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Blood Lead Levels in Copper Smelter Workers in Japan

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Abstract: Lead exposure of workers in a Japanese copper smelter was assessed by determining lead levels in blood, air and flue cinder at the copper smelting processes. All the samples were analyzed for lead by atomic absorption spectrometry. Mean lead levels of air were highest at the anode department followed by the converter, smelter and blend departments. The mean level of blood lead of the workers in the anode department was also the highest among the four smelting departments. The mean blood lead levels of the workers in each department were positively correlated with their air lead levels (r=0.99, p<0.01). This study indicates therefore that workers in copper smelters have been exposed to lead in their workplace. Though this finding has already been reported in preceding studies, the Ordinance on Prevention of Lead Poisoning in Japan has not included copper smelter into its target job categories if their lead concentration in the raw material is less than 3%. The limitation of the present Ordinance which defines the targets by the types of job and not by the actual exposure, is discussed.

Key words: Blood lead level, Air lead level, Flue cinder, Copper smelter, Biological monitoring

Introduction

There have been a number of studies that have reported increased blood lead levels in copper smelter workers[1–9]. Lilis *et al.*[1] reported that mean blood lead levels in copper smelter workers was 31.1 μg/dl. Gerhardsson *et al.*[4] reported that the mean blood lead concentration for northern Swedish copper smelter workers was 58.2 μg/dl in 1950 compared with 33.6 μg/dl in 1974. Apparently workers in copper smelters are exposed to higher level of lead. Gagne *et al.*[8] reported elevated lead levels in the soil of a residential district near a copper smelter in Canada, and Andrzejak *et al.*[9] reported elevated air lead concentrations of a village near a copper smelter in Poland. However there has been no reported study concerning lead exposure in copper smelter workers in Japan.

OSHA (Occupational Safety and Health Administration) in the USA regulates the measurement of lead levels in air in primary copper smelters[10]. Its 8-hour time-weighted-average permissible exposure limit (TWA PEL) of air lead levels has been 50 μg/m^3 since 1978[11]. The Japan Society for Occupational Health changed the occupational exposure limit of air lead level from 150 μg/m^3 in 1961 to 100 μg/m^3 in 1982[12]. However, the present Ordinance on Prevention of Lead Poisoning in Japan applies not to the air lead level but to the work type listed in the Ordinance. For a work handling molten ore in the process of smelting, the Ordinance is not applied if the raw material contains less than 3% lead[13].

The primary objective of this study is to examine whether the current Japanese regulation, which is based not on the actual exposure but by job type, is effective in controlling the lead exposure of workers in copper smelters in Japan.

Materials and Methods

Processes of copper smelting

The principal processes in copper production include mining, concentrating, and smelting. The initial operation in the smelting process is blending, where the copper ore is blended and the ore concentrate is stored. Subsequently,

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the ore concentrate is fed to the smelter process. This features a reverberatory furnace, which contains recycled precipitates, converter slag, flue dust, limestone, and silica flux. Matte, a mixture of cuprous and ferric sulfide, is formed along with a slag of various metal silicates. The matte contains 35% copper and is charged by ladle to the converter process, where blister copper is produced with siliceous flux. Air is blown into the hot metal through tuyeres, and metal silicate slag is formed on top of the cuprous sulfide. The slag is recycled periodically to the smelting operation to recover the remaining copper. The blister copper formed in the converter is purified to approximately 98.5%. This intermediate is further refined in a gas-fired furnace and the copper casted by pouring into molds. Copper is further refined in the anode process, where anode is casted by a gas-fired furnace followed by molds of anode. This product is refined electrolytically in a copper sulfate-sulfuric acid electrolyte bath with the blister copper as the anode and a thin plate of pure copper as the cathode. The anode copper is deposited as 99.5% copper on the cathode.

Subjects

One hundred and sixty-six male workers at a copper smelter in Japan were requested to participate in the study. The subjects age ranged from 21 to 60 years (mean 42.9).

Five groups of workers stationed at five departments of distinct operational areas were selected for this study. The departments were office (control), blend, smelter, converter and anode, with the workers numbering 43, 13, 51, 28 and 31, respectively. The latter four departments comprised smelting processes, and there a three-shift system of 8 h is employed.

Sample collection

Blood Sampling: Venous blood was obtained from workers of the four copper smelting departments and the office. Blood samples were drawn from the arm of each worker, using a sterile plastic syringe containing heparin as anticoagulant. Samples were stored in a refrigerator until analyses.

Air and flue cinder particulate: Particulate matter was collected onto a 44-mm diameter membrane filter of 7.07 μm pore size. The filters were mounted in a filter holder which was attached to an air sampler. The air sampler with a filter was held 1.0–1.5 m above the floor and air was drawn at 15 l/min for 10 min in the four smelting departments.

The air lead levels in each department were demonstrated using the geometric mean of the measurements at five points ($\mu g/m^3$ lead of the sampled air), and those in flue cinder was indicated by the arithmetic mean (% of the sampled flue cinders) from duplicated measurements. There was no flue installed in the anode department. The relationships between lead levels in blood and air or in flue cinder in each department were analyzed.

Sample analysis

Blood samples were prepared as described previously[14]. The analysis of lead levels in blood was performed by flame atomic absorption spectrometry (Hitachi 6100). The detection limit was 1.0 μg/dl blood.

Filters were dissolved in nitric acid by heating to total destruction, and the lead levels in the diluted solutions were measured. Unexposed membrane filter papers were similarly treated and analyzed as blanks. All the measurements were checked with standard solutions.

Questionnaire

Workers of the anode department were asked to complete a questionnaire, regarding age, job history, working hours, smoking habits, frequencies of hand- and face-washing before meals during on-duty hours, taking off protective gloves, eating between meals during working hours, and smoking at work. The responses to the behavioral questions asking for frequencies were classified into 2 categories each, except for smoking (3 categories).

Statistical analysis

Statistical analysis was performed using the PC-SAS. The Wilcoxon test was used to analyze the differences between two groups and multiple group means were compared by the ANOVA. A level of $p<0.05$ was regarded as statistically significant.

Results

In Table 1, the mean values of lead levels in air and flue cinder in each copper smelting department are shown. Mean lead levels in both air and flue cinder increased in the ascending order of the four copper smelting departments of blend, smelter, converter and anode. The geometric mean lead levels in air in the anode department was 313 $\mu g/m^3$, which was the highest among the four copper smelting departments. The percentage of lead contained in flue cinder in the converter department was the highest (26.2%) among the three (blend, smelter and converter).

The total arithmetic mean of blood lead levels was 13.7

Table 1. Geometric mean of lead concentrations in air and average percentage of lead in flue cinders of the copper smelting processes

Jobs Department	Air Pb; $\mu g/m^3$ (range)	Flue cinder Pb; % (range)
Blend	7 (5–8)	0.3 (0.32–0.36)
Smelter	29 (6–67)	6.5 (6.23–6.67)
Converter	41 (17–78)	26.2 (25.6–26.9)
Anode	313 (165–436)	—

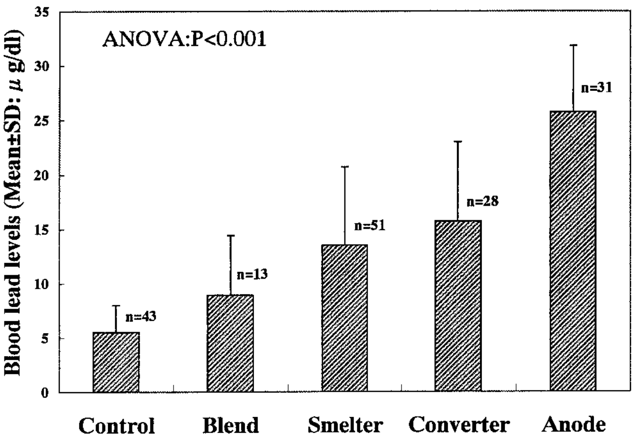

Fig. 1. Blood lead levels of workers in the four copper smelting departments and the control group (Mean ± SD).
Mean blood lead level of workers in the anode department (25.7 ± 6.1 $\mu g/dl$) was significantly higher than that in any other three copper smelter departments and control (office) workers (Wilcoxon test: $p<0.001$).

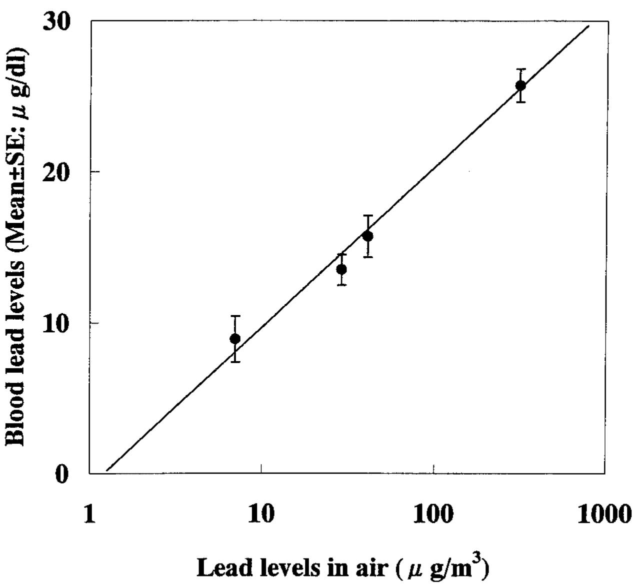

Fig. 2. Relationship between lead levels in blood and air.
($y= - 0.64 + 10.34 \log x$, $r=0.99$, $p<0.01$)

Table 2. Factors contributing to blood lead levels (Anode employees)

		N	Mean ± SD of blood Pb levels; $\mu g/dl$	(range)
Smoking	Smokers	21	26.3 ± 6.2	(14–39)
	Ex-smokers	3	21.0 ± 1.6	(19–23)
	Nonsmokers	7	25.9 ± 6.2	(15–34)
Face-wash (before meals)	(+)	27	26.1 ± 6.3	(14–39)
	(–)	4	22.5 ± 3.0	(19–27)
Snack (during work hours)	(+)	28	26.6 ± 5.7*	(14–39)
	(–)	3	17.3 ± 2.6	(15–21)
Glove (taking off)	≦9/day	9	26.0 ± 6.7	(16–39)
	≧10/day	22	25.5 ± 5.8	(14–36)

Face-wash (+): include 'sometimes', Snack (+): more than once/day. *; $p<0.05$, Wilcoxon-test.

$\mu g/dl$ (Fig. 1). The mean ± SD of blood lead levels in each group were 5.5 ± 2.5 $\mu g/dl$ for control (office), 8.9 ± 5.5 for blend, 13.5 ± 7.2 for smelter, 15.7 ± 7.3 for converter, and 25.7 ± 6.1 for anode. The mean blood lead levels in the anode department was significantly higher than any other departments (Wilcoxon test: $p<0.001$).

Fig. 2 presents the relationship between lead levels in blood and those in air. The mean blood lead levels were positively correlated with the log-transformed lead levels in air ($r=0.99$, $p<0.01$).

As shown in Table 2, the mean blood lead levels in 28 anode workers who eat snacks between meals during working hours (26.6 $\mu g/dl$) was significantly higher than that of the 3 non-eating workers (17.3 $\mu g/dl$). None of the other categories obtained by the questionnaire, including length of service, smoking, face washing and wearing gloves, showed a significant correlation with the blood lead levels.

Discussion

The blood lead level is commonly used as an indicator of the lead exposure in humans[15, 16]. Among the subjects of this study, the mean blood lead level in the anode department was the highest, in addition, the air lead level in the same department was also the highest of the four copper smelting departments. We found a good correlation between log-transformed lead levels in air and those of blood as shown in Fig. 2. As for the correlation between air and blood lead levels, preceding reports have shown conflicting results and for the copper smelters in particular, no detailed analysis for such a correlation has been performed[1–9]. Never the less the results of the present study suggest that workers in the smelting departments have higher blood lead levels than the office workers. Due to lead fumes and flues cinder, copper smelter workers are continuously exposed to lead in their working environment. Lilis *et al.*[3] demonstrated that median blood lead levels of Canadian copper smelter workers was 30.4 μg/dl, and in 16.7% of cases, it was 40 μg/dl or higher. In our study, the blood lead level was not as high as indicated in their report, but it would seem that insufficient attention has been directed to the lead exposure of copper smelter workers in Japan.

Recently there has been increased concern about low level exposure to lead. Lilly *et al.*[1] reported effects of low lead exposure on renal function among Canadian copper smelter workers. Gerhardsson *et al.*[4] reported that a retrospective cohort study of lead-exposed Swedish copper smelter workers showed considerable excess of deaths for total mortality, malignant neoplasms especially lung and stomach cancer, ischemic heart disease, and cerebrovascular diseases when compared with the general population. The effect of low level lead exposure on health has probably been underestimated. At lower levels of lead exposure, there can be nonspecific early symptoms that mimic many other types of illness. For example, disorders in neurological and systemic functions have been identified at low levels of lead exposure, i.e., 20 μg/dl of lead level in blood[18–20]. WHO/IPCS reported that clinical lesions are caused when lead levels in blood become 20 μg/dl or higher[21]. Recently, the recommendation value for biological exposure index for lead by ACGIH has been reduced to 30 μg/dl in blood[22]. Accordingly, biological monitoring of lead exposure in the copper smelter workers may be indispensable, although it is not presently required by the regulations in Japan.

In spite of the preceding reports on the elevated blood lead levels in copper smelters[1–9], the Ordinance on Prevention of Lead Poisoning in Japan does not include copper smelting into its target job categories if the lead concentration in the raw materials is less than 3%. In general, copper ores contain 0.003 to 1.3% of lead[17]. In this study, the lead composition in the raw materials of the smelter was 0.3%, well below the level set by the Ordinance to be regulated as a lead-handling job. However, the melting point of lead is low (327.5°C) and fumes are easily volatilized. The lead levels in air and flue cinder in the copper smelting processes can thus increase and as a consequence, blood lead levels of workers may be elevated.

Under the Ordinance on Prevention of Lead Poisoning in Japan, whatever the level the actual exposure is, a work handling molten ore in the process of smelting is not regulated so long as the raw materials contain lead less than 3%[13]. The results of the present study suggest that workers with high lead exposure were not regulated by the Ordinance, hence, it is not effective in controlling the lead exposure of workers in copper smelters in Japan. Thus, to include smelting jobs into the target job list of the Ordinance regardless of the lead composition of the ore, may be the only solution. However, it may not be practical to keep up with the rapid changes of the job type in accordance with the rapidly developing technology and the changing manufacturing process. We suggest that the Japanese Ordinance should change from the job list system to an actual exposure system in defining the target job. By doing so, the employers appreciate the harmful exposure to workers, and are placed under an obligation to change the system to remedy the consequences of the harmful effects. If environmental management is practiced efficiently and sufficiently, the employers would not face a legal requirement for blood lead examination. The essential issue for the managers becomes the strong motivation for environmental control.

In the mean time, prior to the reform of the legal system, there are number of measures needed to be performed to reduce the exposure of lead to the smelter workers. For the workplace, isolation of high lead area, installation of local and/or general local ventilators, and improvement of existing facilities and work process or procedures are essential. For the workers, effective protective devices such as respirators and cloths should be provided. In addition, education of workers to improve their hygienic behavior may be needed because, our result suggests that eating between meals during working hours increased the blood lead levels, although the number of comparisons was small. The main route of lead absorption may be through inhalation, however, inadequate hand- and face-washing and unrestricted smoking and eating at the workplace may result in subclinical lead poisoning[14]. Finally, employers have to measure the level of lead regularly in the workplace in order to evaluate the effectiveness of the measures and control the working environment.

In conclusion, our study has demonstrated elevated blood lead levels in Japanese copper smelter workers for whom the Ordinance on Prevention of Lead Poisoning in Japan does not apply. This study indicates that the Ordinance does not meet the actual need, and hence suggests a necessity to amend it from the job list system to the one based on the actual exposure.

References

1) Lilis R, Valsiukas JA, Weber JP, Malkin J, Selikoff IJ (1984) Epidemiologic study of renal function in copper smelter workers. Environ Health Perspect **54**, 181–92.
2) Lilis R, Valciukas JA, Weber JP, Fischbein A, Nicholson WJ, Campbell C, Malkin L, Selikoff IJ (1984) Distribution of blood lead, blood cadmium, urinary cadmium, and urinary arsenic levels in employees of a copper smelter. Environ Res **33**,76–95.
3) Lilis R, Valciukas JA, Weber JP, Malkin J (1985) Effects of low-level lead and arsenic exposure on copper smelter workers. Arch Environ Health **40**, 38–47.
4) Gerhardsson L, Lundstrom NG, Nordberg G, Wall S (1986) Mortality and lead exposure: a retrospective cohort study of Swedish smelter workers. Br J Ind Med **43**, 707–12.
5) Valciukas JA, Lilis R (1982) A composite index of lead effects. Comparative study of four occupational groups with different levels of lead absorption. Int Arch Occup Envion Health **51**, 1–14.
6) Andrzejak R, Antonowics J, Bolanowska B, Hebdzinski L, Kabacinska-Knapik D, Smolic R (1996) Thyroid function in smelters after long-term exposure to heavy metals. Med Pr **47**, 253–8.
7) Glickman L, Valciukas JA, Lilis R, Weisman I (1985) Saccadic eye movements among copper smelter workers. Am J Ind Med **7**, 141–53.
8) Gagne D, Letourneau G (1993) Determining soil contamination patterns in a residential district near a copper smelter. Arch Environ Health **48**, 181–3.
9) Andrzejak R, Antonowicz J, Tomczyk J, Lepetow T, Smolik R (1993) Lead and cadmium concentrations in blood of people living near a copper smelter in Legnica, Poland. Sci Total Environ **Suppl Pt 1**, 233–6.
10) OSHA (1991) 29 CFR Section 1910, 1025-Lead, Appendix A and B. Washington DC, OSHA.
11) U.S. Federal Register (1978) Occupational exposure to lead: Final Standard Washington DC, Part IV 52952–53014.
12) Japan Association of Industrial Health (1982) Recommended occupational exposure limits of lead in the air. Jpn J Ind Health **24**, 542–4.
13) Japan Industrial Safety and Health Association (1991) Industrial safety and health law and related legislation of Japan. Daiichi-Houki, English pp. 502.
14) Karita K, Shinozaki T, Yano E (1997) Possible oral lead intake via contaminated facial skin. Sci Total Environ **199**, 125–31.
15) Tang N, Nakatsuka H, Watanabe T, Liu SJ, Qu QS, Liu YT, Cai SX, Jin C, Ikede M (1990) Blood lead levels of Beijing (China) citizens. Sci Total Environ **90**, 31–9.
16) Berlin A (1989) Lead control in the working environment. In: Encyclopedia of occupational health and safety, 2 (L-Z), 1206–9, ILO, Geneva.
17) Nelson KW, Varner M, Smith TJ (1977) “Nonferrous metallurgical operations.” In: Air pollution, ed. by Stern A, 3rd ed., Vol. 4, Academic, New York.
18) Yokoyama K, Araki S, Aono S (1988) Reversibility of psychological performance in subclinical lead absorption. Neurotoxicology **9**, 405–10.
19) Hirata M, Kosaka H (1993) Effect of lead exposure on neurophysiological parameters. Environ Res **63**, 60–9.
20) Murata K, Araki S, Yokoyama K, Uchida E, Fujimura Y (1993) Assessment of central, peripheral, and autonomic nervous system functions in lead workers: neuroelectrophysiological studies. Environ Res **61**, 323–36.
21) IPCS (1995) Inorganic lead. World Health Organization, Geneva.
22) ACGIH (1999) TLVs and BEIs; Threshold limit values for chemical substances and physical agents. Biological exposure indices. Cincinnati: ACGIH.