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HEALTH EXAMINATION OF ALUMINUM REFINERY WORKERS EXPOSED TO FLUORIDE. SERUM AND URINARY LEVEL OF FLUORIDE AND ITS EFFECT ON THE VENTILATORY LUNG FUNCTION

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ABSTRACT

Health examinations were made among aluminum smelting workers engaged at two different plants. The air concentrations of fluorides at the respective work places were below the threshold limit values. X-ray and physical examinations of the subjects revealed no abnormalities. The serum and urinary fluoride concentrations, as an index of the fluoride body burden, were higher in exposed workers than in workers of the control group. The urinary fluoride levels were in proportion to the air concentrations of fluorides.

Ventilatory function tests were carried out in combination with a survey of respiratory symptoms. Persistent cough and phlegm was observed in two workers in a relatively highly fluoride-exposed group. No differences in FVC % and $FEV_{1.0\%}$ between the groups were found, but measurements of the maximal expiratory flow curve ($\dot{V}_{max}$) revealed that $\dot{V}$ 50 and $\dot{V}$25 were significantly lower in exposed workers than in the control group. Correlations between these indices and those of the fluoride body burden were well recognized.

Aluminum is produced by the electrolytic reduction of alumina in molten cryolite. The main problem for occupational health in the electrolytic extraction of aluminum is the effect of fluorides on the human body which is caused by the leakage of dusts and gases in industrial processes. Highly reactive materials like gaseous fluoride and hydrogen fluoride cause bronchial irritation. Water-soluble particulate fluorides as well as gaseous ones become rapidly absorbed through the lung to be partly deposited in the hard tissues selectively. Furthermore, if sufficiently large amounts of fluorides are present, osteosclerosis may occur.

There have been several reports related to the effects of fluorides on the human body since Møller and Gudjonsson[8] described the osseous changes observed during the radiologic examination of Danish workmen exposed to fluoride. In recent aluminum plants of Japan, however, the quality of the air at work places has been so improved that there are now few plants in which the

threshold limit values are exceeded[2]. Therefore nowadays much attention is given to the effects on human health due to a low concentration of fluorides exposure.

The aim of this study was to evaluate the health of workers in aluminum plants. The aluminum smelters referred to in this study are of the Söderberg type.

SUBJECTS AND METHODS

The subjects examined formed three groups of male workers living in different parts of Japan, two groups consisted of employees from two different aluminum plant (plant A-Group A, plant B-Group B), while the third group (Group C) was chosen as a control and consisted of residents living in an urban area with no particular sources of airborne fluorides. The number of subjects, their average age, and duration of employment are listed in Table 1.

TABLE 1
Number of subjects in each group, age and duration of employment (mean ± S.D.).

	Groups under study		
	A	B	C
Number of subjects	42	43	110
Average age of group	42.3 ± 8.3	49.5 ± 4.3	39.4 ± 4.1
Average time of employment (years)	17.7 ± 8.3	12.9 ± 6.7	–

Fluorides in the air were collected on membrane filters of 0.8 μm pore size and alkali-limed filters of 47 mm in diameter by means of a low volume air sampler. The air concentrations of fluorides were determined separately for gaseous and water-soluble particulate fluorides by means of a fluoride specific electrode. Fluoride concentrations in serum and urine were determined by the same method[4]. Twenty-four-hour samples of urine were collected from all subjects examined.

All subjects were medically examined. The examination included clinical and occupational history and physical examination. An X-ray examination of the chest was made of all subjects, and roentgenograms of the pelvis and lumbar spine were taken of fluoride-exposed workers.

The prevalence of respiratory symptoms was surveyed by means of a BMRC[3]. In addition ventilatory pulmonary function tests were carried out using a flow-volume curve recorder.

RESULTS

General health

There were no findings or disorders apparently due to fluorides exposure. A few subjects had headache, diarrhea, hypertension, albuminuria and glycosuria.

The general health of both the exposed and the control workers was found to be good.

In X-ray examinations, no lung or osseous changes were noted.

Environmental fluorides data

The concentrations of gaseous fluorides at the work places of plant A were five to ten times higher than those in plant B (Table 2). Water-soluble particulate fluorides concentrations were approximately of the same level in workers from both plants. Air fluorides concentrations in these plants were kept lower than the proposed standard levels. In contrast, airborne fluoride concentrations in the control plant were under 0.05 μg F/m³, as seen in other non-polluted areas in Japan[5].

TABLE 2
Air concentrations of fluoride in each group (μg F/m³).

	Groups under study		
	A	B	C
Gaseous fluoride	200–1 000	20–200	< 0.05
Particulate fluoride	50– 250	20–400	< 0.05

Body burden of fluoride

Fluoride concentrations in serum are shown in Figure 1. The percentage of subjects with more than 20 μg/l of serum fluoride concentrations was 52.4% in plant A and 41.8% in plant B. However, almost all individuals in the control group had values under 20 μg/l.

Urinary fluoride concentration and amounts of excretion per day are given in Figures 2 and 3, respectively. The mean values of potroom workers were higher than those of the control group, the differences being statistically significant at 0.1% level. In addition, it was found that urinary fluoride levels in workers of group A were significantly higher than in those of group B.

Effects on the respiratory system

The prevalence of respiratory symptoms is listed in Table 3. As a rule, respiratory complaints were more frequent in plant A group than in the two other groups. Persistent cough and phlegm, which is related to chronic bronchitis, were observed in two workers from group A.

The results of the measurement of the lung function are shown in Table 4.

No differences in FVC% and $FEV_{1.0}$% values were noted between the fluoride-exposed groups and the non-exposed group, but measurements of $\dot{V}_{max}$ revealed that $\dot{V}50$ and $\dot{V}25$ were significantly lower in exposed workers than in workers of the control group. Moreover, $\dot{V}50/\dot{V}25$ of exposed workers was higher than that of the control group.

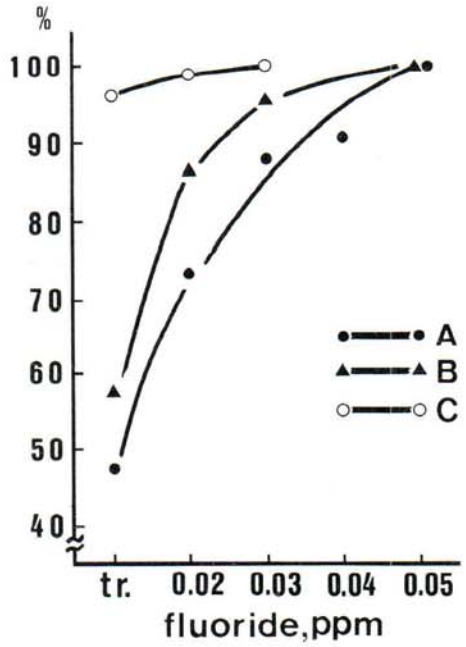

FIG. 1 – Cumulative frequency curves of serum fluoride concentration.

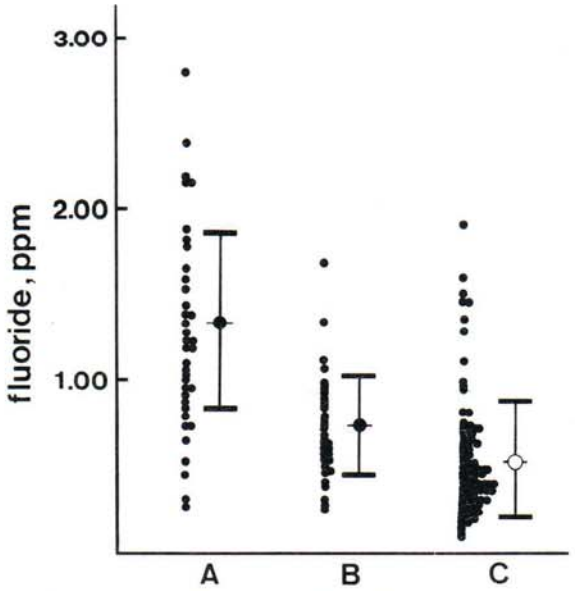

FIG. 2 – Urinary fluoride concentration: mean ± 1 S.D. is 1.35 ± 0.52 ppm in group A, 0.74 ± 0.29 ppm in group B and 0.54 ± 0.34 ppm in group C, respectively.

TABLE 3
Prevalence of respiratory symptoms

Symptoms	Code*	Groups under study		
		A	B	C
Cough, a.m., winter	(1)	7 (16.7)**	4 (9.3)	3 (2.7)
Cough, day/night, winter	(3)	8 (19.0)	3 (7.0)	3 (2.7)
Persistent cough (for 3 months in a year)	(5)	5 (11.9)	1 (2.3)	0 (0.0)
Phlegm, a.m., winter	(6)	9 (21.4)	5 (11.6)	9 (8.2)
Phlegm day/night, winter	(8)	11 (26.2)	3 (7.0)	12 (10.9)
Persistent phlegm (for 3 months in a year)	(10)	5 (11.9)	1 (2.3)	4 (3.6)
Exacerbations of cough and phlegm	(12b)	3 (7.1)	1 (2.3)	1 (0.9)
Phlegm with blood	(13)	0 (0.0)	0 (0.0)	2 (1.8)
Dyspnea, grade 4	(14c)	2 (4.8)	0 (0.0)	1 (0.9)
Wheezing	(15a)	2 (4.8)	1 (2.3)	0 (0.0)
Cough, grade 1	(1/3 + 5)	1 (2.4)	0 (0.0)	0 (0.0)
Cough, grade 2	(1 + 3 + 5)	3 (7.1)	1 (2.3)	0 (0.0)
Phlegm, grade 1	(6/8 + 10)	0 (0.0)	0 (0.0)	1 (0.9)
Phlegm, grade 2	(6 + 8 + 10)	5 (11.9)	1 (2.3)	1 (0.9)
Persistent cough and phlegm	(5 + 10)	2 (4.8)	0 (0.0)	0 (0.0)
	(1/3 + 5 + 6/8 + 10)	1 (2.4)	0 (0.0)	0 (0.0)
	(1 + 3 + 5 + 6 + 8 + 10)	1 (2.4)	0 (0.0)	0 (0.0)
Exacerbations of persistent phlegm	(6/8 + 10 + 12b/c)	2 (4.8)	0 (0.0)	0 (0.0)

* Numbers after symptoms are questionnaire code numbers.
** Numbers in brackets indicate percentage.

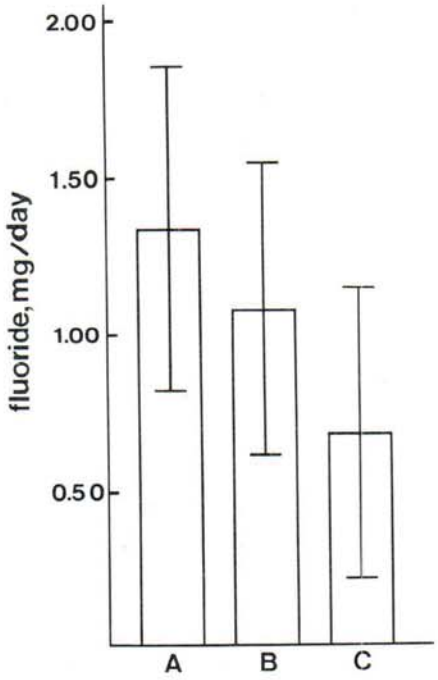

FIG. 3 – Urinary fluoride excretion; mean ± 1 S.D. is 1.34 ± 0.51 mg/day in group A, 1.08 ± 0.46 mg/day in group B and 0.68 ± 0.47 mg/day in group C, respectively.

TABLE 4
Pulmonary function tests (means ± 1 S.D.).

		Groups under study		
		A	B	C
FVC%	%	100.1 ± 12.6	100.8 ± 11.3	106.8 ± 15.4
$FEV_{1.0}$%	%	79.6 ± 6.7	80.8 ± 4.7	81.0 ± 6.8
P.F.	l/s	8.9 ± 1.6	9.3 ± 1.6	8.9 ± 1.5
P.F./Ht.	l/s m	5.4 ± 0.9	5.7 ± 0.9	5.4 ± 0.9
$\dot{V}50$	l/s	3.5 ± 1.2**	3.8 ± 1.1**	4.4 ± 1.1
$\dot{V}50$/Ht.	l/s m	2.2 ± 0.7**	2.3 ± 0.7**	2.7 ± 0.7
$\dot{V}25$	l/s	1.2 ± 0.5**	1.1 ± 0.4**	1.6 ± 0.5
$\dot{V}25$/Ht.	l/s m	0.7 ± 0.3**	0.7 ± 0.2**	1.0 ± 0.3
$\dot{V}50/\dot{V}25$		3.1 ± 0.7*	3.7 ± 1.3**	2.8 ± 0.7

FVC% = % Forced vital capacity; $FEV_{1.0}$% = Forced expiratory volume, in 1sec.%; P.F. = Peak flow; $\dot{V}50$ = Flow at 50% of vital capacity; $\dot{V}25$ = Flow at 25% of vital capacity; Ht. = Height; *P < 0.05, **P < 0.01.

DISCUSSION

Osteosclerosis of fluoride-exposed workers has been usually discussed in reference to the air concentrations of fluorides and urinary fluorides levels. Kaltreider and co-workers[6] have stated that 96% of the potroom workers examined exhibited osteosclerosis in the fluoride air concentrations ranging between 2.4–6.0 mg F/m³. In the study of Agate and co-workers[1] only "a few" workers showed any "abnormality" in exposures ranging from 0.14–3.4 mg F/m³. Largent[7] states that if urinary fluoride levels rarely or never exceed 4 mg per liter, there is little likelihood that even the slightest increase in bone opacity will occur. Tsunoda[9] has emphasized the significance of the urinary fluoride level as an index of the fluorides body burden.

Comparing the indices of the fluoride body burden in the control group with those in the exposed worker groups, potroom workers showed apparent excessive fluoride exposure, but there was little risk to induce osteosclerosis as estimated by their urinary fluoride levels. In fact, none of the examined workers was found to have increased bone density.

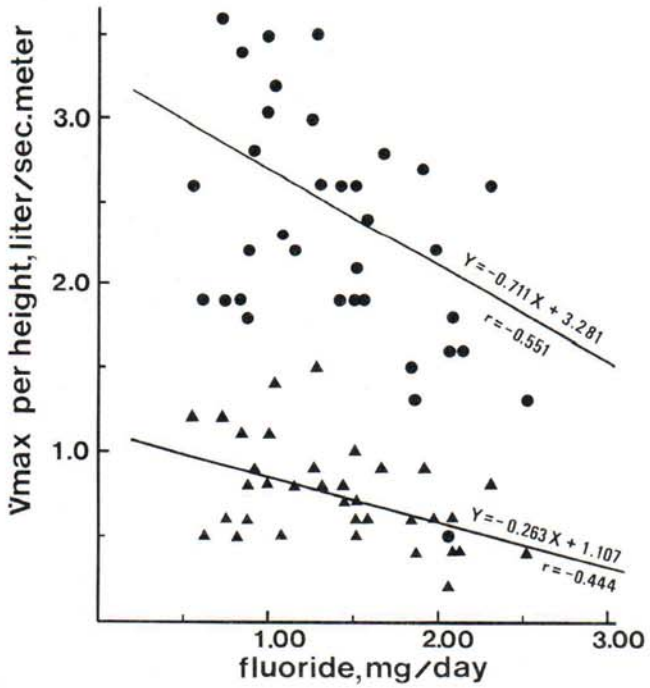

FIG. 4 – Correlation of $\dot{V}_{max}$ per height and urinary fluoride excretion in group A
V̇50 per height vs. urinary fluoride excretion
V̇25 per height vs. urinary fluoride excretion.

However, in the group of potroom workers from plant A with a relatively high exposure to fluoride, the prevalence of respiratory symptoms and $\dot{V}_{max}$ values differed from the other two groups. These results revealed the effects of fluorides on the respiratory system. As shown in Figure 4 $\dot{V}50$ and $\dot{V}25$ decreased in workers from group A as the amounts of urinary fluoride excretion increased. The coefficients correlation was statistically significant ($P < 0.01$). This tendency was not observed in the other groups. These findings suggest that fluoride inhaled through the respiratory tract has an effect on the peripheral airways[10] which can be detected by recording the flow-volume curve.

The results imply that the ventilatory functions are already affected by fluoride, in ranges between 0.2–1.0 mg/m^3 for gaseous fluoride and 0.05–0.25 mg/m^3 for particulate fluorides, although at that level of exposure osteosclerosis was not observed.

REFERENCES

1. *Agate, J.N., Bell, G.H., Boddie, G. F., Bowler, R.G., Buckell, M., Cheeseman, E. J., Douglas, T.H.J., Druett, H.A., Garrad, J. Hunter, D., Perry, K.M.A., Richardson, J. D. and Weir, J.B. deV.* Industrial Fluorosis. A study of the hazard to man and animals near Fort William, Scotland. Med. Res. Council Memorandum No. 22, H.M. Stationery Office, London, 1949.
2. *Committee on Threshold Limit Values.* American Conference of Governmental Industrial Hygienists, Cincinnati, 1970, p. 27.
3. *Committee on the Aetiology of Chronic Bronchitis.* Report to the Medical Research Council. Definition and classification of chronic bronchitis for clinical and epidemiological purposes. Lancet, **1** (1965) 775–779.
4. *Ishikawa, S.* Significance of urinary fluorine content as an index of fluorine body burden. J. Iwate Med. Assoc., **28** (1976) 207–222.
5. *Itai, K.* Concentration of fluorides in the air of various communities in Japan. J. Jpn. Soc. Air Pollut., **11** (1975) 233–234.
6. *Kaltreider, N. L., Elder, M. J., Cralley, L. V. and Colwell, M.O.* Health survey of aluminum workers with special reference to fluoride exposure. J. Occup. Med., **14** (1972) 531–541.
7. *Largent, E. J.* Fluorosis. The Health Aspects of Fluorine Compounds. Columbus, Ohio State University Press, 1961.
8. *Møller, P.F. and Gudjonsson, S.V.* Massive fluorosis of bones and ligaments. Acta Radiol., **13** (1932) 269–294.
9. *Tsunoda, H.* Effects of airborne fluoride on human health. Rodo No Kagaku, **31** (1976) 10–16.
10. *Yazaki, K., Nakaya, S., Suzuki, Y. and Tsunoda, H.* Epidemiologic study on the human effects of air pollution by airborne fluoride. Part 2. Proceedings of 18th Annual Meeting of Japan Society of Air Pollution, 1977, p. 250.