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Dental amalgam and mercury

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Summary. Mercury concentration in intraoral air and urine of seven females with dental amalgam was measured before and after intake of one hard-boiled egg. A considerable decrease in mercury concentration in intraoral air was found. Twenty women with about equal dental amalgam status, with or without subjective symptoms related to dental amalgam, were also studied. Mercury concentrations in intraoral air and urine were measured. For all the 27 women the basal intraoral air concentration of mercury ranged over 0.6–10.4 μg/m^3 (median value 4.3 μg/m^3). This corresponds to a release of 0.02–0.38 ng/s (median value 0.16 ng/s). In urine, the mercury concentration varied from <0.8–6.9 μg/g creatinine (median value 1.9 μg/g creatinine). Data from both parameters were significantly correlated to the total number of teeth areas with dental amalgam. Protein values in urine indicated no renal damage. Maximum concentrations of mercury vapour in intraoral air for the 27 women who had chewed chewing gum for 5 min varied between 2–60 μg Hg/m^3 (median value 19 μg Hg/m^3). This corresponds to 0.07–2.20 ng Hg/s and a median value of 0.70 ng Hg/s.

Key words: Dental amalgam fillings — Mercury — Intraoral air — Kidney — Urine

Introduction

Dental amalgam has been used for more than 100 years and is still the most frequently used material for dental restoration. The alloy consists of mercury, silver and tin (50%, 35% and 15%, respectively). During the last decade silver has been partly replaced by copper in some modern amalgams. Whether mercury released from amalgam fillings may cause adverse health effects or not has been widely discussed during the last century (Stock 1939; Frykholm 1957; Nylander et al. 1987; Clarkson et al. 1988). A possible influence of the intake of certain foodstuffs on the release of mercury from amalgam fillings into intraoral air has been suspected. The possible influence was tested using intake of hard-boiled egg, Swedish crispbread, apple, orange, orange juice, soft drinks, coffee and tea with and without sugar, and hot flavoured food. The results indicated that hard-boiled egg had a marked influence on the mercury concentration in intraoral air. The aim of this study was to determine whether or not hard-boiled eggs would decrease mercury concentration in intraoral air. Mercury levels in urine were also determined.

A second study was performed to compare mercury concentrations in intraoral air and urine and total protein in urine in two groups of women (cases and controls) with comparable dental amalgam status. The cases were all individuals who complained of subjective symptoms related to their dental amalgam. They had contacted their dentist for treatment.

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Materials and methods

Experimental groups. Study I consisted of seven healthy women, 43–60 years of age, with dental amalgam fillings. Each subject had a total number of teeth areas with amalgam ranging over 33–53 ($\bar{x}=39$; SD = 6.9). In order to ensure minimal release of mercury from the amalgam fillings, the women were not allowed to eat breakfast, brush their teeth or smoke before basal mercury levels were determined on the same morning when mercury concentrations in intraoral air were to be mea-

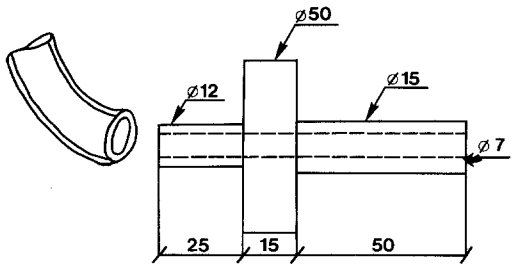

Fig. 1. Special tube in plexiglass for measurements of intraoral mercury vapour. Dimensions are given in mm

sured. Concentrations of mercury were measured after allowing subjects to chew sugar-free chewing gum (V6; Fertin Laboratories A/S Vejle, Denmark) for 5 min. The mercury concentration was registered with the mouth closed and nose-breathing. This procedure was followed by intake of one hard-boiled egg. Mercury levels were registered immediately afterwards and after 1, 2, and 3 h without any intake of food or drink. Study II consisted of two groups (cases and controls) with 10 women in each group. All the women had a comparable dental amalgam status and age. For the control group, the number of teeth areas with amalgam ranged over 37–53 ($\bar{x}=43$; SD = 5.5) and ages ranged over 32–45. The cases were 31–58 years of age with a total number of teeth areas containing amalgam ranging over 24–63 ($\bar{x}=43$; SD = 14.0). They considered themselves to have subjective symptoms that were related to their amalgam fillings. One person with no amalgam fillings served as a blank. For study II basal and stimulated mercury levels (no egg intake) were obtained as described for study I.

Urine analysis. Urine was collected in plastic bottles over a period of 24 h and kept at +4° C until required for further analysis. Amidosulphonic acid (1 mg/ml) was added to subsamples of 100 ml urine and kept frozen until mercury was analysed according to the method of Einarsson et al. (1983). A Goldberg refractometer was used to determine specific gravity (density). Urine was analysed for pH, protein, glucose and blood content with Hema-Combistix^R. Creatinine was determined according to Hare (1950) with picric acid. Protein determination was performed with a biuret method (Piscator 1962) using albumin as standard. Agarose electrophoresis was used for studies of protein in urine (Mikyaka et al. 1977). For protein staining, amido-black was used.

Mercury in intraoral air. A special tube (Fig. 1) was constructed to standardize air sampling from each participant in the studies. The end plate of the tube rests against the lips, while the other end of the tube is about 4 cm inside the oral cavity, i.e. at about the centre of the oral cavity. The tube is located so that the orifice cannot come into contact with the tongue, palate or teeth. After the saliva is swallowed, the air is drawn for 1 min via the nose through the oral cavity into the tube and from there to the measuring instrument. Intraoral air was analysed for mercury concentration by cold vapour atomic absorption spectrophotometry (CVAAS). Released mercury vapour passes into a measurement cell where absorption of mercury light takes place at 253.7 nm (Magos 1971). The method for evaluating the results was modified, due to the different shape of the recorder signal between intraoral measurements and standards. Hence, a digital planimeter (Placom KP-90, Koizumi) was used to measure the total area of all the signals. The amount of mercury vapour in intraoral air during the collection time could then be calculated. Mercury vapour generated in 1 min, and expressed as mass (ng) Hg, was recalculated and expressed as μg Hg/m^3 using an air flow rate of 2.2 L/min. Standards were prepared from mercury chloride ($HgCl_2$) of 25, 50, 80, 100, 150, 200, and 300 ng Hg and were analysed both before and after intraoral investigations.

Saliva. Rate of secretion, and buffering capacity of the saliva were registered with Dentobuff ^{R} (Orion Diagnostica).

Table 1. Results for study I

FP	Dental status		Intraoral air (mean value $n=4$ for K–S)									Urine	
	B	C	K	L	N	O	P	Q	R	S	T	X	Z
1	53	12	5.8	21.6	1.2	2.7	2.6	3.3	1.9	0.4[a]	11.9	4.5	117
2	43	13	2.6	25.9	1.0	2.1	2.1	—	5.3	2.0	17.0	5.1	46
3	33	9	0.8	6.2	0.2[a]	0.7	—	—	0.3[a]	0.2[a]	2.9	2.3	59
4	35	9	0.8	10.0	0.4	0.2[a]	0.4	—	1.0	0.2[a]	4.7	1.5	49
5	35	13	1.8	3.7	1.0	1.1	—	—	2.4	0.9	2.2	0.9	28
6	37	10	1.6	1.4[b]	0.2[a]	0.2[a]	0.2[a]	0.4	0.4	0.2[a]	1.5[b]	<0.8	74
7	40	13	4.8	16.4	0.6	0.8	0.7	0.7	1.3	0.5	10.8	2.2	59
$\bar{x}$	39	11	2.6	12.2	0.7	1.1	1.2	1.5	1.8	0.6	7.3	—	62
SD	6.9	1.9	1.9	9.3	0.4	0.9	1.1	1.6	1.7	0.7	6.0	—	28
Median	37	12	1.8	10.0	0.6	0.8	0.7	0.7	1.3	0.4	4.7	2.2	59
min	33	9	0.8	1.4	0.2	0.2	0.2	0.4	0.3	0.2	1.5	<0.8	28
max	53	13	5.8	25.9	1.2	2.7	2.6	3.3	5.3	2.0	17.0	5.1	117

FP, number of person; B, number of teeth areas with amalgam; C, number of occlusal areas with amalgam; K, basal value (μg Hg/m^3), day 1; L, stimulated value (μg Hg/m^3); N, stimulated value after intake of one egg (μg Hg/m^3); O–Q, stimulated value 1, 2, 3 h after intake of one egg; R, basal value (μg Hg/m^3); S, basal value after intake of one egg (μg Hg/m^3); T, estimated 24-h uptake (μg Hg); X, urine concentration (μg Hg/g creatinine); Z, total protein in urine (mg/g creatinine)

[a] Contains one or several values under detection limit (0.2 μg/m^3)

[b] Temporarily reduced chewing function due to ongoing dental treatment

Dental status and amalgam registration. Each participant's dental and amalgam status was carefully investigated by a dentist. Alginate impressions were taken and models were prepared of each cheek. No functional abnormalities were observed. Amalgam load for all participants was in the range frequently found in Swedish females in these age groups. In study I, occlusal areas with dental amalgam were measured by using digital vernier callipers.

Questionnaire. Each participant in study II received a questionnaire concerning health status, medicine, selenium, vitamin, fish intake, alcohol consumption, and smoking habits.

Estimation of 24-h uptake of mercury vapour released from amalgam fillings. A method described by a Swedish Expert Committee (SOS 1987) was used. It is assumed that the subject's exposure to mercury vapour from amalgam fillings is 4 h/day, i.e. at maximum stimulation during chewing with only mouth breathing. After chewing for 5 min about two-thirds of the 'steady-state' value was reached (Vimy and Lorscheider 1985). Hence, maximum stimulated values recorded were increased accordingly before calculating with the equation: stimulated release = release rate (ng Hg/min) × time (240 min) × 0.5 × 0.8, since it is assumed that 50% of the mercury vapour is inhaled and that 80% of this is absorbed. It is assumed that there is a basal release of mercury vapour during the remaining 20 h. As studies of individual breathing habits indicate high percentages of nasal breathing during rest and sleep (Niinima et al. 1981; Camner and Bakke 1980), it is also assumed that 25% of this release of Hg is inhaled: basal release = release rate (ng Hg/min) × time (1200 min) × 0.25 × 0.8.

Results

The results for study I are presented in Table 1; for study II, results for controls are given in Table 2 and for cases in Table 3. In study I, urinary mercury concentrations varied between <0.8–5.1 μg Hg/g creatinine, with a median value of 2.2 μg Hg/g creatinine. In study II, controls showed levels <0.9–5.8 μg Hg/g creatinine, with a median value of 1.7 μg Hg/g creatinine. For cases in study II levels were <1.2–6.9 μg Hg/g creatinine with a median value of 2.2 μg Hg/g creatinine. In Sweden, for non-occupationally exposed persons, urine values up to approximately 5 μg Hg/g creatinine equivalent to either 5 μg Hg/l or 25 nmol Hg/l or 0.25 nmol Hg/mol creatinine are considered normal. Only one slightly elevated value of 6.9 μg Hg/g creatinine was found. This case was the only person who chewed chewing gum several times a day.

In the present study, mercury excretion in urine could not be related to chewing technique, nor to grinding and/or pressure of teeth, smoking, or selenium intake. Analysis of protein in urine showed no pathological pattern, either in total-protein analyses or using the agarose electrophoretic analyses.

Saliva

Data obtained using a dentobuff test were not adequate as this method was not sensitive enough.

Table 2. Results for study II, controls

FP	Dental status		Intraoral air[a]				Urine	
	B	C	K	L	M	T	X	Z
1	45	15	1.5	4.2	7.7	3.1	1.3	60
2	49	14	7.6	24.1	27.7	13.4	5.8	49
3	38	17	4.9	23.3	22.6	21.5	1.6	26
4	37	14	6.9	14.2	15.5	9.8	3.6	80
5	53	13	4.2	23.1	46.2	12.5	4.5	45
6	40	12	8.5	24.1	40.8	17.4	2.0	53
7	40	15	0.6	3.3	5.6	2.0	1.0	32
8	45	14	8.0	9.5	11.3	8.0	1.3	71
9	37	14	4.3	10.3	16.4	6.8	1.7	66
10	47	17	1.2	2.6	2.9	1.5	<0.9	50
$\bar{x}$	43	15	4.8	13.9	19.7	9.6	—	53
SD	5.5	1.6	2.9	9.1	14.7	6.7	—	17
Median	43	14	4.2	12.2	16.0	8.9	1.7	51
min	37	12	0.6	2.6	2.9	1.5	<0.9	26
max	53	17	8.5	24.1	46.2	21.5	5.8	80

FP, number of person; B, number of teeth areas with amalgam; C, number of occlusal areas with amalgam; K, basal value (μg Hg/m^3); L, stimulated value (μg Hg/m^3); M, stimulated value II (μg Hg/m^3); T, estimated 24-h uptake (μg Hg); X, urinary concentration (μg Hg/g creatinine); Z, total protein in urine (mg/g creatinine)

[a] Detection limit (0.3 μg/m^3), mean value column K ($n=6$), column L ($n=4$), and column M ($n=2$)

Table 3. Results for study II, cases

FP	Dental status		Intraoral air[a]				Urine	
	B	C	K	L	M	T	X	Z
1	47	14	2.5	4.9	7.4	3.8	<1.3	37
2	61	14	10.4	30.6	38.0	17.0	5.6	104
3	43	14	5.8	16.3	30.4	10.5	6.9	85
4	51	12	2.7	6.9	20.3	5.2	<1.9	34
5	63	15	6.3	19.0	21.5	12.4	2.7	—
6	34	10	6.0	16.3	31.4	9.8	2.4	77
7	46	11	5.9	12.3	19.3	8.1	4.4	38
8	36	12	9.1	26.0	36.8	18.6	<1.9	51
9	24	7	2.1	6.3	13.4	4.0	<1.2	63
10	25	10	0.9	17.9	26.5	9.1	<1.7	54
$\bar{x}$	43	12	5.2	15.7	24.5	9.9	—	60
SD	14	2.5	3.1	8.4	10.0	5.1	—	24
Median	45	12	5.9	16.3	24.0	9.5	2.2	54
min	24	7	0.9	4.9	7.4	3.8	<1.2	34
max	63	15	10.4	30.6	38.0	18.6	6.9	104

FP, number of person; B, number of teeth areas with amalgam; C, number of occlusal areas with amalgam; K, basal value (μg Hg/m^3); L, stimulated value I (μg Hg/m^3); M, stimulated value II (μg Hg/m^3); T, estimated 24-h uptake (μg Hg); X, urinary concentration (μg Hg/g creatinine); Z, total protein in urine (mg/g creatinine)

[a] Detection limit (0.3 μg/m^3), mean value column K ($n=6$), column L ($n=4$), and column M ($n=2$)

Mercury vapour concentration in intraoral air

For the calibration between known amounts (ng Hg) of standards added and the areas (cm^2) of the recorder signals at 5 mV range and a chart speed of 60 mm/min the following equations were obtained:

Study I: area (cm^2) = 0.88 × Hg (ng) + 0.88

Study II: area (cm^2) = 0.87 × Hg (ng) + 0.46

The detection limit was calculated to be 0.2 μg/m^3 for study I and 0.3 μg/m^3 for study II. All signals were below detection limit for the amalgam-free person (blank).

In study I, the basal mercury vapour concentration (individual mean) in intraoral air was measured on two different days. Interindividual (mean = 2.2, range = 0.6–4.0 μg Hg/m^3) as well as intraindividual variations (day 1, mean = 2.6; day 2, mean = 1.8 μg Hg/m^3) could be detected.

The stimulated values (range 4–50 μg Hg/m^3 for maximum values) found after subjects had chewed chewing gum for 5 min were elevated in all subjects compared to the basal values. However, the intraoral mercury concentration (mean = 0.7 μg Hg/m^3, SD = 0.4 μg Hg/m^3) decreased in all subjects to a level below the basal level after intake of one hard-boiled egg. This happens even when stimulation occurred immediately before intake (Fig. 2). The influence of the egg intake persisted up to 3 h later.

In study II, the basal values (individual mean) varied over 0.6–8.5 μg Hg/m^3 with a mean of 4.8 μg/m^3 for the control group, and ranged over 0.9–10.4 μg Hg/m^3 with a mean of 5.2 μg/m^3 for the cases.

No significant ($P=0.79$, $n=20$ Mann Withney test) difference between the group means could be found. After stimulation, the intraoral Hg concentrations increased for both cases and controls

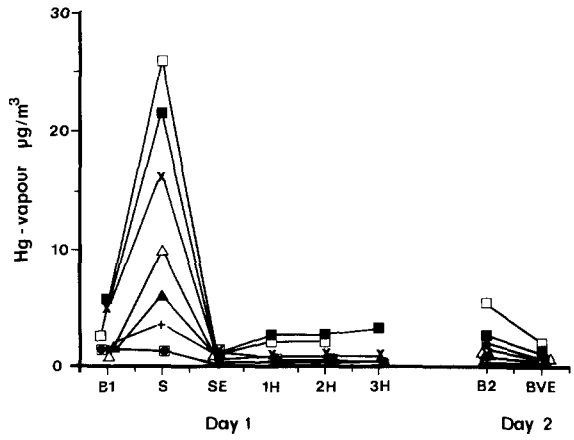

Fig. 2. Concentration of mercury vapour in intraoral air (μg Hg/m^3) plotted for individual persons ($n=7$). Bl = basal value day 1; S = stimulated value; SE = stimulated value immediately after intake of one egg; 1H, 2H and 3H = concentration after 1 h, 2 h and 3 h; B2 = basal value day 2; BVE = basal value after intake of one egg

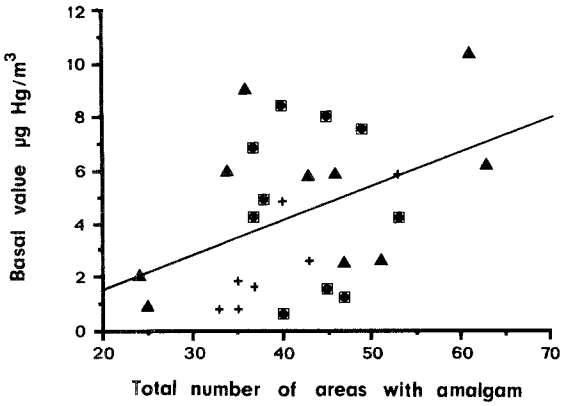

Fig. 3. Basal concentration of mercury vapour in intraoral air as µg Hg/m^3, plotted against total number of teeth areas with dental amalgam. $y=-1.0764+0.129x$; $n=27$; $r=0.41$; $0.05>P>0.02$, 95% confidence interval regression coefficient (k) $0.01<k<0.25$. (+) Study I; (■) study II, control; (▲) study II, cases

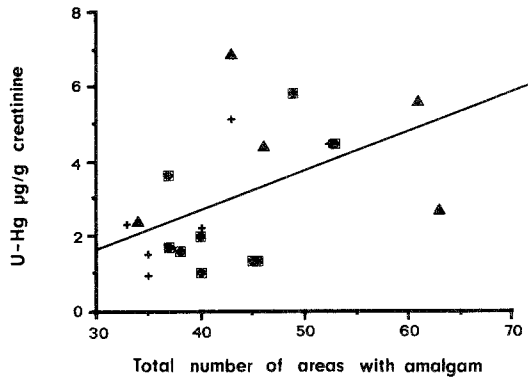

Fig. 4. Mercury in urine (U-Hg), expressed as µg Hg/g creatinine, plotted as a function of total number of teeth areas with dental amalgan. $y=-1.5219+0.1054x$; $n=20$; $r=0.50$; $0.05>P>0.02$, 95% confidence interval regression coefficient (k) $0.01<k<0.20$. (+) study I; (■) study II, control; (▲) study II, cases

compared to the basal level. The maximum values were between 3–60 µg Hg/m^3 for the control group and between 8–45 µg Hg/m^3 for the cases. No difference was found among the groups.

Mercury uptake estimated from intraoral air

For study I ($n=7$) an uptake of 1.5–17.0 µg Hg ($\bar{x}=7.3$ µg Hg; SD 6.0 µg Hg) was calculated. For study II, the control group ($n=10$) values were 1.5–21.5 µg Hg ($\bar{x}=9.6$ µg Hg; SD 6.7 µg Hg). In the case group ($n=10$) similar values were 3.8–18.6 µg Hg ($\bar{x}=9.9$ µg Hg; SD 5.1 µg Hg).

All subjects, from both studies, had comparable dental amalgam status and were pooled in order to obtain a larger group with a total of 24–63 teeth areas with dental amalgam. Pooled plottings, statistical tests (Student's t-test; 5% significance level, two-tailed test) and regression analyses were performed with the data obtained.

There was a statistically significant ($0.05>P>0.02$, $n=27$) correlation between the total number of areas with amalgam and the basal intraoral mercury-vapour concentration (Fig. 3). However, the estimated 24-h uptake of mercury from dental amalgam was not significantly related to the total number of teeth areas with amalgam ($0.5>P>0.1$, $n=27$).

A significant correlation ($0.05>P>0.02$, $n=20$) for mercury in urine (µg Hg/g creatinine) as a function of the number of teeth areas with

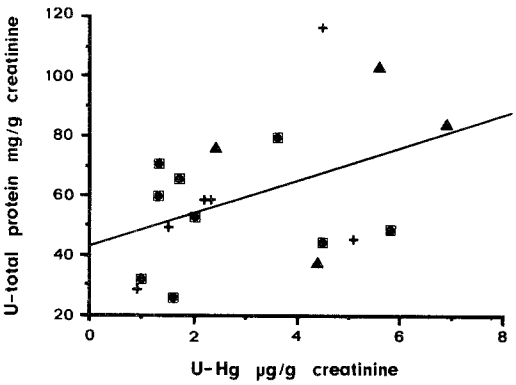

Fig. 5. Total protein concentration, expressed as mg/g creatinine, in urine plotted as a function of mercury concentration, expressed as µg Hg/g creatinine in urine. $y=42.9805+5.865x$; $n=19$; $r=0.43$; $0.1>P>0.05$, 95% confidence interval regression coefficient (k) $-0.45<k<11.62$. (+) study I; (■) study II, control; (▲) study II, cases

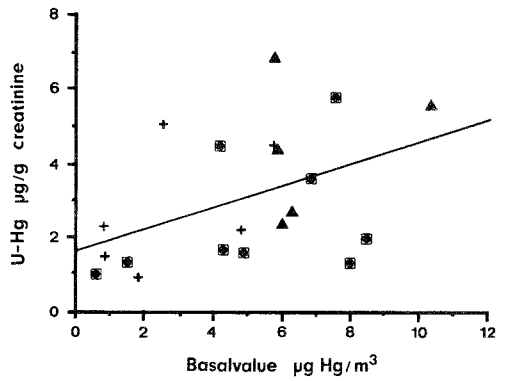

Fig. 6. Mercury concentration in urine (U-Hg), expressed as µg Hg/g creatinine, is plotted as a function of basal value of mercury in intraoral air (µg Hg/m^3). $y=1.6189+0.2966x$; $n=20$; $r=0.45$; $0.05>P>0.02$, 95% confidence interval regression coefficient (k) $0.01<k<0.58$. (+) Study I; (■) study II, control; (▲) study II, cases

amalgam is shown in Fig. 4. Values below detection limit were excluded. This relationship became slightly stronger when the mercury in urine concentration was expressed as μg Hg/L adjusted for group mean specific gravity (1.016 g/ml). However, the correlation was weaker and non-significant when the mercury content was given as excreted mass (μg)/24 h.

No renal effects were found in either of the groups when determined from total amount of protein excreted in the urine. The total amount of protein in urine (mg protein/g creatinine) is shown in Fig. 5. No significant correlation was found when protein was plotted as a function of urinary mercury (μg Hg/g creatinine). Values below detection limit were excluded. However, a significant ($0.05 > P > 0.02$, $n = 20$) correlation was found (Fig. 6) between μg Hg/g creatinine in urine and the basal values of μg Hg/m^3 in intraoral air. Urinary mercury values below detection limit were not included.

Discussion

Differences in the effects of mouth and nose breathing are not discussed in this article. Air from the oral cavity was collected with closed mouth only. After intake of one hard-boiled egg, a lower concentration of mercury was found in the intraoral air. This might be explained by the sulphur content of the egg. Mercury has a very high affinity for sulphur. Furthermore, a layer of mercury sulphide might be formed on the amalgam surface and consequently result in a decrease in the release of mercury vapour.

No major differences in concentration of mercury in intraoral air and in urine were obtained for the groups in study II. Therefore, subjective symptoms related to amalgam in dental fillings for these women could not be explained by these results. However, the uptake rate to the central nervous system or sensitivity to mercury may differ for different individuals and mercury-related symptoms cannot therefore be excluded.

For all the 27 women the basal intraoral air concentration of mercury ranged over 0.6–10.4 μg Hg/m^3 (median values 4.3 μg Hg/m^3). This corresponds to mercury release of 0.02–0.38 ng Hg/s (median value 0.16 ng Hg/s) which coincides with recently published data (Berglund et al. 1988).

No damage in renal function was indicated as the total excretion of protein is within the normal range found. Agarose electrophoretic pattern was normal.

More detailed information is given in a Swedish Master Thesis in Toxicology at Karolinska Institute (Aronsson 1988). This investigation was carried out on a rather small population. A larger group needs to be studied before any firm conclusions can be drawn.

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