Industrial Health 2006, **44**, 64–68 **Original Article**

Health Effects of Occupational Exposure to Fluorine and Its Compounds in a Small-Scale Enterprise

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Received August 31, 2005 and accepted November 24, 2005

Abstract: A 7-yr study was conducted to evaluate the respiratory effects of fluorine compounds on exposed workers in a small-scale enamel enterprise. Air monitoring was done and 75 pairs, exposed and non-exposed workers were examined. The applied tests were: an epidemiological questionnaire, the bronchitis record, clinical examinations and urinary fluorine. Linear regression analysis was done. The values of fluorine in the air of workplaces ranged from 0.1 to 3.7 mg/m^3 air during the study. Twenty five point and seven percent of exposed workers presented chronic rhinitis, laryngotracheitis and bronchitis. The smoking habit was similar in both groups. The incidence of chronic bronchitis was significantly higher in exposed workers compared to the non-exposed ones. The values of urinary fluorine were higher in the exposed versus the control group. Linear regression analysis has shown positive correlations between the fluorine exposure and incidence of chronic bronchitis (r=0.75), as well as the incidence of chronic respiratory diseases (r=0.71). Fluorine exposure may be responsible for the high incidence of chronic irritative respiratory diseases, especially for chronic bronchitis in exposed workers. For diseases prevention it is advisable to reduce the levels of fluorine in the air of workplaces and to decrease the concentration of fluorine compounds in the composition of enamel.

Key words: Occupational exposure to fluorine compounds, Small-scale enamel enterprise, Health effects, Chronic bronchitis

Introduction

Enameling is an old and widely adopted technology. Enamel or vitreous enamel is the colorful result of fusion of powdered glass (which contains silica, rich in fluoride salts) to a substrate through the process of firing, usually between 750 and 850 °C. The powder melts and flows to harden as a smooth, durable vitreous coating on metal, glass or ceramic. The durability of enamel has given its many functional applications including: interior walls of ovens, speckleware cooking pots, exterior walls of high quality kitchen appliances. In above mentioned small-scale enamel enterprise enameling is used to make interior and exterior walls of ovens. Color in enamel is obtained by the addition of various minerals, often based on the elements as cobalt, iron, etc. In enameling process, fluorine and its compounds always are present in the air of the workplaces. Fluorine and fluorides (the ionic form of fluorine) are combined with the watereous-vapours from the air resulting anhydrous hydrogen fluoride and aqueous hydrofluoric acid.

Limited data exist on the toxicity of fluorine and its compounds. Here are a few notes about the hazards and risks associated with fluorine. Skeletal and dental fluorosis has been observed in workers exposed to hydrocarbon fluoride and fluoride dusts. Chronic exposures to hydrogen fluoride and cryolite dust have resulted in impaired lung function in workers. The observed respiratory effects are attributed to its highly corrosive properties. Fluorine and hydrogen fluoride are highly reactive chemicals; direct contact can result in severe damage to the skin or eyes—directly related to the concentration and duration of the exposure. Fluorine (F_2) gas is a severe irritant to the eyes, skin and respiratory

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tract; intermittent exposures to 10–15 ppm causing irritations at the above mentioned levels. Fluoride, hydrogen fluoride, hydrofluoric acid and fluorine are extremely irritating chemicals and can cause tissue damage after direct contact. The respiratory tract is the primary target of toxicity following inhalation exposure to hydrogen fluoride and fluorine. They cause upper respiratory tract irritation in exposures to fluoride $\subseteq$ 0.5 ppm or fluorine $\subseteq$ 10 ppm. At higher concentration pulmonary congestion, necrosis and/or edema have been observed. Repeated exposures may cause pulmonary and nasal irritations.

Fluoride ion is a very useful component of drinking water (in very low concentrations) since it renders tooth enamel (replacing the OH group of hydroxyapatite) relatively immune to bacteriological attack. The average dietary intake (including water) of fluoride ranges between 1.4–3.4 mg/d (0.02–0.048 mg/kg/d) for adults, living in areas with 1.0 mg/l fluoride in the water. Fluoride salts (fluorides) are naturally occurring components of rocks and soil (ex. sodium fluoride).

A seven-year follow-up study was conducted to evaluate the health status of workers occupationally exposed to fluorine and its compounds in a small-scale enamel enterprise. In this study we were focusing mainly on the irritative effects of fluorine and its compounds on eyes, skin and upper respiratory system in long term exposure.

Materials and Methods

Long-term air monitoring in the workplaces concerning fluorine and its compounds was done - during a 7-yr period: 1995 - 2001. The levels of fluorine and its compounds in the air of all workplaces were measured twice in a year. The yearly medium values were established to evaluate the degree of exposure. Seventy-five pairs of workers, exposed and non-exposed were examined. Their mean age was 39.9 ± 3.3 yr and their mean length in work (exposure time) was 17.1 ± 1.9 yr.

The applied tests consisted of:

- an epidemiological questionnaire regarding the fluorine and its compounds' effects on health status of the exposed workers;
- the bronchitis record for detecting the occurrence of chronic bronchitis in exposed and non-exposed workers; frequency of occurrence of chronic bronchitis was connected with smoking habit and age;
- the usual clinical examinations and functional ventilatory tests, height and weight measurements and rhinolaryngologic examinations (in both groups);
- yearly urinary fluorine determinations (in both groups);
- investigations regarding the smoking habit of the workers —using the Comparative Score Method (in both groups);
- Linear Regression Analysis was done to find out the relationship between the exposure to fluorine and its compounds and health effects, especially the incidence of chronic bronchitis and chronic respiratory diseases in the ocupationally exposed workers.

Results

Long-term air monitoring in the workplaces has shown that the values of fluorine in the air of all workplaces ranged from 0.1 to 3.7 mg/m^3 air. The maximal admissible concentration (MAC) of fluorine compounds is 2 mg/m^3 air. The yearly values of fluorine and its compounds during the study are shown in Fig. 1.

Figure 2 represents the medium values (the medium of the 7-yr) of fluorine and its compounds for all the six studied workplaces.

The values of urinary fluorine were higher in the exposed (6–8 mg/l) versus the control group (1–3 mg/l), which attest the existence of exposure to fluorine and its compounds in this small-scale enterprise (the water fluoride-content being 0.9–1.1 mg/l).

During this long-term study, the workers were exposed chronically mainly to intermittent low and medium concentrations of fluorine. The epidemiological questionnaire and the usual clinical examinations revealed the occurrence

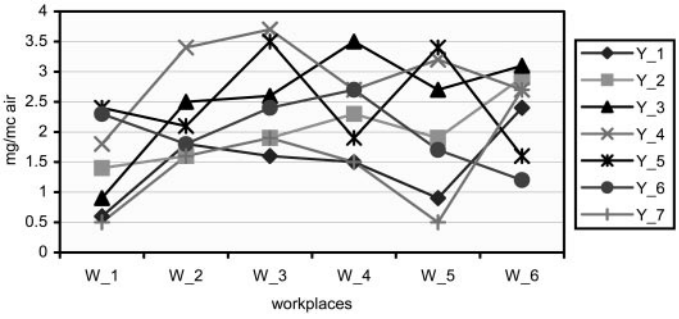

Fig. 1. Long-term air monitoring of the workplaces concerning fluorine and its compounds.

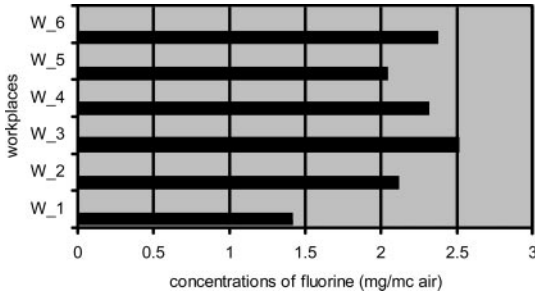

Fig. 2. Medium values of fluorine in the air of workplaces during the study.

of skeletal and rare dental flourosis in the exposed workers, as well as irritations in eyes, skin and the respiratory tract. In this study we were focusing on the irritative effects of fluorine and its compounds at different levels, and mainly, we were searching for the occurrence of chronic respiratory diseases, such as chronic rhinitis, chronic laryngotracheitis and chronic bronchitis. Figure 3 represents the incidence of chronic irritative effects of fluorine and its compounds: 25.7% irritations at the level of upper respiratory tract, 14.9% eyes irritations, 1.8% skin irritations and 3.8% others.

To find out the contribution of smoking in the occurance of chronic respiratory diseases, mainly to the occurrence of chronic bronchitis in exposed and non-exposed workers, the smoking habit was evaluated in both groups. The epidemiological questionnaire, which we used, also contains a part which investigated the negative habits of the exposed and non-exposed workers, such as smoking (light, medium and heavy smokers), alcohol intake (sortiment and quantity) and coffee consumption. All the questions were quoted with a number and a scor was calculated for each negative habit. The negative habits of the studied groups were compared with the Comparative Score Method. The smoking habit was similar in both groups. The majority of the workers were non-smokers; their prevalence was over 60% in both, exposed and non-exposed groups. Thirty eight percent of exposed workers were found smokers: light (8%), medium (19%) and heavy (11%) smokers. The percentage of the number of cigarette smoking was very similar in the non-exposed workers.

History, clinical examinations (physical examination with height and weight measurement, respiratory function tests and rhinolaryngologic examinations) and the chronic bronchitis record were used to find out the occurence of chronic bronchitis and other chronic respiratory (upper tract) diseases in both studied groups. Figure 5 presents the details about the incidence of chronic bronchitis and other chronic respiratory diseases in both groups. The incidence of chronic bronchitis was significantly higher ($p<0.05$) in exposed workers compared to the non-exposed ones. There is also a statistically significant difference ($p<0.05$) between the incidence of chronic rhinitis in exposed and non-exposed workers.

The respiratory tract is the primary target of toxicity following inhalation exposure to hydrogen fluoride and fluorine. They cause upper respiratory tract irritation in exposures to fluoride $\subseteq$ 0.5 ppm or fluorine $\subseteq$ 10 ppm. Repeated exposures may cause chronic pulmonary, bronchial and nasal irritations.

Linear Regression Analysis was done to find out the relationship between the exposure to fluorine and its compounds and health effects, especially the incidence of chronic bronchitis and chronic respiratory diseases in the occupationally exposed workers. There were two correlations between the observed exposures and health effects:

1) Relationship between the fluorine exposure and incidence of chronic bronchitis
2) Relationship between the fluorine exposure and

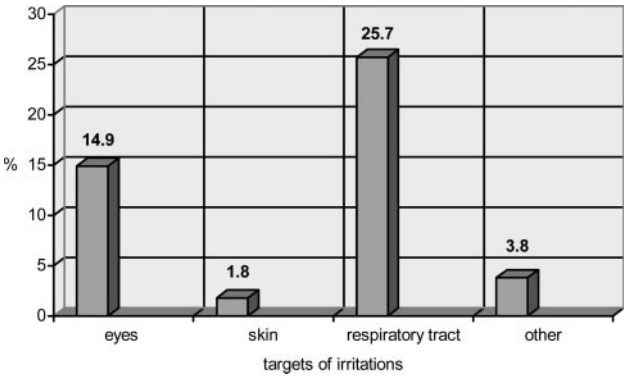

Fig. 3. The irritative effects of fluorine and its compounds in exposed workers.

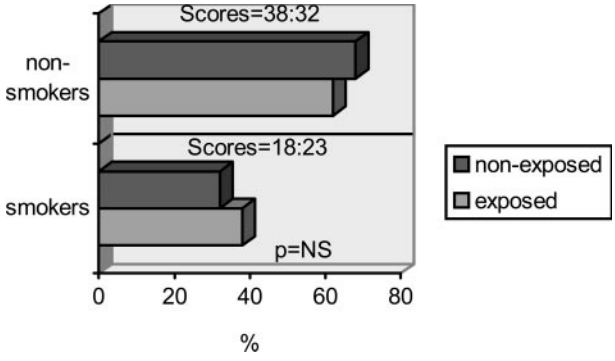

Fig. 4. The smoking habit in the studied groups.

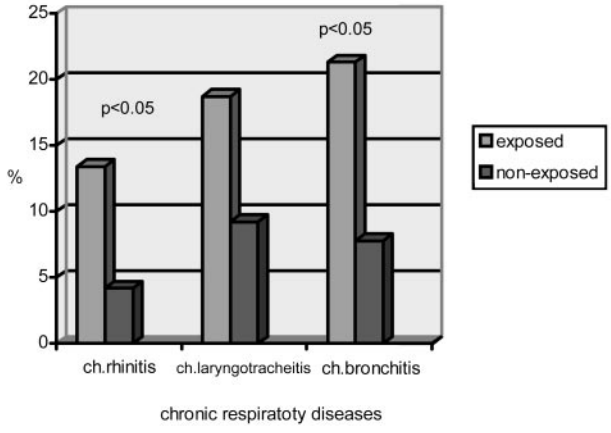

Fig. 5. The incidence of chronic bronchitis and other chronic respiratory diseases.

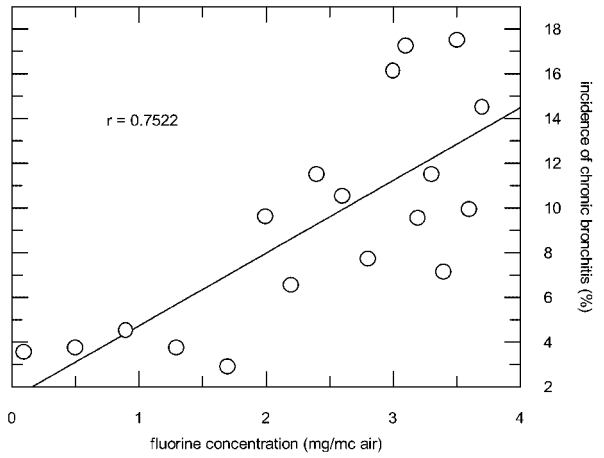

Fig. 6. Relationship between the fluorine exposure and incidence of chronic bronchitis.

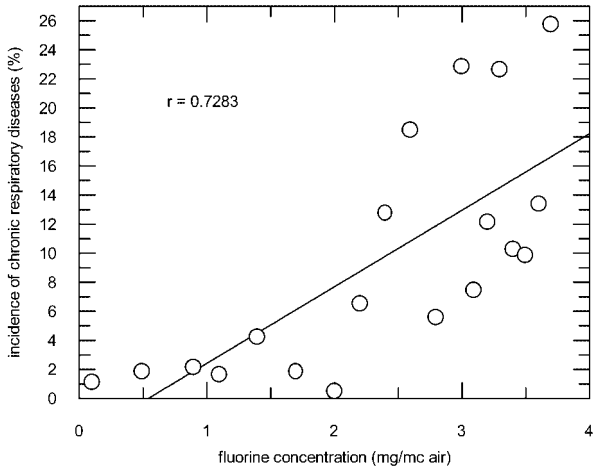

Fig. 7. Relationship between the fluorine exposure and incidence of chronic respiratory diseases.

incidence of chronic respiratory diseases

Following the above mentioned Figure (Fig. 6), we may observe a direct, close relationship between the exposure to fluorine and its compounds and the incidence of chronic bronchitis in the exposed workers, the Correlation Coefficient (r) being 0.7522. The curve of the correlation has a positive and ascendent way (with appropriate Intercept and Slope).

The situation is very similar regarding the relationship between the exposure to fluorine and its compounds and the incidence of chronic respiratory diseases in exposed workers (Fig. 7). The shape of linear regression curve in this case is also positive and ascendent; the Correlation Coefficient being 0.7283 (with appropriate Intercept and Slope).

Discussion and Conclusions

The present study focused only on the irritative effects of fluorine and its compounds in the exposed workers, because of the length and complexity of the problems. This follow-up study also needs to be completed and deepened in following years. Initially this enameling factory was a big plant, but after the economic-political changes it has became a small-scale enterprise according to the general trends in our country.

This seven-year follow-up study was conducted during the period between 1995 and 2001, which represents only about a half time from the mean exposure time of these 75 studied workers (17.1 ± 1.9 yr). Thus, the real exposure time of about 15 yr was enough for the fluorine and its compounds to produce chronic irritations, as chronic respiratory diseases—even at the intermittent low and medium concentrations of fluorine.

The values of concentration of fluorine and its compounds (which represents the yearly average of these concentrations) was fluctuate according to the year in the studied workplaces.

These fluctuations are determined by a lot of factors, as shown in Fig. 1.:

- the concentration of fluorine compounds in the composition of enamels (the raw materials were procured not constantly, depending on the cost-effectiveness relation from different sources)
- the intensity of the working process in the given/ controlled period (working with total or partial capacity)
- the functionality of their ventilation-system
- the outside weather
- the time of measurements/determinations

It was also noticed on the same figure, that the values of concentration of the fluorine compounds in workplace-1 (W-1) were comparatively lower than the values obtained in other workplaces. The explanation for this finding may be the fact that the enameling process begins in W-1 with the handling of powdered glass in appropriate recipients. There is no process of firing, no high temperatures, no irritant fluorine compounds formation in a considerable amount at workplace. Because of the powdered glass dust, the windows were usually wide opened here.

All the 75 studied exposed workers were distributed proportionally in the above mentioned workplaces, as follow: W-1 (10), W-2 (11), W-3 (14), W-4 (13), W-5 (14), W-6 (13). We have also tried to analyze the incidence of chronic respiratory diseases according to the differences of exposing duration and amount, but the results were not statistically significant, compared with the recent study. All the 75 workers were exposed mainly to intermittent low and medium concentrations of fluorine and in long-term exposure these exposing amounts/values have shown to be able to induce chronic irritations, as chronic respiratory diseases.

Our conclusions of this part of the study were the followings:

1) Long-term fluorine exposure may be responsible for the high incidence of chronic irritative respiratory diseases, especially of chronic bronchitis in exposed workers.

2) To prevent chronic respiratory diseases, mainly the chronic bronchitis in exposed workers to fluorine and its compounds, it is advisable:

- to reduce the levels of fluorine and its compounds in the air of all workplaces by improving their ventilation systems;
- to use the individual protection equipment in correct and appropriate manner according to the General and Special National Work Protection Standards;
- to decrease the concentration of fluorine compounds in the composition of enamels.

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