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Evaluation of Biological Monitoring Among Stainless Steel Welders

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Summary. Ten manual metal arc (MMA) high alloy stainless steel (SS) welders were studied during one week and the concentrations of chromium (Cr) and nickel (Ni) were determined in their urine and blood. Stationary and personal air samples were collected from the immediate work environment; they covered the entire work period. Spot urine samples were collected during the follow-up period. Whole blood and plasma samples were taken from the workers before and after one shift, and the retention rate of magnetic dust in the lungs was estimated with magnetopneumography. On the basis of the results, indices of short-term exposure to Cr and Ni were evaluated. Urinary Cr and Ni concentrations (corrected to creatinine) reflect both the body burden caused by long-term and short-term exposure to easily soluble fractions of these metals. The results indicated that the use of Cr and Ni urinary analyses as indices of short-term exposure is not as dependable as previously assumed. The Cr and Ni concentrations in whole blood and plasma did not correlate with the measured exposure, but the daily mean increase in the Cr concentration reflected exposure to total Cr and Cr(VI) very well. The large variation in the Cr concentration of the morning urine (0.01–2.7 μmol/l) and blood (0.05–1.43 μmol/l) indicated large personal variations of body burden among the exposed welders. The retention rate of magnetic dust in the lungs correlated well ($P < 0.01$) with the daily mean increase of Cr in blood. Very good correlations ($P < 0.001$) were found between the retention rate of magnetic dust and the personal air samples of Cr and Cr(VI).

Key words: Biological exposure tests – Chromium – Nickel – Stainless steel welding

Introduction

The occurrence of chromium (Cr) and nickel (Ni) compounds in industrial metal aerosols has risen during the last decade as a result of the increased use of alloyed

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steel. Stainless steel (SS) welding fumes have been recently characterized in detail (Stern 1977a; Ulfvarson et al. 1978; Kimura et al. 1979; Koshi 1979; Lautner et al. 1978; Malmqvist et al. 1980).

The total Cr concentration in stainless steel manual metal arc (MMA/SS) welding fumes varies between 1.5 and 5.2% by weight (Stern 1977a; Ulfvarson et al. 1978; Koshi 1979; Kimura et al. 1979; Malmqvist et al. 1980; Wilson et al. 1981), and the hexavalent chromium [Cr(VI)] fraction from 15 to 90% of the total Cr content (Stern 1977b; Koshi 1979; Malmqvist et al. 1980; Wilson et al. 1981). The concentration of Ni in MMA/SS welding fumes is usually small (0.5%–1.0%), and Ni has been presumed to be present in a poorly water-soluble form (Stern 1977b).

MMA/SS welders are exposed to Cr and Ni compounds, which are known to have a potency to induce occupational health hazards. The most common measure of individual exposure to Cr is the determination of its concentration in urine (Gylseth et al. 1977; Tola et al. 1977; Mutti et al. 1979a; Kalliomäki et al. 1981b). A correlation of 0.88 between the daily mean exposure to water-soluble Cr compounds and urinary excretion corrected to creatinine in a spot sample taken after the work shift has been reported for a group of MMA/SS welders by Tola et al. (1977). A better correlation of 0.95 was found between exposure and the difference in the urinary Cr concentrations before and after the workshift. Gylseth et al. (1977) found an equally high correlation between exposure to total Cr and urinary Cr concentration excreted by three groups of MMA/SS welders after the workshift. Mutti et al. (1979a) reported correlation coefficients of about 0.92 for after-shift Cr concentrations in the urine or increased urinary excretion during the workshift and exposure to water-soluble Cr in MMA/SS welding fumes. The correlation was, however, dependent on previous exposure to MMA/SS welding fumes (Mutti et al. 1979a).

Only a few investigators have studied exposure to Ni compounds present in MMA/SS welding fumes (Kalliomäki et al. 1981b). In estimating the occupational exposure to water-soluble Ni compounds, analyses of Ni concentrations in both plasma and urine have been used (Bernacki et al. 1978; Tola et al. 1979).

The magnetic method has proved suitable for estimating lung contaminants in persons exposed to particulates containing a magnetic component (Kalliomäki et al. 1978a, b, 1979; Freedman et al. 1980; Kalliomäki et al. 1980, 1981a). Many industrial metal dusts and fumes have been shown to contain a magnetic component, which can serve as a tracer of lung retention (Kalliomäki et al. 1981a).

The present study was undertaken to compare the concentrations of metals in urine, plasma and whole blood, and the measured average magnetic field of the chest areas as indices of the individual exposure of MMA/SS welders to welding fumes and grinding dusts containing Ni and Cr compounds. Another purpose was to examine the level of exposure among the welders in a workshop.

Subjects and Exposure

The ten subjects studied were healthy male MMA/SS welders with an average age of 39, SD ± 6 years. Three of them were smokers. Their mean work times as welders was 13, SD ± 6 years. They had only welded stainless steel, using mainly the MMA method. During previous

years, the composition of the alloyed steel welded and electrodes used varied. Three persons working in the same factory, but not exposed to welding fumes, served as controls.

The duration of the welders' normal daily shift was 7 h, during which they did welding, grinding and assembly work. The welding work represented, on the average, one third of the working hours.

During the study period, process containers for chemical industry and heat exchangers were being welded. The materials to be welded were mainly heat resistant, high-alloy steel (x10NiCrAlTi2032, DIN), containing 19–23% Cr and 30–34% Ni and 18/8 stainless steel (x10CrNiTi189, DIN). The most commonly used electrodes were S-NiCr19Nb (DIN) and ETi/99nC23 (DIN).

The workplace was a shop measuring about 3 200 m^2 and 76 700 m^3. It was equipped with general mechanical ventilation. Fresh air (150 000 m^3/h) was blown via 18 grills in the floor, and the same volume was removed with 7 fans on the roof. No local exhaust systems were used at the welding sites. The maximum number of welders working during a shift was 30. The welding, grinding and assembly of process containers took place at this location.

Methods

Personal air, blood, plasma and urine samples were collected during one workweek. Each subject was monitored for 2 consecutive days, 4 persons a day. Urine samples were collected during the follow-up period (48 h) as spot samples taken each time the subjects urinated. Blood samples were taken before and after one workshift during the follow-up period. Personal air samples were collected during the week so that the sampling times would cover the entire work period (7 h).

Dust Samples

Sampling. The personal breathing-zone air samples were collected on cellulose acetate membrane filters (Millipore filter AAWP 037; 3-piece filter holder) and on PVC filters (Nuclepore 0.8 μm) with constant-flow personal sampling pumps (Wärtsilä 8077 DS). The filter holder was placed on the welders collar outside of his mask. The sample flow rate was 2.0 l/min (±5%). The pump was equipped with a turbine transducer with optical speed registration (accuracy ±2%). The sampling time ranged between 350 and 450 min and air volumes between 700 and 900 l. Sampling was stopped only for the lunch break (30 min).

Stationary air samples were collected in the work areas and corridors with the same filter types used in the personal sampling with stationary sampling pumps equipped with a dry gas meter. The nominal flow rate was 20 l/min, and the sampling time varied between 360 and 450 min. Twenty-two personal samples and 12 stationary samples were collected.

The samples for the electron microscopic studies were collected on PVC filters. The air volume sampled ranged between 2 and 10 l. Personal and stationary air samples were analysed for total particulates, main metals and total and hexavalent chromium. Electron microscopic studies were made to estimate the fractions of welding fume and grinding dust particles in the samples.

Metal Analysis. Total particulate concentrations were determined by weighing. Prior to the initial weighing and after the sample collection, the filters were kept in a desiccator for at least 24 h. They were cut into two pieces for different types of analyses. One half of the filter was treated in glass with 10 ml of concentrated nitric acid and 2 ml perchloric acid and boiled in a sand bath until the filter material was dissolved and the solution volume was about 2 ml. The solution was then transferred to a 10-ml test bottle, and water was added until there was 10 ml of the solution. Total chromium (Cr), nickel (Ni), manganese (Mn) and iron (Fe) were determined with an atomic absorption spectrophotometer (AAS) (Perkin Elmer 503). For comparison, the other half of some filters was analysed for total Cr in another laboratory by treating the filter with fusion techniques. The filter with the sample was low-temperature ashed. The ashed filter was fused with a 3 : 1 mixture of sodium carbonate and boric acid. The extract was analysed for total Cr with AAS.

Total Cr(VI) was analysed with the carbonate leaching method described by Thomsen and Stern (1979). The analysis was made from samples taken both on cellulose acetate and PVC filters.

Scanning electron microscopy and energy dispersive X-ray analysis (EDXA) were used to identify the fume and grinding dust particles.

Biological Samples

Sampling. All polyethylene bottles were allowed to stand in a 20% Deconex solution overnight and were rinsed thereafter with distilled water. Urine samples were collected in 100-ml polyethylene bottles for analysis of Cr and Ni. Before the sampling, all subjects showered to avoid contamination.

Whole blood samples were collected in 50-ml heparinized (200 µl heparin/50 ml) polyethylene bottles. Separate blood samples were taken for the whole blood and plasma analyses. The plasma samples were centrifuged (3000 rpm, 15 min) and transferred into clean polyethylene bottles.

Analysis of the Blood and Urine Samples. The urinary concentrations of Cr and Ni were corrected to specific gravity for 10 welders and to creatinine for 5. The concentrations are indicated in micromoles per litre and microgrammes per gramme of creatinine respectively. The urine creatinine concentrations were analysed by the Jaffe reaction with an FP-901 chemistry analyser (Labsystems Oy, Finland). The specific gravity was measured by a SPR-T2-meter (Atago, Japan).

The determinations of Cr and Ni in urine, blood and plasma were performed with direct nitric acid dilution employing AAS. The urinary Cr analysis used was a modification of the procedure of Tola et al. (1977). The method used to analyse Ni in urine in the present study has been found to correlate with the IUPAC-reference method (Brown et al. 1981).

All chemical determinations were made with a model 4000 AAS equipped with a graphite furnace (model HGA 400 and autosampling system AS-40) and a model 56 recorder, all from Perkin-Elmer Corp., Norwalk, CT, USA. The urine and blood samples were diluted with Hamilton's digital diluter before the Cr and Ni analyses.

Magnetic Measuring Method

Measurement of the contaminants retained in the lungs is based upon the measurement of the remanent magnetic field due to ferrimagnetic particles retained in the lungs. This method has been previously used to investigate the pulmonary retention of occupational particulates in mild steel welders, foundry workers, iron and steel plant workers, and stainless steel welders (Kalliomäki et al. 1979, 1980; Koponen et al. 1980; Kalliomäki et al. 1981a, b). The average remanent magnetic field (B), as well as the retention rate of magnetic dust in the lungs (B/t_{ex}) (the average remanent magnetic field divided by the length of the exposure period in years), depends on the magnetic properties of the welding fumes. The properties of these fumes have been studied earlier (Kalliomäki et al. 1981b; Moilanen et al. 1982).

Statistics

A lognormal distribution has been used in the calculations of the average values and the standard deviations. In this study such parameters are referred to by the expressions mean and σg as a distinction from the arithmetic average and standard deviation (SD).

Results

The elemental composition of breathing zone air samples and stationary air samples are shown in Table 1. For total Cr, the samples were analysed in two laboratories. The difference between the results of the two analyses was not significant. The Cr(VI) fraction in the breathing zone was 35% (range 13–63%) of the

Table 1. Composition of manual metal arc (MMA) stainless steel (SS) welding fumes according to the present study and papers published previously

	Total *particulate* mean (mg/m^3)	Cr (%)	Cr(VI)/Cr (%)	Ni (%)	Fe (%)	Mn (%)	Number of samples
Present study[a]							
Personal air samples	5.4	3.6	35	3.3	7.9	2.8	22
Stationary air samples	0.8	3.4	–	3.7	12.2	1.7	10
Tola et al. (1977)[b]							
Personal air samples	7.4	2.4	67	–	–	–	
Mutti et al. (1979a)[a]							
Personal air samples	17.4	3-6	67	–	–	–	
Stern (1977b, c)	–	3.8	–	0.5	6.3	2.7	
Ulfvarson et al. (1978)							
Stationary air samples	–	4.0	–	0.5	8.0	6.0	
Malmqvist et al. (1980)[c]		5.1		0.4	4.0	6.1	

[a] Welders did welding and grinding
[b] Welders did mostly welding and some grinding
[c] MMA/SS welding fumes were generated in a laboratory environment

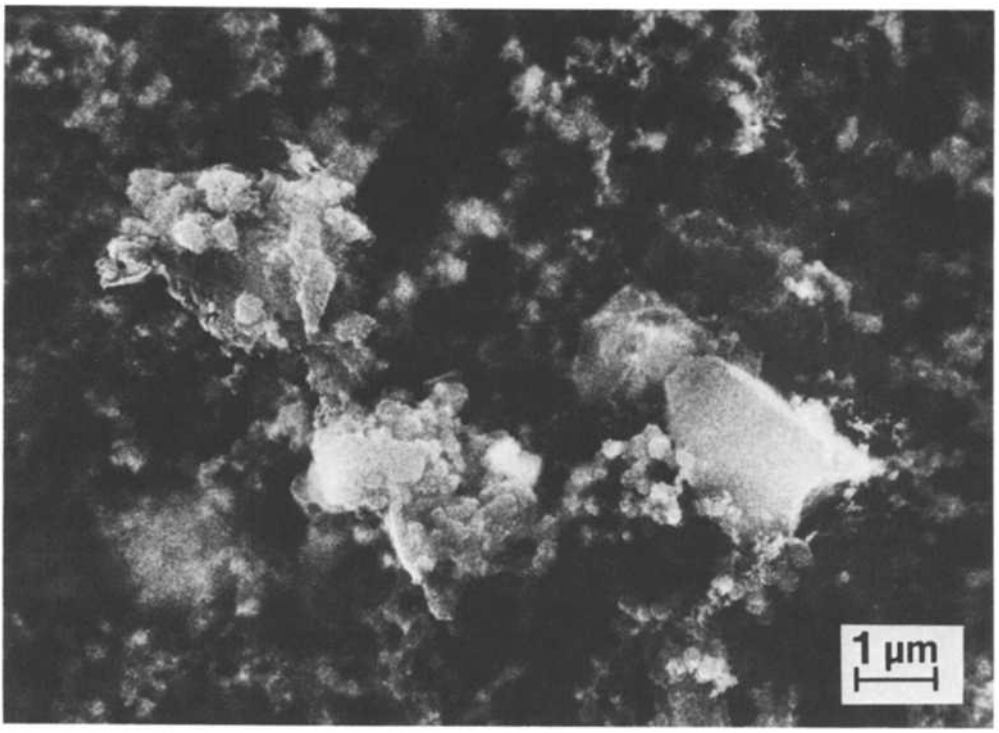

Fig. 1. SEM micrograph of a breathing zone air sample

total Cr, while the concentration of the total Cr was 3.6% of the total particulate. The concentration of Ni in the breathing zone was 3.3%. In the stationary air samples, the concentration of Cr was 3.4%, and that of Ni was 3.7%. A scanning electron micrograph of a breathing zone air sample is presented in Fig. 1. Both welding fume and grinding dust particles can be seen.

The results of the measurements of Cr and Ni in air and in the fluids of the welders and 3 controls are presented in Table 2. The air samples were taken from the breathing zone, unless otherwise stated. In the subjects the mean concentration of Cr was 0.24 µmol/l in whole blood and 0.19 µmol/l in plasma. The lowest concentrations of Cr observed both in whole blood (0.05 µmol/l) and in plasma (0.02 µmol/l) were the same as those of nonexposed persons, while the highest concentrations of Cr were 1.35 µmol/l in whole blood and 0.82 µmol/l in plasma. Thus 57% of the total Cr in whole blood was bound to cells and 43% was in the plasma. The Ni concentration in whole blood hardly differed from the control values.

In urine the mean concentration of Cr was 0.51 µmol/l when corrected to specific gravity or 37.8 µg/g creatinine when corrected to creatinine. The corresponding concentrations of Ni in urine were 0.22 µmol/l when corrected to specific gravity and 11.5 µg/g creatinine when corrected to creatinine. In Table 2 the mean detected concentrations of the metals for before and after the shift are also given.

Table 2. Chromium and nickel concentrations in air and in the whole blood, plasma and urine of the 10 or 5 manual metal arc (MMA) stainless steel (SS) welders and 3 controls. The mean detected concentrations of the metals are given for before (b.s.) and after (a.s.) the shift

Medium		Controls	MMA/SS welders					
		Mean	Mean	σg	Range	b.s.	a.s.	n
Air	Cr (mg/m^3)		0.15	3.5	0.03–0.96			10
	Ni (mg/m^3)		0.15	4.0	0.03–1.78			10
Whole blood	Cr (μmol/l)	< 0.05	0.24	2.7	< 0.05–1.35	0.23	0.25	10
	Ni (μmol/l)	0.018	0.02	1.6	0.01–0.05	0.016	0.020	10
Plasma	Cr (μmol/l)	< 0.02	0.19	2.8	< 0.02–0.82	0.18	0.20	10
Urine	Cr (μg/g creatinine)	< 0.9	37.8	1.6	19.3–67.2	36.3	49.7	5
	Ni (μg/g creatinine)	5.2	11.5	1.6	7.8–26.5	12.0	12.5	5

Table 3. Correlations between the concentrations of chromium and nickel in air and in human fluids of manual arc (MMA) stainless steel (SS) welders. Correlations between the metals and the retention rate of magnetic dust in the lungs (B/t_{ex} are given; (creat—urinary concentration corrected to creatinine; grav—urinary concentration corrected to specific gravity))

In air	In blood	Δ in blood	In plasma	Δ in plasma	In urine		Δ in urine creatinine	B/t_{ex}
					grav	creat		
Cr	0.35	0.89***	0.43	0.83**	0.44	0.91*	0.87	0.92***
Cr(VI)	0.32	0.92***	0.39	0.85**	0.39	0.91*	0.93*	0.89***
Ni	0.56	0	–	–	0.95***	0.97**	0.95**	0.28
Total particulate								
B/t_{ex}	0.31 (Cr)	0.89*** (Cr)	0.36 (Cr)	0.74* (Cr)	0.36 (Cr)			

* = $P < 0.05$; ** = $P < 0.01$; *** = $P < 0.01$

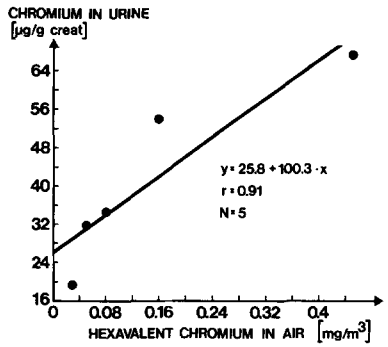

Fig. 2. Relationship between the hexavalent chromium concentration in air and urine (creatinine-corrected)

The concentration of Cr and Ni in the fluids of the welders increases during the workshift. The increase is diminished by the clearance of the metals. By calculating the amount of clearance, the increase in the Cr and Ni concentration can be determined. The mean increase of Cr concentration in whole blood was 12.5%. In plasma it was 21% and in urine 46%. The mean increase of Ni concentration in urine was 11%.

The average decrease of urinary Cr concentration (in percent) between two successive shifts was $20 \pm 9\%$. By applying a single exponential function fit for these two points, the biological half-time of urinary clearance was 2.0 ± 0.5 d. The corresponding biological half-life of Cr was 5.5 ± 1.5 d in blood and 4.0 ± 0.5 d in plasma. The half-life of Ni in urine was 4.5 ± 1.0 d.

The mean remanent magnetic field of the chest area was 1.1 nT ($\sigma g = 2.6$), ranging from 0.26 nT to 3.48 nT, and the retention rate of magnetic dust in the lungs (B/t_{ex}), calculated from of the magnetic signal, was 0.1 nT/year ($\sigma g = 1.8$). The range of the retention rate was 0.07–0.28 nT/year.

The mean concentrations of Cr and Ni in urine (creatinine-corrected) correlated with the concentrations of Cr ($P < 0.05$), Cr (VI) ($P < 0.05$) and Ni ($P < 0.01$) in air (Table 3). In Fig. 2 the regression line between water-soluble Cr (VI) in air and the average creatinine-corrected Cr concentrations in urine is shown. A correlation ($P < 0.05$) was observed between the Cr concentration in the urine spot samples (creatinine-corrected) taken after working hours and the Cr concentration in air.

The best correlations were found between the daily mean increase in the Cr concentration in whole blood and the total Cr ($P < 0.001$), as well as between the former and Cr (VI) ($P < 0.001$) in air. When the concentration of the metals is compared with the similar increase in plasma, the corresponding correlations are not as strong (Table 3).

The concentrations of Ni in air correlated well with the daily mean increase in the Ni concentration in urine during the working hours ($P < 0.01$) and with the urinary concentration of Ni corrected to specific gravity ($P < 0.01$) (Fig. 3) and to creatinine ($P < 0.01$).

The retention rate of magnetic dust in the lungs (B/t_{ex}) correlated with the daily mean increase of Cr in blood surprisingly well ($P < 0.01$). Furthermore very

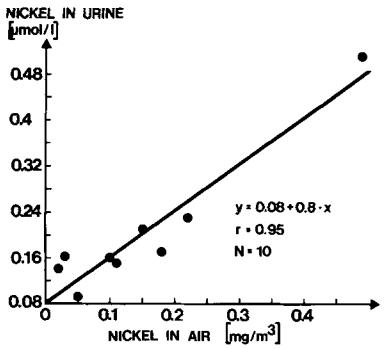

Fig. 3. Relationship between the nickel concentration in air and urine

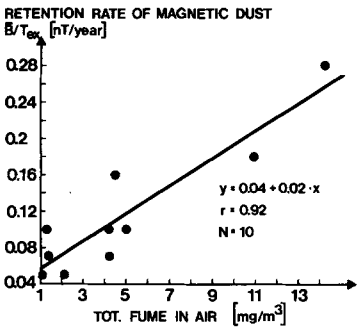

Fig. 4. Relationship between the total particulate content in air and the retention rate of magnetic dust in the lungs

good correlations were found between the retention rate and the personal air samples of Cr and Cr(VI) (Table 3; Fig. 4).

Discussion

In the actual work environment, welders are exposed to a rather complex composition of particulates containing welding fumes and grinding dusts. In the present study, the analysis of the welding fumes showed that only 35% of the total Cr concentration is water-soluble Cr(VI). This is half of the corresponding levels (67–90%) determined by other researchers [Tola et al. 1977; Mutti et al. 1979a; Table 1)]. This low level, as well as the high amount of Ni (3.3%) found in the fumes, is probably due to the especially high Ni concentration of the steel and the type of work. The actual welding time represented about one-third of the working hours. According to the particle morphology, roughly one-third of the particles on filters were slag, metal and abrasive wheel particles (Fig. 1).

Exact comparison of the Cr and Ni concentrations determined in human fluids in one study with the concentrations reported in other investigations is difficult due to the variations in exposure, the different sampling methods, and the difficulties incurred in the analyses of Cr and Ni (Stern and Thomsen 1982). Nevertheless, we have compared the results of our study with those of Tola et al.

Table 4. Comparison of the mean chromium and nickel concentrations determined in air and in blood and urine by various investigators (MMA = manual metal arc; SS = stainless steel)

Study	Studied occupation	Chromium			Nickel	
		Air (mg/m^3)	Whole blood (μmol/l)	Urine (μg/g creatinine)	Air (mg/m^3)	Urine (μmol/l)
Present study	MMA/SS welders	0.1 d	0.33	37.8	0.15	0.22
Tola et al. (1977)	MMA/SS welders	0.1	0.51	48.4	–	–
Mutti et al. (1979a)	MMA/SS welders	0.04	–	24.2	–	–
Kalliomäki P-L et al. (1981)	MMA/SS welders	–	–	18 g/l	–	0.08
Bernacki et al. (1978)	MMA welders[a]	–	–	–	0.006	0.11
Tola et al. (1979)	Ni platers[b]	–	–	–	0.08	0.83
Høgetveit et al. (1980)	Ni refining workers[c]	–	–	–	0.24	0.54

[a] Welded material was high-alloy Ni
[b] Subjects were exposed to aerosols containing $NiSO_4$ and $NiCl_2$ in an electroplating shop
[c] Subjects were exposed to dusts with water-insoluble Ni compounds in roasting-smelting departments in a Ni refining factory
[d] Number of subjects was 5

(1977, 1979), Mutti et al. (1979a), Bernacki et al. (1978) and Høgetveit et al. (1980) with regard to the concentrations of metals detected in blood and urine (Table 4).

Even with the difficulties mentioned, the Cr concentrations of our samples (0.33 μmol Cr/l blood and 37.8 μg Cr/creatinine) seem to agree well with those of Tola et al. (1977) (0.51 μmol Cr/l blood and 48.4 μg Cr/g creatinine) and Mutti et al. (1979a) (24.4 μg Cr/g creatinine). Both had studied MMA/SS welders as we had.

A comparison of the Ni concentrations determined for occupational groups exposed to different Ni compounds is even more difficult due to the different solubility properties of the various Ni compounds. Tola et al. (1979), for instance, studied workers exposed to water-soluble $NiSO_4$ and $NiCl_2$ in an electroplating shop. Kalliomäki et al. (1981b) studied MMA/SS welders. The Ni concentration they reported (0.08 μmol Ni/l urine) was lower than the one we obtained (0.22 μmol Ni/l urine), mainly due to the higher Ni content in the steel welded in the present study.

The same level of exposure to welding fumes has been found to lead to a higher excretion in experimental animals and in MMA/SS welders with an increased body burden of chromium (Franchini et al. 1978; Mutti et al. 1979a, b). When the MMA/SS welders were divided into two groups according to clearance, the slope of the correlation curve was significantly different between the groups (Mutti et al. 1979a). The increased excretion with approximately the same exposure during the follow-up period may be a result of a higher body burden and decreased clearance caused by the kidney damage found in previously exposed workers (Mutti et al. 1979a). Therefore, the effect of the individual body burden upon the excretion rate, and also the blood concentrations of Cr and Ni, obviously complicates the use of urinary excretion for monitoring of Cr exposure among MMA/SS welders.

The determination of the amounts of Cr and Ni excreted in urine during a certain time period before and after work might be adequate to eliminate the effect of body burden. However, this method still requires further research before being put to practical use.

The Cr concentrations in whole blood and plasma did not correlate with the exposure to Cr(VI), but the daily mean increase in the Cr concentration of the blood and plasma reflected the exposure to total Cr and Cr(VI) very well (in blood $P < 0.001$, in plasma $P < 0.01$). The clearance correction used for the daily mean increase in the Cr and Ni concentrations is based on one exponential function model. A multiexponential clearance model could not be applied because the number of points was limited. Nevertheless, the biological half-life of Cr in whole blood (5.5 ± 1.5 d) obtained in this study is about the same as that reported by Kalliomäki et al. (1983) for rats after one-month's inhalation of MMA/SS welding fumes.

The large variation in the Cr concentration detected in the morning urine (0.01–2.7 μmol/l) and blood (0.05–1.43 μmol/l) samples indicates different body burdens of the metals. Perhaps the high correlations between the retention rate of the magnetic dust (B/t_{ex}) and the amount of particulates in air and, on the other hand, between the retention rate and the daily mean increase in the Cr concentration of the blood can partly be explained by fixed work habits of the welders. It

should be emphasized that B/t_{ex} is an index of long-term exposure. Although the magnetic measuring method indicates body burden in vivo, it also seemed to indicate the Cr exposure among the MMA/SS welders in the present study.

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