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Research letter

Biological monitoring of occupational exposure to dust among aluminium foundry workers

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Abstract: *Aim* — The purpose of the study was to evaluate the biological monitoring of occupational exposure to aluminium (AL) dust in foundry workers in south of Tehran.
Material and Methods — This cross-sectional study was carried out on 63 workers in A and B foundries and 50 unexposed individuals as the control group. AL dust were sampled using Higgins-Dewell cyclone (HD) and cellulose ester membrane filter (MEC) with a flow of 2.2 l/min in the breathing zone of workers for 4 hours. Urinary samples were taken at the end of work shifts per week and were analyzed using graphite furnace atomic absorption. Data were analyzed using SPSS v.21 and statistical methods including t-test, one-way ANOVA and linear regression.
Results — Airborne concentration of AL aerosols differed statistically significant in occupational groups ($P<0.05$). There were no significant differences between urinary AL concentrations in different occupational groups ($P>0.05$). However, there was a significant difference between urinary concentrations of the exposed group and the control group ($P<0.05$) and there was no significant correlation between the AL concentration in workplace air and urinary concentration of AL in the exposed group ($P>0.05$).
Conclusion — Determination of AL concentration in urine is not enough to serve as a biomarker. Estimation of AL nanoparticles in the air and biomarkers that determine the actual absorption rate seems to be an adequate method for occupational exposure monitoring of AL.

Keywords: biological monitoring, air monitoring, foundry workers, occupational exposure, aluminium dust

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Introduction

Aluminum (AL) is one of the most abundant compounds in the earth crust [1]. Exposure to AL widely occurs due to its presence in the environment and its use in everyday products such as beverage cans, cooking utensils, cosmetics, antiperspirants, sunscreens and food additives [2]. The primary source of exposure to AL for most people is through food [3]. AL compounds may be added to processed foods such as flour, baking powder, food coloring and anticaking agents, whereas unprocessed foods (fresh fruits, vegetables, meats) typically only contain trace amounts. On average, an adult consumes 7-9 mg of AL per day through ingestion of food [3].

Due to the widespread presence of AL in the environment, the toxicity of AL has been subjected to many studies [4]. In some studies, AL is considered as a non-toxic metal, but long-term dialysis as well as chronic exposure to dust, fumes and oxides of AL was reported to be responsible for pathological changes in certain situations [1, 4]. Some other studies have shown that there is a relationship between high levels of AL absorption in the body and increased risk of neuro-degeneration disorders such as encephalopathy, Alzheimer's disease and Parkinson's disease [5]. AL can accumulate in various body organs such as brain, bone, liver and kidneys and lead to kidney failure in the long term [1, 6]. In recent toxicology studies, the respiratory system is considered as the main route of exposure to AL compounds in industries, but the mechanism of the AL movement obtained from this route is not clearly determined and many ambiguities are proposed considering the direct absorption and metabolism of the AL from upper and lower extremity of the lung epithelium layer [7, 8].

Considering that AL bioavailability depends on its particle size and specific features, AL is not uniformly distributed in all tissues after absorption and is excreted primarily through the kidneys. AL half-life is very different in the urinary of exposed individuals depending on AL type and duration of exposure in a range of days, months and years. Since AL stored in the body is slowly excreted, so it can exist in the urinary many years after cessation of occupational exposure [9]. AL in human serum and urinary is an indicator of AL absorption rate and it is recommended to determine the urinary concentration of the AL to assess the occupational exposure of exposed workers [4].

Biological monitoring (BM) is an occupational exposure assessment method, by which hazardous substances or metabolites are determined through the analysis of blood, urine, hair or breath. Biological monitoring reflects the general absorption of a chemical substance through an absorption pathway (respiratory, digestive, skin) or a combination of these pathways; so they show the true

exposure level and biological monitoring helps identify hazardous substances in the body before causing any adverse health impacts [9, 10]. The results of biological monitoring can provide better risks values compared with the environmental monitoring (EM) and are considered as a complementary method. This evaluation method is used as an integral part of occupational health and safety strategy for the control of hazardous substances in the workplace, especially when the exposure occurred through different paths or occurs unusually [11].

Many methods have been proposed for determining the AL content. Graphite furnace atomic absorption (GFAA) is a method often used to measure the AL level in biological samples such as urinary and has shown excellent results. In this method, fluid samples can be directly inserted into the device and solid samples can be directly measured using new nuclear absorption devices [12]. Blood and urinary samples are accepted indicators, which can be widely used for occupational exposure. Blood has an undeniable advantage, i.e. it is in balance with the organs and tissues and thus is widely used for different research purposes; however, it is an invasive method and has practical and ethical limitations, particularly for sensitive populations [6, 10]. For practical reasons include available and non-invasive sampling, determination of urinary AL concentrations seems to be useful for risk assessment related to occupational exposure to Al dust than measurements of blood samples. Therefore, biological monitoring hasn't been done in Iran for the aim of evaluation of occupational exposure to AL so far. Assessment of occupational exposure to Al dust has been conducted merely on the basis of measurements of their concentrations in the air workplace and comparison with the values of occupational exposure limits (OEL) in Iran.

The purpose of this study has been to investigate of occupational exposure to AL among the foundry workers by using environmental and biological monitoring.

Material and Methods

Subjects

This cross-sectional study was carried out on workers who were exposed to AL aerosols in A and B foundries in south of Tehran (Iran). 63 healthy male workers at two AL foundry plants (A and B) participated in this study. This study was also performed on 50 non-exposed workers as the control group who did not have any history of occupational exposure to AL. Exposed and control groups were similar in demographics (gender, age) and socioeconomic status. Census sampling method was used. Based on inclusion criteria of this study, smokers, workers less than a year as work history and those using therapeutic drugs, were excluded.

Industrial processes

In the process of foundries, AL ingots are melted in open-hearth furnaces, then the molten metal is manually poured by the operator into the press molding (the shape of the desired piece) and cooled to solidify. Finally, the formed piece of metal isolated from a mold, sanded, then packaged and brought to the market. The widespread use of AL in various industries due to its special characteristics, such as lightweight and resistant to corrosion has increased the Al smelting operation. This process of work exposes the foundry workers to various types of hazardous harmful agents [13, 14]. In each of these foundries, three occupational tasks were defined as casting worker, assembly line worker and polishing worker. Since these groups have the same exposure conditions, they classified as similar exposure groups (SEG).

Measurements of study variables

A questionnaire was designed and data about the demographic characteristics of workers included age, height, weight, work history, worker's activity, history of disease or history of the specific drug were collected. According to the inclusion criteria, the purpose of the research was completely explained to subjects and if they wished to continue to cooperate, all participants complete and signed an informed consent form before the commencement of the study.

This study was approved by the ethical committee of the Shahid Beheshti University of Medical Sciences (Tehran, Iran) under the principles of the Declaration of Helsinki.

In order to measure the exposure of workers with AL dust, the respirable personal sampling in the breathing zone of workers was measured in different units. The concentration of dust was measured using NIOSH 7013 as a specific method for AL and its compounds. Air sampling was carried out in the worker's breathing zone for 4 hours using Higgins-Dewell cyclone (HD), Mixed Cellulose Ester filter (MEC) with a pore size of 0.8 and sampling pump with a flow of 2.2 L/min [15]. Graphite furnace atomic absorption spectrometry (Aurora Model) was used for the analysis of samples.

Biological monitoring

To determine urinary AL, urinary samples were collected by using clean plastic bottles. The plastic bottles were first rinsed with deionized water and were soaked in 10% nitric acid for 24 hours. To reduce the possibility of contamination of these plastic bottles, the displacement was carried out using gloves. At the end of work shift and after changing their uniforms, the workers were asked to wash their hands with liquid hand wash. Latex gloves were put at the disposal of workers and about 50ml of urinary was taken from the all exposed and control groups after giving necessary explanations to them. Urinary samples were collected at the end of the workweek.

Preparation of standard solutions for urinary samples

To prepare standards, AL sulfate-18-hydrate and worker's urinary were used. Solutions with a volume of 10 ml, concentrations of 10, 20, 30, 40, 50, 60 and 70 ppm were prepared. Standards were dried on the hot plate at 70°C. 6ml of 65% concentrated nitric acid, 6 mL of concentrated hydrochloric acid for digestion proposes and 6mL of oxygenated water for transparency proposes were added to each standard and was dried in three stages. 0.2 ml (200 µl) of potassium solution with a concentration of 50,000 ppm was added to each solution. Solutions were later reached to the intended volume using 10% nitric acid in the 10 ml volumetric flask. The solution was later transferred to falcon tubes and was centrifuged for 10-15 minutes at a speed of 6,000 rpm. Finally, to prepare a calibration curve and to carry out the atomic furnace analysis, 20 µl of each solution was injected into the flask. To prepare urinary samples, 10ml of each sample was prepared using the above-mentioned methods and graphite furnace atomic absorption spectrometry (Aurora model, made in Canada) was used for the analysis of samples with high sensitivity. The device was set at 309.3 nm in wavelength, deuterium lamp intensity of 4 milliamperes (mA) and voltage of

725 v and the temperature program used to analyze biological samples is presented in *Table* 1 [16].

Table 1. Temperature program used to analyze biological samples

Phase	t, °C	Ramp, °C/s	Hold time, s	GF, L/min
Drying	85	5	0	2
Incineration 1	110	15	10	2
Incineration 2	500	5	5	2
Incineration 3	1500	15	5	2
Atomic absorption	2700	0	2	2

t, temperature; GF, gas flow.

Table 2. Demographic data of studied subjects

Variables	*Exposed group (n=63)*	*Control group (n=50)*	*P-value*
Age, years	36.2±7.5	35.7±7.3	0.685
Work experience, years	12.3±8.2	12.2±7.2	0.724
BMI, kg/m^2	24.2±3.9	25.3±4.3	0.321

Data presented as mean with standard deviation – M±SD.
BMI, body mass index.

Table 3. Airborne and urinary concentrations of AL in occupational groups in Foundries

Foundry	Work unit	n	Air conc. of AL, mg/m^3	Ur. conc. of AL, µg/L
A	Casting w-r	13	3.77±2.79	43.4±18.2
	Assembly line w-r	11	1.94±1.71	45.6±15.9
	Polishing w-r	9	3.93±1.83	49.6±13.9
B	Casting w-r	12	4.18±2.40	47.3±22.3
	Assembly line w-r	10	1.98±1.58	40.7±17.0
	Polishing w-r	8	3.73±1.85	54.5±13.8

Data presented as mean with standard deviation – M±SD.
n, sample number; conc., concentration; Ur., urinary; w-r, worker.

Table 4. Comparison of air and urinary concentrations of AL in the corresponding occupational groups

Variables	*W.u.*	*Cast.*	*P*	*Ass. l.*	*P*	*Pol.*	*P*
A.c.AL, mg/m^3	A	3.77	0.726	1.94	0.957	3.93	0.827
	B	4.18		1.98		3.73	
Ur.c.AL, µg/L	A	18.2	0.85	45.6	0.674	49.6	0.562
	B	22.3		40.7		54.5	

W.u., work unit; Cast., Casting; P, P-value; Ass. l., Assembly line; Pol., Polishing; A.c.AL, air concentration of AL; Ur.c.AL, urinary concentration of AL.

Table 5. Comparison of AL airborne and urinary concentrations in occupational groups

Variables	*Casting (n=25)*	*Assembly line (n=21)*	*Polishing (n=17)*	*P-value*
A.c.AL, mg/m^3	3.95	1.96	3.83	0.004
Ur.c.AL, µg/L	45.28	43.24	51.9	0.287

A.c.AL, air concentration of AL; Ur.c.AL, urinary concentration of AL.

Table 6. Comparison of urinary concentrations between exposed and unexposed groups

Variable	*Exposed group (n=63)*	*Unexposed group (n=50)*	*P-value*
Ur.c.AL, µg/L	46.38±17.28	7.40±2.18	0.001

Data presented as mean with standard deviation – M±SD.
Ur.c.AL, urinary concentration of AL.

Statistical analysis

Statistical analysis was performed by using SPSS software (version 21, SPSS Inc., Chicago, IL, USA). The Kolmogorov-Smirnov test was used to investigate the normality of data distribution. As the variables including age, work experience, body mass index (BMI), air AL concentration and urinary AL concentration were normal, the differences between demographic variables of subjects and comparison differences between the groups were analyzed independent t-test. The analysis of variance (ANOVA) was used to study the air and urine concentrations of AL in various occupational groups of each foundry. Also, the relationship between air and urinary AL concentrations in the exposed workers were analyzed by linear regression test. The level of statistical significance was considered at P<0.05.

Results

The demographic data of both exposed workers to AL aerosols and unexposed group are provided in *Table* 2. The mean age of foundry workers in the A and B plants were 36.1±7.5 and 36.3±7.1 years, respectively. The mean work experience of exposed workers in A and B foundries were respectively determined as follows: 11.9±8.4, 12.7±8.1 years. The statistical test shows no significant differences between the demographic variables of the two foundry group and control group regard to age, work experience and BMI (P>0.05).

Average respirable AL dust in Foundries A and B was respectively calculated 3.21±2.33 and 3.31±2.15 mg/m³ that shown in *Table* 3. The statistical analysis showed no statistically significant difference between their exposures (P=0.853).

The mean urinary AL concentrations of exposed workers in the A and B Foundries were 45.8±17.02 and 47.02±18.26 µg/L respectively. The mean with standard deviation of AL airborne and urinary concentrations of both foundries are separately provided in *Table* 3. The statistical analysis showed that there was no statistically significant difference between the urinary AL concentration workers in the two Foundries of A and B (P=0.784).

Air and urinary concentrations of AL in the corresponding occupational groups in both A and B work units are separately presented in *Table* 4. The results of the statistical analysis showed that there was no significant statistical difference between the corresponding occupational groups in terms of the air and urinary concentrations of AL.

The relationship between demographic variables and the level of exposure was investigated using Pearson correlation, which showed no significant relationship between the concentration of AL in the urinary of exposed individuals and their age, work experience and BMI (P>0.05).

Both air and urinary concentrations of AL in different occupational groups were investigated (*Table* 5). There was a statistically significant difference between air AL concentrations in occupational groups. The differences were statistically significant between casting and polishing groups with assembly line working group (P<0.05). In addition, there was no statistically significant difference between different occupational groups in terms of urinary AL concentrations.

The AL urinary concentration in the exposed and unexposed groups is presented in *Table* 6. The total mean urinary concentration of AL in the exposed groups was 6 times higher than the urinary concentration of AL of the control group (P<0.05).

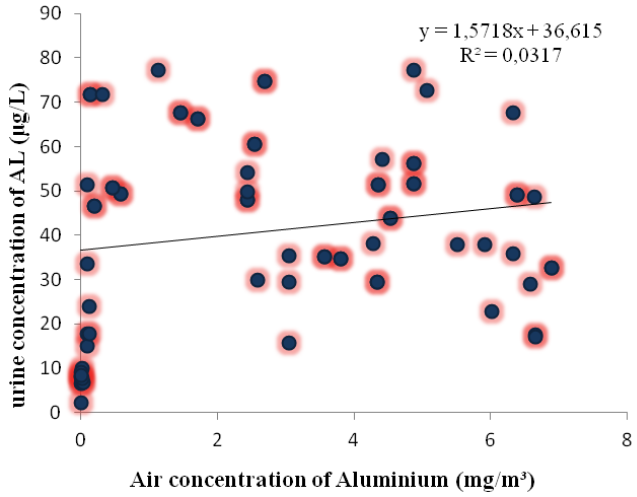

Figure 1. Data scatter plot between the urinary and air concentrations of AL

Correlation of AL airborne and urinary concentrations were investigated in the exposed group using the linear regression test, which showed that there was no statistically significant relationship (P=0.130). *Figure* 1 shows the scatter plot between the concentrations of AL in the air and urine.

Discussion

Biological monitoring is an important tool in the occupational health to evaluation workplace pollutants. By using the biological monitoring determined occupational exposure limits to various contaminants which can lead to reduce exposure and prevent adverse health effects. In this study, three occupational groups with the similar exposure conditions were evaluated in two different work units. Based on the results of air monitoring, exposure rate to respirable aerosols of AL was determined in the range of 0.10 to 6.89 mg/m³ and an average of 3.26 mg/m³, which is higher than the occupational exposure standard of the American Conference of Governmental Industrial Hygienists (ACGIH) [17] and the occupational exposure limits (OEL) recommended by the Technical Committee of Health Professionals of Iran. Previous studies suggested that AL concentrations in urine or plasma can be appropriate for assessment of the chronic occupational exposure of subjects with AL [18, 19]. On the other hand, excretion of urinary AL has been shown more sensitive to the plasma AL concentration [9]. In this study, urinary excretion of AL was measured in workers who occupationally exposed to dust and non-exposed subjects. Based on several previous studies, the concentration of urinary AL in the general population with no history of occupational exposure to the AL is less than 15 µg/L [9]. In this study, the urinary concentration of AL in the control group was less than the prescribed limit and in workers exposed to AL dust was higher than normal limit of urinary AL (≥25 µg/l) [19]. In our study, the reasons for the high exposure are working in units that are not enclosed and without any suitable protective equipment.

Workers in corresponding occupational groups experienced the similar exposure conditions (*Table* 4). There was no significant relationship between the urinary concentration of AL with age, work experience and the BMI of exposed workers. In a study, Ogawa and Kayama showed that there no significant relationship between urinary concentrations of AL, with age and work experience, which is consistent with the result obtained in the present study [6]. In a previous study, those who had less than a year of exposure to AL, elimination half-life of AL in urine was within a few days, but subjects with work experience of over 10 years have can have the half-life up to several months [20]. Therefore, the longer working experience could prolong the elimination time of AL from the body. Because AL has a strong tendency to accumulate in bone tissues and can be maintained for a long time in human bones. Moreover, AL could partly accumulate in the lung alveoli, especially while inhalation is the main route of exposure to AL [21,22].

Statistical analysis of the present study shows, there was a significant statistical difference between the groups in terms of occupational exposure to respirable aerosols, and the average concentration of AL measured in the Casting and the Polishing groups is 3 times more than of the Assembly line group. However, there was no difference between the AL urinary concentrations of these exposed groups. In this study, all the subjects were exposed to AL airborne aerosol higher than occupational exposure limits. While subjects are exposed to excessive amounts of AL, it seems that the body absorption of AL is higher than its ability to eliminate it. Therefore, subjects will show the same urinary excretion and in the long-term occupational exposure also will be prolonged the half-life of the elimination of AL in the body. On the other hand given that occupational exposure to AL, breathing is the main path for AL absorption, and biological absorption of AL is dependent on the type and size of inhaled particles [9]. We cannot ignore the impact of nanoparticles on the absorbed AL.

The level of urinary AL concentrations in the present study shows that the AL concentrations in the exposed group were statistically significantly higher than the control group (about 6 times more), which indicates the exposure to airborne dust and fumes of AL can significantly increase the concentration of AL in urine. Similarly, Mussi et al. showed that occupational exposure to AL fumes and dust led to a significant increase in urinary concentrations of AL in the exposed group [23]. Rollin et al. showed that exposure to Al airborne concentrations statistically increased the urinary of exposed workers to AL dust compared to controls [24]. Liao et al. also demonstrated that urinary concentrations of 103 exposed workers increased, but the correlation of their external with internal exposures was not statistically significant [25]. Sinczuk also showed that the AL exposure level was estimated to be 1.5 mg/m³ in foundries and urinary concentration of AL in the exposed group was significantly higher than the control group [4].

In a longitudinal study carried out by Rossbach et al. showed that welders had approximately same constant exposure to AL dust, a close relationship between internal exposure and total dust concentration of AL was not found, and it was suggested that the respirable concentration of AL should be measured [22]. Sjögren study showed that there is a positive relationship between the urinary concentrations of AL after the work shift, air concentration of AL and exposure time [26]. But Sinczuk observed no significant relationship between the concentration of AL oxide in workplace air and urinary concentrations of AL [4]. In this study, the concentration of respirable AL air was measured, but there was no significant relationship between the AL air concentration and urinary concentration of AL in the exposed groups. Based on these findings, we can not specify that AL absorbed in the body is only

affected by airborne breathable dust because airborne nanoparticles seem to have a large share of particles absorbed in the body. Only breathable particles can be collected and analyzed by cyclone HD and sampling can't be carried out on nanoparticles; so, airborne AL measurement cannot reveal the true exposure rate of the staff. In the study of Hull & Abraham on subjects exposed to welding fumes, the results showed that most of the particles were accumulated and the average diameter of the AL particles was 0.34 µm and there were also the singlet particles as small as 10 nm in diameter [27]. The toxicokinetic of AL in human by inhalation, which is the main route of exposure in occupational workplaces, has not received much attention [28]. It may be difficult to estimate the actual body burden due to the reabsorption of inhaled AL in the lung [9]. Even though the role of nutritional factors in the detoxification of AL cannot be ignored [10, 29].

According to the results of the present study, it is recommended biological monitoring can be assessed as a complementary method to investigate the actual amount of exposure rate by the workers. In order to correct the effects of diuretic dilution, urinary concentration of AL should be investigated according to the creatinine.

Conclusion

The results of this study show that airborne concentrations of AL were statistically significantly different in three occupational groups. However, there were no statistically significant differences between urinary concentrations of AL in different occupational groups. Therefore, occupational monitoring of workers sampling respirable Al dust (with a diameter of less than 10 microns) in the work environment may not be a good indicator to determine the exposure rate. Although determining urinary concentrations of AL can be considered a practical indicator of AL absorption in the body, this method is time-consuming and expensive and with a high probability of error. It is recommended to carry out studies on AL nanoparticles in the air as well as investigating other biomarkers for determining the actual absorption level of airborne Al.

Limitations

The limitations of this study include the relatively small number of samples, lack of selection of the control group from the same industrial site and the further investigation of long term health effects of the workers.

Conflicts of interest

The authors declare that there are no conflicts of interests regarding the publication of this article.

Ethical approval

All procedures performed in studies involving human participants were in accordance with the ethical committee of the Shahid Beheshti University of Medical Sciences (Tehran, Iran), No. 157373, complied with the principles of the declaration of Helsinki (1964) and its later amendments.

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