

# Urinary fluoride concentration as an estimator of welding fume exposure from basic electrodes

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**ABSTRACT** Urinary fluoride concentrations have been measured in electric arc welders using basic electrodes. The fluoride concentration and the total welding fume concentration in air showed a linear relation with postshift urinary fluoride concentration. The measured concentrations were below internationally recommended postshift urinary fluoride concentrations believed to cause fluorosis. Biological monitoring by postshift urinary fluoride measurements is evaluated for the prediction of total welding fume exposure, when a specific basic electrode was used, by means of confidence limits and tests of validity.

Electric arc welding generates fume in which most of the particles are smaller than 1  $\mu\text{m}$ .<sup>1</sup> Particulate fluorides are emitted from basic electrodes coated with calcium fluoride and such electrodes are commonly used in arc welding. Some earlier studies have shown a higher excretion of fluorides in urine among welders exposed in this way as compared with non-welding referents.<sup>2-4</sup> Most studies of the correlation between air and urinary fluoride concentrations, however, have been conducted in other industrial settings where exposure comes from the manufacture of fertilisers,<sup>5</sup> the electrolytic production of aluminium,<sup>6</sup> and from hydrofluoric acid steeps.<sup>7</sup>

The purpose of this study was primarily to determine the relation between the particle fluoride concentration in air and the postshift fluoride concentration in the urine and, secondly, to investigate the relation between the total concentration of welding fume particles and the postshift concentration of fluoride in the urine of welders using a specific electrode with a known composition of calcium fluoride. If such a strong relation exists, then postshift urinary fluoride concentration could be used for biological monitoring of welding fume exposure.

This study was a part of the Swedish welding project in which several hygienic and health aspects of welding<sup>8,9</sup> have been investigated.

## Material

In the first study in which airborne particulate fluoride and urinary fluoride concentrations were measured, 11 welders from four different companies participated. Their mean age was 42 (range 28-63). The welders worked with different types of basic electrodes in stainless steel and their welding exposure time varied from eight to 42 years (mean 18 years).

The second study in which total welding fume exposure and urinary fluoride concentrations were measured involved 64 railway track welders. The mean age in this group was 48 (range 28-59) and all these welders worked with the same basic electrode (OK 74-78), the fume from which consists of 18-20% fluorides, according to the manufacturer (ESAB). The welding exposure time in this group varied from five to 41 years (mean 18 years).

The referent subjects were 70 non-welding workers employed by the Swedish State Railways. They were occupied with electric and signal installations and track construction. Their mean age was 47 (range 27-59) and they lived in the same areas as the railway track welders.

All participants, welders and referents, lived in different parts of Sweden, from Malmö in the south to Boden in the north. All were men.

## Methods

The welding fume samples were collected on celu-

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Table 1 Postshift urinary fluoride concentrations ( $\mu\text{mol/l}$ ) unadjusted for standard density.

		Median	Range
All welders	(n = 75)	58	10-221
Referents	(n = 70)	26	<10-137

lose ester membrane filters (Millipore filter AAWP  $\phi$  37 mm, mean pore size  $0.8 \mu\text{m}$ ) with personal sampling pumps (MSA Monitaire sampler pump, model G). The air flow was checked several times during the sampling period and the filters were placed inside the welding helmet. The sampling time was at least five hours for total particle measurements and at least four hours for fluoride measurements. Total particle welding fume concentrations were measured gravimetrically.

Fluoride concentrations in air were measured by treating the filters with sulphuric acid, the evaporated gas being collected in a solution of sodium hydroxide. Buffer was added and the fluoride concentration was determined with an ion specific electrode.<sup>10</sup> The detection limit of fluoride on the filter was  $0.4 \mu\text{g}$  and the coefficient of variation was about 5%.

At the end of the work shift, welders and referents collected urine into polyethylene bottles. The density of the freshly voided urine was measured with a refractometer. Equal parts of urine and a buffer of sodium acetate and citrate (pH 5.25) were mixed and an ion specific electrode was used for the determination of fluoride.<sup>10</sup> The detection limit was  $10 \mu\text{mol/l}$  and the coefficient of variation was 2-5%.

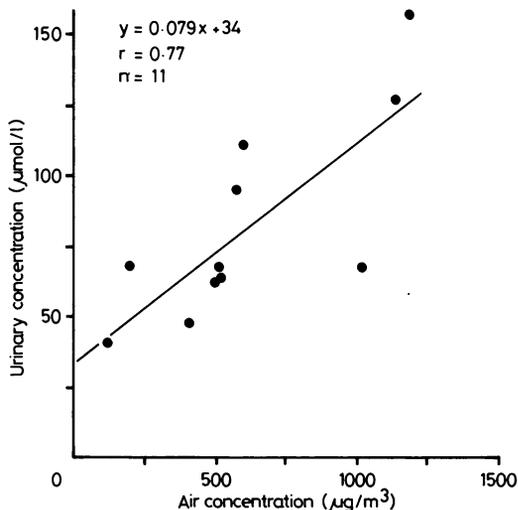


Fig 1 Air and unadjusted postshift urinary concentrations of fluoride.

Urine concentrations were calculated unadjusted as well as adjusted to a standard density of  $1.024$ .<sup>11</sup>

Individual air and urine concentrations were measured on the same workday. This day was arbitrarily chosen in the workweek to represent normal working conditions although Mondays were excluded for technical reasons.

Differences of frequencies were tested by chi<sup>2</sup> test and equations of linear regression were calculated by the method of least squares. The p values shown later in the text refer to two tailed tests.

## Results

The welders had higher postshift urinary concentrations of fluoride compared with the non-welding referents (table 1). This difference is highly significant when the welders and the referents were divided at the median of all urinary concentrations and the frequencies tested by chi<sup>2</sup>-test ( $p < 0.001$ ); 95% of the referent values were below  $75 \mu\text{mol/l}$ .

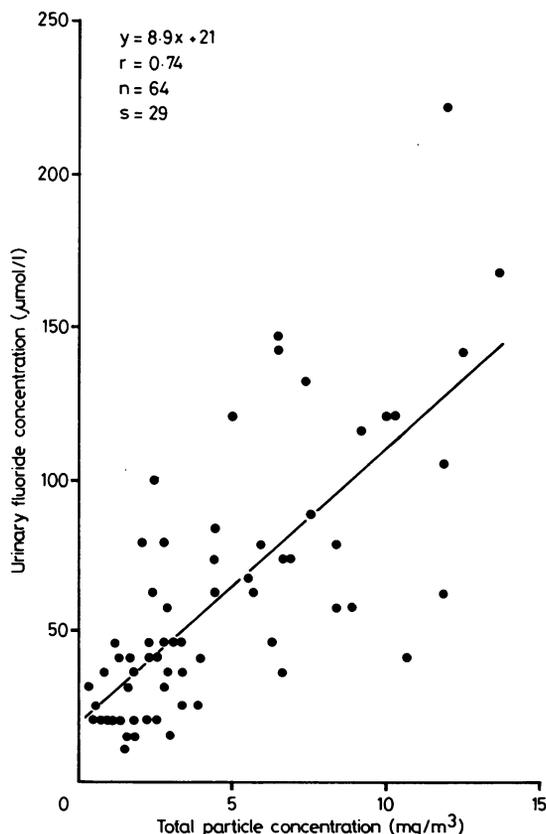


Fig 2 Eight hour TWA total particle concentrations and unadjusted postshift urinary fluoride concentrations.

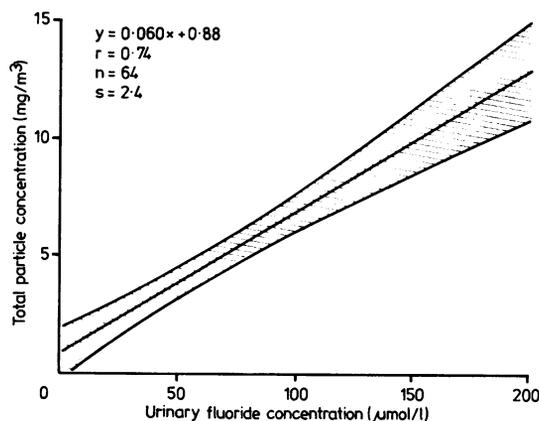


Fig 3 Ninety five per cent confidence limits for eight hour TWA of mean total particle concentrations in relation to unadjusted postshift urinary fluoride concentrations.

There was no relation between age and urinary fluoride excretion in either welders or referents.

The postshift urinary fluoride concentrations showed a linear relationship with the airborne concentration of particulate fluoride for the welders working with different types of basic electrodes ( $r = 0.77$ ,  $p < 0.01$ ) (fig 1). A similar correlation coefficient was obtained after adjustment of the urinary concentrations to standard density ( $r = 0.76$ ,  $p < 0.01$ ) while the equation of regression was  $y = 0.069x + 38$ . These regression lines became steeper ( $y = 0.09x + 39$ ) when the air concentrations were calculated as eight hour time weighted averages.

There was also a linear relationship between postshift urinary fluoride concentration and total welding fume concentration, calculated as eight hour time weighted averages ( $r = 0.74$ ,  $p < 0.001$ ) in welders using a specific basic electrode (OK 74.78) (fig 2). When the urinary concentrations were adjusted to a standard density the correlation coefficient was somewhat lower ( $r = 0.70$ ) while the equation of regression was  $y = 8.4x + 27$ . The postshift urinary fluoride concentrations were similar during the workweek (from Tuesday to Friday), suggesting that there was no tendency for accumulation.

As a linear relationship existed between the total particle welding fume and the urinary fluoride concentrations, it was possible to use the latter as an estimate of exposure to total particle welding fume. A regression line was calculated with urinary fluoride concentration as the independent variable ( $x$ ) and total particle welding fume concentration as the dependent variable ( $y$ ). Figure 3 shows the 95% confidence limits for the prediction of mean air particle concentrations.

## Discussion

The major source of fluoride for most occupationally unexposed people is the drinking water. The absorption of fluoride ions from an ingested dose of soluble fluoride salts is rapid and nearly complete. Between 20% and 33% of an absorbed dose is found in the urine within three to four hours. Less soluble fluoride compounds are absorbed to a lesser degree.<sup>12</sup> When a steady state is reached after multiple oral doses, about 50% of the given amount is excreted in the urine.<sup>13</sup> Children excrete in the urine less fluoride from a given dose compared with adults, which might be explained by a faster uptake of fluoride in the growing skeleton.<sup>14</sup> In adults about 99% of the total body burden of fluoride is found in the skeleton.<sup>15</sup> No relation, however, between age and urinary fluoride concentration was found in this study.

### FLUORIDE IN AIR AND URINE

Some earlier studies have shown an increased excretion of fluorides among welders working with basic electrodes,<sup>2-4</sup> the fluorine compounds in welding fume being mainly in the particulate form. In most investigations the gaseous fluorine compounds have varied from non-detectable concentrations to 10% of the particulate fluorides.<sup>16-19</sup> The linear relationship between particulate air fluoride and urinary fluoride concentrations in welders in this study has not been described previously.<sup>4</sup> The regression equation is reasonably congruent with equations for other industrially exposed groups.<sup>5-7</sup> A validity test for the prediction of air fluoride concentration from urinary concentration has not been made as the number of subjects is too small.

### TOTAL WELDING FUME AND URINARY FLUORIDE LEVELS

A linear relationship was found between the total welding fume concentration and the postshift urinary fluoride concentrations when a specific electrode (OK 74.78) was used. A previous study of aluminium smelter workers showed an increasing excretion of fluorides in postshift samples during the first three days of a workweek despite approximately constant air concentrations.<sup>20</sup> No such tendency was observed in this study which may be due partly to the fact that Mondays were excluded, and partly to the comparatively low concentrations of fluoride in the air.

### PREDICTION OF TOTAL WELDING FUME

As a linear relationship existed between the total welding fume concentration and the postshift urinary fluoride concentration, it was possible to pre-

dict exposure to total welding fume from the urinary fluoride concentration. The prediction is made from a regression line with urinary fluoride concentration as the independent variable and total welding fume concentration as the dependent variable<sup>21</sup> (fig 3). The regression line indicates that the lower 95% confidence limit of the mean air concentration of particles at 5 mg/m<sup>3</sup>, the occupational exposure limit for non-specific respirable particles in Sweden and the United States, corresponds to a urinary fluoride concentration of about 80 µmol/l; this value was exceeded by two of the referents. Welding fumes sometimes contain high fractions of other metals such as manganese or chromium. On these occasions, the occupational exposure limit for non-specific particles is often inadequate, as the occupational exposure limits for these specific chemicals are much lower than for the non-specific particles.

Our results can also be evaluated in terms of sensitivity and specificity. For this purpose, 5 mg/m<sup>3</sup> was chosen as the total air particle concentration limit and 80 µmol/l as the postshift urinary fluoride concentration limit (table 2). Tables constructed for 100% sensitivity (table 3) and 100% specificity (table 4) show that a postshift urinary fluoride concentration below 35 µmol/l will correspond to a total air particle concentration below 5 mg/m<sup>3</sup>, and that a urinary concentration above 105 µmol/l will correspond to an air particle concentration above 5.0 mg/m<sup>3</sup>. Nevertheless, when the urinary fluoride concentrations are between 35 and 104 µmol/l, total air particle concentrations are difficult to predict.

These predictions are valid only if the non-occupational burden of fluoride is approximately equal for the referent group and the welding fume exposed group. The median urinary fluoride concentration for the referent group, 26 µmol/l (table 1), is close to the intercept of the regression equation of the railway track welders (21 µmol/l). This intercept will theoretically reflect the non-occupational concentration of urinary fluoride excretion. Comparing the two welding groups, the stainless steel welders (intercept 34 µmol/l) has, presumably, a higher non-occupational burden of fluoride than the railway track welders.

These proposed predictions are applicable only for groups welding with electrodes emitting 18–20% fluorides in the fume. Other regression lines and confidence limits for the prediction of exposure must be calculated if the electrode composition differs from those used here. The slope of the regression line for the railway track welders (8.9 µmol·m<sup>3</sup>·mg<sup>-1</sup>·l<sup>-1</sup>) differs by a factor 1.8 from the theoretical slope calculated from the slope of the stainless steel welders (0.2 × 79 µmol·m<sup>3</sup>·mg<sup>-1</sup>·l<sup>-1</sup>). This discrepancy may be explained by the relatively

Table 2 Tests of validity for the estimation of total air particle concentrations from postshift urinary fluoride concentrations

	Total air particle concentration (mg/m <sup>3</sup> )		
	≥5.0	<5.0	
Urinary fluoride concentration (µmol/l)	≥80 <80	12 12	2 38
Sensitivity: $\frac{12}{24} = 50\%$			
Specificity: $\frac{38}{40} = 95\%$			
Positive predictive value: $\frac{12}{14} = 86\%$			
Negative predictive value: $\frac{38}{50} = 76\%$			

Table 3 Specificity and predictive value for the estimation of total air particle concentrations from postshift urinary fluoride concentrations after enforcement of 100% sensitivity

	Total air particle concentration (mg/m <sup>3</sup> )		
	≥5.0	<5.0	
Urinary fluoride concentration (µmol/l)	≥35 <35	24 0	22 18
Sensitivity: $\frac{24}{24} = 100\%$			
Specificity: $\frac{18}{40} = 45\%$			
Positive predictive value: $\frac{24}{46} = 52\%$			
Negative predictive value: $\frac{18}{18} = 100\%$			

Table 4 Sensitivity and predictive value for the estimation of total air particle concentrations from postshift urinary fluoride concentrations after enforcement of 100% specificity

	Total air particle concentration (mg/m <sup>3</sup> )		
	≥5.0	<5.0	
Urinary fluoride concentration (µmol/l)	≥105 <105	11 13	0 40
Sensitivity: $\frac{11}{24} = 46\%$			
Specificity: $\frac{40}{40} = 100\%$			
Positive predictive value: $\frac{11}{11} = 100\%$			
Negative predictive value: $\frac{40}{53} = 75\%$			

small number of measurements in the stainless steel welding group. The slopes of the regression lines from previous studies have varied by a factor of 10 for different occupational exposures.<sup>6,7</sup>

#### OSTEOSCLEROSIS

The critical effect of fluoride exposure seems to be osteosclerosis which will not appear if the urinary

fluoride concentration is lower than 260  $\mu\text{mol/l}$  (5 mg/l).<sup>22</sup> The NIOSH criteria document<sup>23</sup> recommends pre-shift concentrations below 210  $\mu\text{mol/l}$  (4 mg/l) and post-shift concentrations below 370  $\mu\text{mol/l}$  (7 mg/l) to avoid negative health effects. Similar limits have recently been suggested by others.<sup>7</sup> The urinary fluoride concentrations found in this study are lower than the levels reported to cause adverse effects.

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