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Biological monitoring of glazers exposed to lead in the ceramics industry in Iran

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Exposure to heavy metals, particularly lead, takes place in the ceramics industry. Lead is used in glaze to produce smooth and brilliant surfaces; thus, there is a likelihood of occupational adverse effects on humans. Urine samples were collected from 49 glazers at the start and end of the work shifts (98 samples). Solid phase extraction was used for separation and pre-concentration of the analyte. Samples were analysed by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). Lung function tests were performed on both control and lead exposed subjects. Statistical analysis of covariance (ANCOVA) was used to evaluate the data obtained. The concentration of lead in glazers was 6.37 times higher than in the control group. Lung functions were significantly lower in the glazers compared to the control group ($p < 0.001$). Results showed that poor ventilation systems, overtime work and work history are effective determinants of high exposure levels.

Keywords: glazer; urine lead; ceramics industry; spirometry; inductively coupled plasma-atomic emission spectroscopy

1. Introduction

Lead compounds are used in the production of glazes. Such compounds can decrease the melting point, increase surface smoothness and brilliance, and reduce cracking. The main hazard of using lead in glazes is its adverse effects on the body system. All glazers are in contact with lead during the mixing, storing and handling of the glazes in the powdered form. They are also exposed to lead in the process of glaze application to the ceramic wares. Cooking, serving and storage of food in glazed ceramic dishes can release lead into the foodstuff.[1,2] Acidic or spicy foods such as tomatoes and chilli may increase excreted lead, because lead is more soluble at low pH.[3,4]

Groups with a high risk of exposure to lead are the relevant industry workers and young children. Smelters, battery makers, ceramic makers, ship burners and construction workers can be considered as groups at risk in industry. Sources of lead exposure for children include soil, dust, drinking water, ceramics, lead-based paint, painted glass, industrial emissions and toys.[5] Lead is brought home on the skin, hair and clothing of the workers, therefore, this metal can be ingested or inhaled by children as household dust.[6,7] This metal affects the central and peripheral nervous systems, renal, gastrointestinal, cardiovascular, skeletal and reproductive organs.[8–10] Symptoms of exposure to lead include muscle weakness, arthralgia, abdominal pain, general fatigue, appetite and weight loss, nausea, vomiting, diarrhoea, constipation, insomnia, irritability and headache.[9,11] Based on symptoms observed, the International Agency for Research on Cancer (IARC) has classified lead as possibly carcinogenic to humans (Class 2B).[5]

The objectives of this study were to determine the lead levels in the urine of glazers in the ceramics industry and to find out whether a relationship exists between work history, work shift, using mask, kind of job, overtime work, ventilation system, age, weight and height, and the urinary concentration of lead. Investigation of occupational exposure to lead and lung function in glazers is another objective of this study. Many approaches have been considered to extract different samples before analysing the compound of interest.[12,13]. The extraction procedure optimized in our previous studies was applied to make samples as purified and concentrated as possible.[14–19]

2. Method

A case-control study was carried out in the glazing units of the ceramics industry. The study group contained 49 workers employed in 15 tile factories and 6 pottery workshops; the control group consisted of 55 office workers. The control group had never been occupationally exposed to lead. The range of glazer ages was 22–50 years with a mean

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value of 30.67 years. For the control group, the range was 23–50 years with a mean value of 32.6 years. The work shift length was 8 h with two shifts per day.

Questionnaire forms were completed for both case and control groups. Different variables including overtime work, work history, work shift, kind of job, using mask, ventilation system, age, weight and height were evaluated. There were no significant differences between glazers exposed to lead and the control group concerning age, weight and height. Urine samples were taken at the start and end of the work shifts of glazers (98 samples). The control groups were sampled just once (55 samples).

Samples were collected from tile and pottery glazers before the start of work and at the end of the work shift. At the end of shift, workers were asked to remove their work clothes and advised to wash their hands before sampling to reduce the potential for any contamination; 20 ml urine was collected in acid-washed polyethylene containers and stored at 4°C until the analysis time.

A solid phase extraction system was used for separation and pre-concentration of lead. Cartridges were filled with 500 mg amberlite XAD-7 resin. In order to remove organic and inorganic contaminants, the amberlite XAD-7 resin was washed with ethanol, water and 1 M HCl. Urine samples (2.5 ml) were adjusted at pH 9 and the lead present in the samples was chelated with ammonium pyrrolidine dithiocarbamate (APDC). Then, samples were diluted to 25 ml with distilled water. The solution was then passed through the sorbent at a flow rate of 5 ml/min under gravity force. The column was then washed out with water and 1 M NaOH. The retained analyte was eluted by 15 ml of 2 M HNO_3.

Determination was performed by the use of inductively coupled plasma-atomic emission spectroscopy (ICP-AES) (SPECTRO, ARCOS, Germany). Urinary lead concentrations were recorded in terms of μg/L.

Lung function tests were performed on both control and lead exposed subjects using a Vitalograph 2120 spirometer (Vitalograph, Buckingham, UK). The spirometer was calibrated daily with a one litre calibration syringe and operated within a temperature range of 20–25 °C. Tests were performed in a standing position and conducted according to American Thoracic Society (ATS) recommendations. Throughout this experiment, forced vital capacity (FVC), forced expiratory volume in 1 s (FEV_1) and forced expiratory flow ($FEF_{25–75}$) were measured. The FEV_1/FVC ratios were calculated in terms of percentage.

Statistical analysis was performed using SPSS 11.5 software. Comparisons between control group and glazers were made with independent samples *t*-test. Paired *t*-test was used to assess the changes between pre- and post-shift measurements for the glazers. After controlling for the pre-shift, analysis of covariance (ANCOVA) was used to evaluate the effect of each factor on post-shift measurement.

3. Results

Important demographic characteristics and the results of the lung function tests for both glazers and controls are shown in Table 1. There were no significant differences for age, height and weight between case and control groups. Mean values of FVC, FEV_1, FEV_1/FVC and $FEF_{25–75}$ were lower in the exposed group compared to the control group (Table 1). Analysis of the lung function tests revealed significant differences in all spirometric parameters between the glazers and the control group ($p < 0.001$) (Table 1).

Urinary lead levels at the start of the work shift had limitation from 29 to 156 μg/L, with a mean value of 72 μg/L. At the end of the work shift, samples were limited from 38 to 393 μg/L with a mean value of 132.82 μg/L. A significant difference was found in the before and after shift values. As can be seen in Table 2, there were significant differences in the mean concentration of lead between the exposed and the control groups. Table 3 shows the results obtained for lead concentration based on the different parameters considered in this study.

Glazers working in the morning shift have slightly higher urinary lead compared to the evening shift (134.87 μg/L versus130.84 μg/L). ANCOVA showed that after adjusting for base line urinary lead level (pre-shift value) no significant cross shift differences in urinary lead levels

Table 1. Characteristics of the study groups.

Variables	Glazer (n = 49) $M \pm SD$	Control (n = 55) $M \pm SD$	p
Age (years)	30.67 ± 5.88	32.60 ± 7.10	0.12
Height (cm)	174.50 ± 8.60	176.42 ± 9.06	0.26
Weight (kg)	77.44 ± 11.70	78.25 ± 10.60	0.70
BMI	25.59 ± 4.39	25.33 ± 4.29	0.76
Overtime work (h)	10.05 ± 14.90	0.83 ± 2.74	<0.001
Work history (months)	51.53 ± 33.44	76.32 ± 67.92	0.02
FVC (L)	4.24 ± 0.67	4.74 ± 0.66	<0.001
FEV1 (L)	3.50 ± 0.70	4.25 ± 0.70	<0.001
FEV1/FVC (%)	80.99 ± 8.16	88.85 ± 5.60	<0.001
FEF25–75 (L)	3.47 ± 0.80	4.49 ± 0.69	<0.001

Note: BMI = body mass index; FVC = forced vital capacity; FEV1 = forced expiratory volume in 1 s; FEF = forced expiratory flow.

Table 2. Urinary lead concentrations in pre-and post-shift samples in the glazers and control group.

Group	$M \pm SD$	t	p
Pre-shift glazer	72 ± 34.47	8.74	0.005
Control	20.84 ± 14.28		
Post-shift glazer	132.82 ± 90.86	4.4	<0.001
Control	20.84 ± 14.28		
Pre-shift glazer	72 ± 34.47	− 3.7	0.001
Post-shift glazer	132.82 ± 90.86		

Table 3. Urinary lead concentrations in the glazers in Meybod, Iran.

Factors		No.	Pre-shift $M \pm SD$ (µg/L)	Post-shift $M \pm SD$ (µg/L)
Work shift	Morning	24	70.29 ± 35.18	134.87 ± 99.13
	Evening	25	73.64 ± 34.42	130.84 ± 84.19
Using mask	No	19	74.84 ± 37.38	167.28 ± 89.2
	Yes	30	70.2 ± 33.03	112.87 ± 87.03
Kind of job	Pottery glazer	16	86.12 ± 32.18	154.3 ± 98.85
	Tile glazer	33	65.15 ± 33.9	125.05 ± 87.97
Ventilation system	Non standard	36	73.14 ± 33.38	160.19 ± 90.75
	Standard	13	68.85 ± 38.57	57 ± 23.06
Work history (months)	≤24	14	75 ± 35.47	75.93 ± 41.36
	25–60	18	62.28 ± 25.07	160.94 ± 92.55
	≥61	17	79.82 ± 41.17	154.5 ± 102.13
Overtime work (h)	No	29	69.35 ± 34.38	106.37 ± 81.95
	1–20	11	72.81 ± 35.63	151.54 ± 105.71
	21–60	9	79.55 ± 36.24	206.25 ± 55.8
BMI	<20	7	72 ± 40.57	122.57 ± 95.69
	20–25.99	18	66.55 ± 26.83	149.76 ± 87.96
	26–30.99	19	72.31 ± 37.7	121.9 ± 99
	31–40.99	5	90.4 ± 42.13	131.5 ± 81.62
Age (years)	≤25	10	76.3 ± 34.14	110.67 ± 66.65
	26–31	22	69.27 ± 34.43	153.1 ± 96.34
	≥32	17	73 ± 36.46	110.1 ± 93.63

Note: BMI = body mass index.

Table 4. Results of analysis of covariance (ANCOVA) for post-shift urine lead by independent factors controlled for pre-shift lead.

Source	F	p
Shift	0.004	0.95
Mask	3.75	0.06
Job	2.48	0.12
Ventilation	13.55	0.001
Work history (months)	4.07	0.02
Overtime work (h)	5.29	0.009
BMI	0.13	0.94
Age (years)	0.55	0.58

Note: BMI = body mass index.

were found (Table 4). During mixing or handling, those glazers not using protective masks and standard ventilation system had higher exposure levels to lead compared to the glazers using with such protections (167.28 µg/L versus112.87 µg/L and 160.19 µg/L versus 57 µg/L, respectively). ANCOVA also showed that the ventilation system effect on post-shift lead was significant, however, as Table 4 shows, the use of a mask was not significant.

The urinary lead level of pottery glazers was higher than the tile glazers (154.3 µg/L versus 125.05 µg/L). ANCOVA showed that the effect of job on post-shift lead was not significant (Table 4). Glazers with work history more than or equal with 61 months had a higher urine lead level as compared to the work history of less than or equal with 24 months (154.5 µg/L versus 75.93 µg/L). A significant difference was found for the effect of work history on post-shift lead (Table 4).

The urinary lead levels were significantly higher in those who worked more than 20 h/month (206.25 µg/L versus 106.37 µg/L). ANCOVA showed that the effect of overtime work on post-shift lead was significant (Table 4). However, there was no significant trend in urinary lead levels according to the BMI and age parameters.

4. Discussion

Lead is a heavy toxic metal with wide universal industrial applications, particularly in Asian countries; therefore, occupational exposure to this toxic metal does occur lead is a heavy toxic metal with wide universal industrial applications, particularly in Asian countries.[20] In this study, 15 tile factories and six pottery workshops were evaluated. Because of differences present in the structure, ventilation systems, machines maintenance and cleaning operations in these plants and workplaces, different lead contents were observed in the urinary samples obtained. The lead concentration in glazers was 6.37 times higher than the control group. Results obtained from this study were similar to the other reported results. Saito et al. [21] reported that the highest blood lead levels were present among those who were working in the mixing section of lead materials. They also reported that blood lead levels were directly related to the amount of lead materials used for ceramic productions. Nagar and Rosa et al. [22,23] reported that, in the ceramics industry, glazers have the most exposure to lead compared to the other groups. Sanai et al. reported high values of lead in urine from workers where metallic lead was molten and used in glaze.[24] Shiri et al. reported that several cases of the occurrence of acute abdominal pain due to lead poisoning in pottery workshops.[20] This symptom was also observed among glazers in our study. Lead poisoning in a ceramic glazer has also been reported.[25] Therefore, all workers exposed to lead in the ceramic industries may show the same symptoms.

Similar to a study of Choy et al., [26] results obtained from this study showed a higher urinary lead level during mixing and handling of lead glazes without the use of personal protective equipment. This emphasises the role of using personal protective equipment in the reduction of lead exposure. Maizlish and Rudolph have reported high blood lead levels in workers not using a respirator.[27] In our study this variable is not significant, probably due to the small number of glazers who protected themselves by using a mask as a protective device.

Similar to our findings, Bache et al. reported that glazers whose workplaces have a ventilation system show significantly lower levels of lead in their hair samples.[28] Maizlish and Rudolph reported the highest blood lead level (10.63 μmol/L) in ceramic tile glaze-sprayer who was working in an unventilated workplace.[27] Therefore, lack of a ventilation system can significantly affect workplace lead concentration. The differences present in the urinary lead between tile and pottery glazers can be related to the differences between the environment and the lead content in the glaze at the two types of jobs.

Urinary lead level was also significantly increased with work history. Similar to our results, Fukaya et al. found a relationship between high blood lead level and the number of years of lead exposure in the ceramics industry.[29]. Subjects who were working overtime for more than 20 h during the past month had the higher urinary lead in comparison to the group without overtime working period, likely due to the more time exposure to lead-containing glazes.[30] In the present study, all spirometric parameters in glazers were significantly decreased as compared to their control group. The decline in FEV_1 and the ratio of FEV_1/FVC were higher in workers exposed to lead compared to those who were not exposed. Our results suggest an obstructive pattern of impairment of lung function among the glazers. Because of the high concentration of lead, these results would be expected, however, in the glazing unit in addition to exposure to lead, exposure to other raw materials, particularly silica, may occur, causing a reduction in lung function.

Halvani et al. and Bahrami et al. studied ceramic workers and discovered that exposure to silica is related to a reduction in pulmonary function and chronic obstructive pulmonary disease (COPD).[31,32] Therefore, in addition to lead, silica can cause similar symptoms. In another study in Iran, ceramic factory workers complained of respiratory disorders and abnormal pulmonary function.[33] Therefore, further studies are required to understand the mechanism of interaction between glaze constituents and silica in impaired lung functions.

It is worth mentioning that the health hazards of lead were not limited to the glazers who used lead in glaze structure. Improper formulation and firing practices of glazes may increase the hazards of lead migration from glazes into food substances and, therefore, the general population is also exposed to lead when they use these wares. In Mexico, the sixth largest lead-producing country in the world, a number of studies have confirmed that cooking and storage of foods in the lead-glazed pottery is a significant determinant for elevated blood lead concentrations.[4,34–36] There was a significant increasing trend in blood lead levels regarding frequency of consumption of food prepared in lead-glazed ceramic.[4] Rothenberg et al. and Farias et al. reported that pregnant women who used lead-glazed ceramics had higher blood lead levels compared to the pregnant women who did not use this type of ceramic.[37,38] Similar results have been observed in New York City, i.e., the use of imported glazed ceramic ware from Mexico was the strongest determinant of lead exposure.[39] Also, other cases of poisoning have been observed after using ceramic vessels.[40,41] Several studies on potters and their family members have demonstrated the high blood lead levels as a result of manufacturing and using lead glazes.[42–44] Children whose mothers use lead-glazed pottery dishes show high blood lead levels.[1,45] Sheets and Klein et al. observed that half of the samples of porcelain dinnerware and earthenware released lead into the food substances.[41,46] Therefore, based on such studies, the presence of lead in ceramic products may cause exposure of the public to this toxic metal.

Further studies need to be developed in order to know more about the adverse effects on workers who are working in the lead industries. Preventive strategies are required to guarantee complete human health.It can be concluded that enclosing the glazing unit, presence of local dust ventilation, use of protective masks or respirators, decrease of exposure time, drinking of milk in workplaces, replacement of lead by another substance and periodic re-deployment can reduce the exposure to lead down to the threshold limit value. Glazers should not eat in places of mixing or handling, or during the glaze firing.

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