in drinking water comes from household plumbing and water supply distribution systems (Moore et al. 1977, 1979; Elwood et al. 1978; Pocock et al. 1983; Monty and Mark 1993; Guo 1997). Exposure to Pb through drinking water accounts, on average, for 10–20 %

of the total exposure (Agency for Toxic Substances and Disease Registry 2005; USEPA 2005). The World Health Organization (WHO) guideline for protection of human health is 10 $\mu\text{g Pb/l}$ in drinking water (World Health Organization 2004).

Whole blood has been the most widely used matrix to monitor for exposure of humans to Pb and has been calibrated to thresholds for toxicity. Recently, concentrations of Pb in human blood and subsequent intoxication have been linked to Pb in drinking water (Triantafyllidou et al. 2006, 2007). In blood, 97 % of Pb is associated with red blood cells (RBCs; Waxter 1998; International Program on Chemical Safety 2000; Arun et al. 2006) and the half life of Pb in RBCs is 2–3 weeks (Timbrell 2002). Clinical manifestations of Pb toxicity include effects on the central and peripheral nervous systems, the hematopoietic system, the renal system, and the gastrointestinal systems. Exposure of children to Pb can adversely affect the developing brain and result in learning defects, such as language skills (Bulletin of the World Health Organization 2000, 2004). Moreover, Pb interferes with synthesis of haem and porphyrin (Agency for Toxic Substances Disease Registrar 1989; Bulletin of the World Health Organization 2000). The result of the effects of Pb on porphyrin synthesis are less hemoglobin and appearance of coproporphyrin aminolaevulinic acid in urine (USEPA 2000). The Centers for Disease Control and Prevention states that concentrations of Pb greater than 10 $\mu\text{g/dL}$ are cause for Concern (Centers for Disease Control and Prevention 1991; Bulletin of the World Health Organization 2000).

In Saudi Arabia, people use two types of water, bottled water and water from the public domestic distribution system (unbottled water). The domestic water network passes water through pipelines twice a week where it is stored in containers on roofs of houses, before use for cooking, washing, and sometimes drinking. These containers can be a source of Pb from dust, pipes, or improperly soldered pipes and construction materials. In this study, concentrations of Pb in domestic drinking water and human blood were measured in two regions of Riyadh, Saudi Arabia. These included Olya, which is more modern and less populated and Al-Batha, which is older and more populous. Concentrations of Pb were determined in both bottled and domestic public (unbottled) water and whole blood was collected from the two areas. Correlations between concentrations of Pb in water and whole blood were determined.

Materials and methods

Sample location

This study was conducted in Riyadh, which is the capital of Saudi Arabia and its largest city. Riyadh is situated in the

center of the Arabian Peninsula, which is arid, but receives some rainfall and hail occasionally falls during winters. Riyadh has a population of approximately 4,260,000, which is 66 % Saudi and 34 % non-Saudi. Riyadh is divided into 15 municipalities, two of which were chosen for this study. The first district was Olya, the commercial heart of the city with accommodation, entertainment, dining, and shopping. In this district, water storage containers are generally well maintained and routinely cleaned. The second area, Al-Batha, is a district that has primarily residential but does have some industry. In Al-Batha, people generally do not maintain their water storage containers as frequently as they do in Olya.

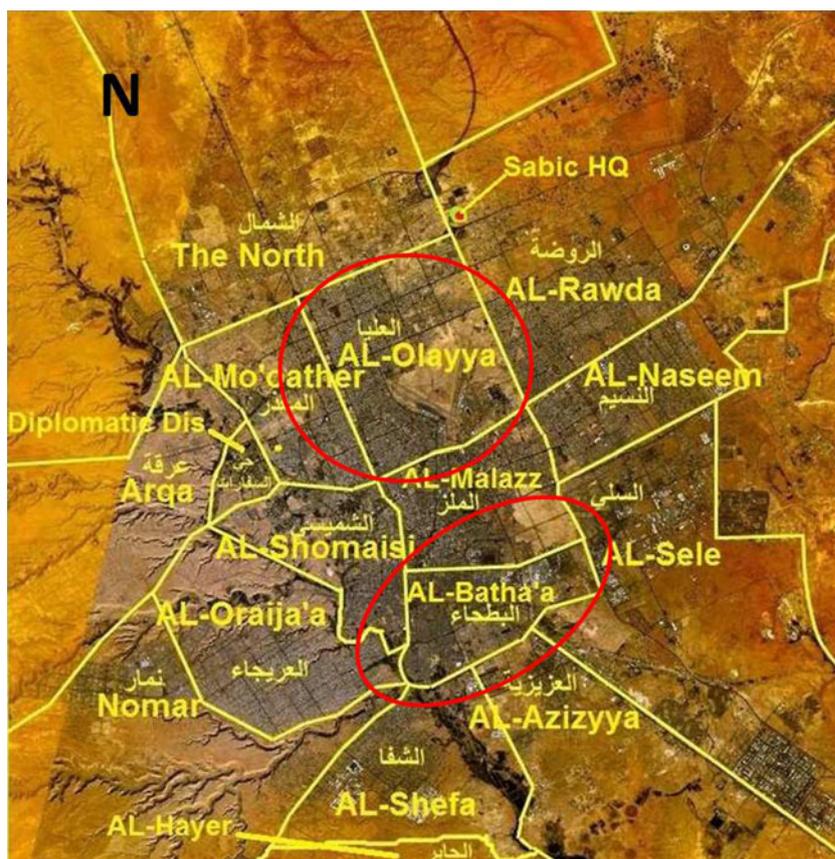
Collection of water samples

A total of 270 water samples (90 bottled and 180 unbottled) were collected from each district, each in three replicates, producing for each district 810 water samples. As shown in Fig. 1, for each district, the water sampling was distributed all over the study area from north to south and from west to east. Also, sampling was extended to cover areas closed to the study area (for Olya district, sampling extended to cover Alworood, El-rahmania, and Alsolimania; for Al-Batha district, sampling was extended to cover Alfoutah, Manfouha, Almarqab, Aderah, Alshemeisi, and Ghubaira). The unbottled water samples also included samples from the outlet of the water treatment units in Riyadh city which feed the two districts. The bottled water samples were collected from supermarkets distributed in all areas under study from the two districts. Samples of unbottled water were collected from houses and mosques (concrete constructed containers and plastic containers). Samples were collected into polyethylene bottles that had been prewashed with acid. Bottled water was purchased from local markets. Water samples were transferred directly to the laboratory, filtered, acidified with concentrated HNO_3 , and refrigerated at 4°C until quantification (Wardencki et al. 2002).

Water sample preparations and analysis

High purity chemicals were used in all tests. All reagents met the specifications of the Committee on Analytical Reagents of the American Chemical Society, which include: nitric acid (ultra pure HNO_3 , 70 %) from Wenlab, UK; hydrochloric acid (36 %), from BDH laboratory supplies, England; hydrogen peroxide from Petrochem, UK; and reagent Water (high purity, 18 M Ω). Water samples were prepared by adding HNO_3 and HCl according to methods described by Hageddorm (2008). A 100-ml aliquot of well-mixed water sample was transferred to a PTFE beaker, 2 ml of concentrated HNO_3 was added followed by 5 ml of concentrated HCl. Samples were covered and heated on a hot plate at 90–95°C until the volume had been reduced to

Fig. 1 Distribution of the water sampling from Olaya and AL-Batha districts



20 ml. Beakers were removed and allowed to cool. Walls of beakers were washed with reagent water then the digested solution was filtered through 0.45 μm filter paper. The final volume was adjusted to 50 ml.

Concentrations of Pb in digested samples were determined by use of inductively coupled, plasma atomic emission spectroscopy (ICPAES). A Thermo Scientific iCAP 6500 ICP-AES with SIK (standard introduction kit); glass concentric nebulizer/cyclonic spray chamber was used (van de Wiel 2003). A study to determine recovery (accuracy) of the analytical procedure was conducted by use of the matrix spiking method where samples were analyzed before and after adding known quantities of Pb. The spiked samples were processed for the analysis by the same procedure as unspiked samples. The mean recovery (accuracy) and precision were 98.5 and 1.6 (SD), respectively.

Collection and preparation of blood

A randomly selected population comprised of 170 (90 male and 80 female) healthy nonsmoking subjects was selected from each district. Each subject signed a consent form that authorized collection of blood and subsequent quantification of Pb. Medical technicians obtained whole venous blood samples using standard techniques. Samples were kept in the freezer till being analyzed.

Blood samples were digested with HNO_3 , HCl, and hydrogen peroxide (30 %) as described by Bukhari et al. (2005). Known volumes of blood samples were taken in a PTFE beaker, then 5 ml of concentrated HNO_3 and 2 ml of HCl were added. Samples were evaporated on a hot plate at medium heat (90–95°C) then a few drops of 30 % H_2O_2 were added. The digestion was continued till a clear solution was obtained. The beakers were removed and allowed to cool. The digested sample was washed from walls of beakers with reagent water then the digested solution was filtered through 0.45 μm filter paper. The final volume was adjusted to 50 ml. Concentrations of Pb in digested samples were determined by use of ICPAES as described above. A recovery study of the analytical procedure was conducted by spiking and homogenizing several already analyzed samples with varied amounts of standard solutions of Pb. The spiked samples were processed for the analysis by the same procedure as described above. The average recovery was 98.7 ± 1.1 %.

Statistical analysis

Results were analyzed by use of SPSS program (version17). Comparison between means was performed using Student's *t* test. Pearson correlation was conducted to determine the association between concentrations of Pb in water and blood.

Table 1 Concentrations of Pb in unbottled, bottled, both unbottled and bottled, and whole blood from Al-Olya and Al-Batha areas of Riyadh

	Al-Batha				Olya				Significance
	Mean	SD	Max	Min	Mean	SD	Max	Min	
Bottled water (mg/l)	0.0038	0.018	0.0061	0.0009	0.0038	0.001	0.0061	0.0009	
Unbottled water (mg/l)	0.03	0.047	0.022	0.0084	0.012	0.018	0.131	0.0047	0.00087
All Used Water (mg/l)	0.017	0.036	0.022	0.0009	0.0079	0.014	0.131	0.0009	0.0016
Sig. (Unbottled & Bottled Water (mg/l))	0.00092				0.0072				
Sig. (All Used & Bottled Water (mg/l))	0.00061				0.0047				
Blood Samples (µg/dl)	25	30	1.5x10 ²	1.4	10.0	10.0	40	0.3	0.00098
Male (µg/dl)	30	31	1.5x10 ²	2.4	14.5	13.0	40	0.3	
Female (µg/dl)	18	18	73	1.4	8.0	5.3	22	1.0	

Results

Concentrations of Pb in unbottled water from Olya and Al-Batha areas of Riyadh city are given (Table 1). Concentrations of Pb in unbottled water from the Olya area ranged from 0.0047 to 0.13 mg/l with a mean concentration of 0.012 mg/l, while concentrations of Pb in unbottled water from the Al-Batha area ranged from 0.0084 to 0.22 mg/l with a mean concentration of 0.03 mg/l. Concentrations of Pb in unbottled water from the Al-Batha area was significantly ($p < 0.00087$) greater than that of Olya. To estimate potential exposure, the average concentration of Pb in bottled and unbottled water was calculated for the two areas. Concentrations of Pb in the water used by the population (all used water) in the Al-Batha area were significantly ($p < 0.0061$) greater than that consumed by people in Olya. Concentrations of Pb in unbottled water from both Olya and Al-Batha were significantly ($p < 0.0072$ and 0.00092 , respectively) greater than concentrations of Pb in bottled water from Olya and Al-Batha, which were 0.012

and 0.03 mg/l, respectively. Unbottled waters used by people in Olya and Al-Batha were significantly ($p < 0.0047$ and $p < 0.00061$, respectively) greater than concentration of Pb, in bottled waters, which were 0.0079 and 0.017 mg/l, in Olya and Al-Batha, respectively. Concentrations of Pb in whole blood of people in Olya ranged from 0.3 to 40 µg/dl with a mean concentration of 10.0 µg/dl (Table 1). Concentrations of Pb in people from Al-Batha population ranged from 1.4 to 1.5x10² µg/dl with a mean of 25 µg/dl. The mean concentration of Pb in whole blood of people in Al-Batha was significantly ($p < 0.00098$) greater than that of people in Olya.

Concentrations of Pb in whole blood of both males and females from Al-Batha and Olya were correlated with concentrations in domestic water (Table 2). Mean concentrations of Pb in domestic water of Al-Batha was significantly correlated with the mean concentrations of Pb in the blood of males in Al-Batha ($r^2 = 0.1406$, $p < 0.0001$) and all people ($r^2 = 0.0650$, $p < 0.015$). There were no significant correlations between concentrations in whole blood of females and

Table 2 Correlation between concentrations of Pb in the blood and in unbottled drinking water of the Al-Batha and Olya and concentrations of Pb in the blood of males and females in Al-Batha and Olya

		Unbottled	Bottled
Al-Batha female (N=80)	Pearson correlation	0.005	0.005
	Sig. (two-tailed)	0.528	0.519
Al-Batha male (N=90)	Pearson correlation	0.005 ^a	0.010
	Sig. (two-tailed)	0.000	0.339
Al-Batha all population blood samples (N=170)	Pearson correlation	0.065 ^b	0.012
	Sig. (two-tailed)	0.015	0.304
Olya female (N=80)	Pearson correlation	0.003	0.016
	Sig. (two-tailed)	0.629	0.246
Olya male (N=90)	Pearson correlation	0.058 ^b	0.008
	Sig. (two-tailed)	0.030	0.425
Olya all population blood samples (N=170)	Pearson correlation	0.000	0.042
	Sig. (two-tailed)	0.903	0.053

^aCorrelation is significant at the 0.01 concentration (two-tailed)

^bCorrelation is significant at the 0.05 concentration (two-tailed)

concentrations of Pb in waters of Al-Batha. The mean lead concentration of Pb in unbottled water from the Olya area was significantly correlated ($r^2=0.0586$, $p<0.03$) with the mean concentration of Pb in whole blood of males in Olya. No significant correlations were found between concentrations of Pb in Olya unbottled water and concentrations of Pb in the blood of females from Olya (Table 2).

Risk categories for concentrations of Pb in the blood of individuals in Al-Batha and Olya are reported (Table 3). In Al-Batha, 52.5 % of the population exceeded the value of 10 μg Pb/dl established by WHO for the protection of human health. In Olya, 37.5 % of concentrations in blood exceeded 10 μg Pb/dl. In Al-Batha, 17.5 % of the population had medium risk (20–40 μg Pb/dl), while 10 % of the Olya population was in this range. In Al-Batha, 22.5 % of the population had concentrations of Pb that were classified as “high risk” (>40 μg /dl) while, none of the concentrations of Pb in Olya were in this range.

Discussion

Humans have suffered from lead poisoning throughout history and research on lead in drinking water has a long history. The first article published on lead poisoning was published in 1914 (Stainthorpe 1914). After that, the number of publications increased mainly during two periods: from 1992 to 1997 and then from 2004 to 2007 (Jie et al. 2010). Due to the lack of suitable water resources for human utilities in Saudi Arabia, it depends on treatment of groundwater, rainwater, and seawater through the public Saudi water network. The water network pumps water to houses only 2 days a week. The residents store water in concrete tanks constructed under the houses, and then the stored water is pumped to tanks on the roofs of houses to provide water pressure. This water is used directly for cleaning and washing, but if it is used for drinking or cooking, it is passed through filters. These filters usually are activated carbon or cotton and very few people use reverse osmosis units in their homes. Activated carbon or cotton filters, can remove impurities and adsorb metals and organic pollutants.

Table 3 Risk categories of based on concentrations of Pb in the blood of the Al-Batha and Olya populations of Riyadh, Saudi Arabia

Categories	Concentration of BLLs in micrograms per deciliter	Al-Batha population (%)	Olya population (%)
Safe	<10	47.5	62.5
Low risk	10–20	12.5	27.5
Medium risk	20–40	17.5	10
High risk	>40	22.5	0

However, the fast saturation of these filters requires frequent replacements (weekly or monthly) depending on their capacities and the intensity of use. In particular, regular filter maintenance in Al-Batha district is seriously lacking.

Concentrations of Pb in unbottled water from the Olya and Al-Batha districts exceeded concentrations recommended by WHO and the USEPA. The significantly ($p<0.00087$) greater concentrations of Pb in unbottled water from Al-Batha might be attributed to deposition and leaching from concrete tanks and corrosion of pipelines (Subramanian and Connor 1991; Conio et al. 1996; Isaac et al. 1997; Korshin et al. 2000). These authors suggested that release of Pb to drinking water is mainly due to corrosion of lead-bearing plumbing. Another explanation might be due to falling dust and impurities during water storage above house roofs. But it cannot be from the sources of water because the Saudi network water are subjected to water treatment units which employ reverse osmosis technique to remove toxic metals. Also, this is confirmed by water sampling from the outlet of the water treatment units and lead was not detected in these samples by our method.

The fact that concentrations of Pb in water were related to concentrations of Pb in the blood of people in both the Al-Batha and Olya districts suggests that people in these areas are exposed to lead and that concentrations of Pb in the blood exceed the guidelines suggested by Bulletin of the World Health Organization (2000) and that these individuals are at increased risks for lead poisoning. Symptoms of lead poisoning include damage to peripheral nerve function (Mayer and Wilson 1998; AOEC 2007). These symptoms have been exposed in occupationally exposed workers with a mean concentration of 36.9 μg Pb/dl in blood, while those that were not occupationally exposed and had a mean concentration of 10.5 μg Pb/dl in blood did not exhibit these effects. Thus, concentrations greater than 40 μg Pb/dl are associated with adverse effects on peripheral nerves (Chia et al. 1996). For this reason in this study, individuals were classified into groups based on the magnitude of adverse effects as follows: <10 μg /dl, “safe”; 10–20 μg Pb/dl, “low”; 20–40 μg Pb/dl, “medium”; and >40 μg Pb/dl, “high”. Based on this classification scheme, the Olya population was at “low” risk of adverse effects from exposure to Pb, while the Al-Batha population had a “high” potential for adverse effects due to exposure to Pb. Since the concentrations of Pb in unbottled water were significantly correlated with concentrations of Pb in the blood of both the Olya and Al-Batha populations, drinking water is a likely source (Table 2). These results are in accordance with Watt et al. (1996) who mentioned that the proportion of concentrations of Pb that are greater than 10 μg /dl that can be attributed to concentrations of Pb in tap water ranges from 63 to 76 %, depending on the type of water sample on which the estimate is based. Also, it has been reported that tap water

remains the most important environmental determinant of Pb in maternal blood (Meyer et al. 1992).

No previous studies of concentrations of Pb in water or human blood have been conducted in Saudi Arabia thus, it is impossible to compare to other regions of the country or to previous times. However, concentrations of Pb in the blood of people in the two regions of Riyadh can be compared to other regions of the world and to occupationally exposed populations. The mean concentration (10.0 µg Pb/dl) in the blood of the Olya population was greater than that observed for other countries. For example, in Australia concentrations were <10 µg Pb/dl, while in and New York the mean concentration was 1.8 µg Pb/dl. The concentration of Pb in the blood of people in Olya was less than that reported for people in Tehran (12.37 µg Pb/dl) and Pakistan (13.9 µg Pb/dl; Farzin et al. 2008). Concentrations of Pb in the blood of people from Al-Batha (25.08 µg Pb/dl) was also greater than those in other areas, but less than occupationally exposed individuals. For example, 35.5 % of concentrations of Pb in the blood of bus drivers in Tehran exceeded 50 µg Pb/dl (Abdollahi et al. 1995). Concentrations of Pb in 94 % of copy center workers, exceeded 49.9 µg Pb/dl (Abdollahi et al. 1996a), while those of 50.7 % of paint factory employees exceeded 50.71 µg Pb/dl (Abdollahi et al. 1996b).

Conclusions

Concentrations of Pb in unbottled water in the Olya and Al-Batha districts of Riyadh, Saudi Arabia, which were 0.0119 and 0.03 mg Pb/l, respectively, were significantly ($p < 0.0072$ and 0.00092 , respectively) greater than those in bottled water and also greater than the concentration of Pb of 0.01 mg/l, recommended for drinking water by WHO and USEPA. Thus, using storage systems in Riyadh results in greater exposure to Pb than if only bottled water were consumed.

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