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Transfer of Cadmium from a Sandy Acidic Soil to Man: A Population Study[1]

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This population study included 230 subjects (age range 20–83 years) who consumed vegetables grown in kitchen gardens on a sandy acidic soil (mean pH ~6.3). The study investigated the association between the Cd (cadmium) levels in blood and urine and the Cd concentration in the soil (range 0.2–44 ppm). Seventy-six subjects were current smokers and 122 participants lived in a district with known Cd pollution. Urinary Cd in the 230 subjects averaged 8.7 nmole/24 hr, (range 1.3 to 47 nmole/24 hr) after age adjustment positively correlated with the Cd level in the soil; a twofold increase of the Cd concentration in the soil was accompanied by a 7% rise in urinary Cd in men ($R^2 = 0.05$; $P = 0.04$) and by a 4% rise in women ($R^2 = 0.02$; $P = 0.05$). Blood Cd averaged 11.5 nmole/liter (range 1.8–41 nmole/liter) and was negatively associated with the Cd level in the soil. After adjustment for significant covariates (smoking and serum γ-glutamyl transpeptidase in both sexes, and age and serum ferritin in women), a twofold increase in the Cd concentration in the soil was accompanied by a 6% decrease in blood Cd in men ($R^2 = 0.03$; $P = 0.09$) and by a 10% decrease in women ($R^2 = 0.06$; $P < 0.01$). In conclusion, in a rural population, consuming vegetables grown on a sandy acidic soil, 2 to 4% of the variance of urinary Cd was directly related to the Cd level in the soil. The negative correlation with blood Cd, a measure of more recent exposure, was biased by the implementation of preventive measures in the polluted district. © 1992 Academic Press, Inc.

INTRODUCTION

Cadmium is an occupational and environmental contaminant that after absorption is efficiently retained by the human body and therefore accumulates throughout life (Bernard and Lauwerys, 1984; Elinder, 1985). The main toxic effects ensuing from long-term exposure to cadmium are renal tubular dysfunction

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(Buchet *et al.,* 1990; Lauwerys *et al.,* 1990) and disturbances of the calcium homeostasis (Kjellström, 1985; Staessen *et al.,* 1991) and bone metabolism (Kjellström, 1985). These effects, which have potentially important implications in terms of public health, have been observed in a random sample of the population (Buchet *et al.,* 1990; Lauwerys *et al.,* 1990; Staessen *et al.,* 1991).

Inhalation of tobacco smoke and the consumption of contaminated foodstuffs constitute the two main sources of cadmium for subjects not exposed at work (Bernard and Lauwerys, 1984; Elinder, 1985). When grown on cadmium-polluted soils, some vegetables amass a considerable amount of cadmium and may represent an important source of dietary cadmium (Elinder, 1985). Although the various pathways of cadmium transfer to the human body are well understood from an operational point of view, their quantitative importance at the population level remains largely unknown.

The present study investigated whether, in the population at large, a positive correlation could be demonstrated between the cadmium concentration in the cultivated soil and the amount of cadmium in the body. The internal dose of cadmium was estimated from the blood cadmium concentration, which is thought to mirror mainly current intake (Lauwerys, 1983) and by measuring the urinary excretion, reflecting lifetime exposure (Lauwerys, 1983). The study was conducted in Belgium, one of the principal cadmium producers in Europe (Lauwerys *et al.,* 1990).

METHODS

Subjects

As described in detail elsewhere (Lauwerys *et al.,* 1990), the CADMIBEL Study (Buchet *et al.,* 1990; Lauwerys *et al.,* 1990; Staessen *et al.,* 1991) was conducted from 1985 to 1989 in four Belgian districts, selected to provide a wide range of environmental cadmium exposure. A random sample of the households was identified in each district with the goal of recruiting an equal number of subjects in six subgroups by sex and age (20–39, 40–59, ≥60 years). All household members with a minimum age of 20 years were invited to participate, provided that the quota of the particular sex–age group to which they belonged had not yet been satisfied. Subjects who had taken part in the CADMIBEL Study were eligible for the present analysis (1) if they were living in a rural area, (2) if they belonged to a family owning a kitchen garden, whereby a family was defined as all subjects living on the same address, and (3) if they had not been exposed to heavy metals at work.

The rural populations surveyed in the CADMIBEL Study lived either in a polluted district (Noorderkempen) or in an area with no known source of cadmium pollution (Hechtel-Eksel). The soil in both areas has a sandy composition with 2 to 3% organic carbon up to a depth of 25 cm (Scokart *et al.,* 1983). From 1984 to 1987 the 95th percentile of the airborne cadmium concentration in Noorderkempen averaged 0.36 nmole/m^3 (40 ng/m^3), whereas in nonpolluted Belgian districts it never exceeded 0.09 nmole/m^3 (10 ng/m^3); from 1981 to 1986 the cadmium concentration in the grass ranged from 1 to 258 μmole/kg dry weight (DW) (0.13 to 29 mg/kg DW) in Noorderkempen, whereas in the nonpolluted district it was not higher than 2 μmole/kg DW (0.2 mg/kg DW) (Lauwerys *et al.*, 1990). The cadmium concentration in the water sampled from 2410 wells in 1983 and 1984 in the polluted rural area ranged from 0.4 to 3559 nmole/liter (0.05 to 400 μg/liter) with a median of 38 nmole/liter (4.3 μg/liter) and a 90th percentile of 222 nmole/liter (25 μg/liter) (Lauwerys *et al.*, 1990).

In an attempt to make an inventory of the cadmium concentration in the soil, LISEC (Research Institute for Ecology and Forestry, B-3600 Genk, Belgium) invited the inhabitants of the Northern part of the Province of Limburg to have the cadmium concentration in the soil of their garden measured. In the polluted district, gardens were only considered for sampling if they were located in an area where, on the basis of a preliminary screen, the cadmium concentration in the soil was suspected to be at least 3 ppm. Soil specimens obtained on a total of 2600 different locations had been measured in 1986. This list of 2600 addresses was consulted, and participants of the CADMIBEL Study, whose kitchen garden had been sampled, were identified for inclusion into the present analysis.

Field Work

Each household was visited several times to obtain all necessary data (Lauwerys *et al.*, 1990). Body weight and height were measured. The participants were asked to complete a self-administered questionnaire inquiring into their medical history, current and past occupations, smoking habits, consumption of alcohol, dietary habits, and intake of medications. They were also asked to collect a 24-hr urine sample in a wide-neck metal-free polyethylene container, after having been instructed how to avoid external contamination of the urine with cadmium. On a separate occasion, but usually within 2 weeks after the urine collection, a physician or nurse visited the households to withdraw 20 ml of venous blood.

Biochemical Measurements

Serum was analyzed for the activity of the enzyme γ-glutamyl transpeptidase (Persyn and van der Silk, 1976) and for its ferritin concentration (Bernard and Lauwerys, 1983). Blood and urinary cadmium was measured by electrothermal atomic absorption spectrometry with the use of a stabilized temperature platform furnace and Zeeman background correction (Lauwerys *et al.*, 1990).

The soil samples were dried for 24 hr at 60°C. The dried samples were sieved on a 2-mm sift and, after a wet destruction with aqua regia, were analyzed for cadmium by atomic absorption spectrometry.

Statistical Analysis

For database management and statistical analysis the Statistical Analysis System (SAS) (SAS Institute, Inc., 1987) was used. The distributions of blood and urinary cadmium, serum γ-glutamyl transpeptidase, serum ferritin, and cadmium in the soil were normalized by a logarithmic transformation; for these measurements the geometric mean and range are reported.

Statistical methods included Student's t test and linear regression analysis. The determinants of blood and urinary cadmium were identified by a stepwise regression procedure, terminating when all regression coefficients in the model were significant at the 5% probability level. Gender, age (linear and quadratic term), smoking, the number of cigarettes smoked per day, serum γ-glutamyl transpeptidase (as an index of alcohol consumption), serum ferritin, social class (coded 1 to 3 for low, middle, and high social class), the use of water from a private well, the cadmium concentration in the soil, and the pH of the soil were considered for entry into the regression model.

RESULTS

Characteristics of the Subjects

In the two rural areas a total of 993 subjects, living in 469 homes, had taken part in the CADMIBEL Study (participation rate 78%). Exposure to cadmium at work was reported by 195 participants. Of the remaining 798 subjects, 230 were eligible for the present analysis because they owned a garden that had been sampled for the determination of cadmium in the soil. One hundred and eight subjects lived in the control district, and 122 lived in the polluted area.

The main characteristics of the men and women included in the study are presented in Table 1. Their ages ranged from 20 to 83 years. The urinary cadmium excretion in all subjects combined averaged 8.7 nmole/24 hr (geometric mean), ranging from 1.3 to 47.1 nmole/24 hr, and the geometric mean blood cadmium concentration was 11.5 nmole/liter (range 1.8–40.9 nmole/liter). All but 6 subjects (97%) reported consumption of homegrown vegetables for a median duration of 30 years (range 1–79 years). Forty-seven subjects (20%) used water from a well for drinking or cooking. Seventy-six subjects (33%) were smokers (median 16 cigarettes/day), and 32 (14%) reported daily intake of alcohol (median 20 g/day).

Cadmium in the Soil

In the nonpolluted area (63 homes) the cadmium concentration of the soil averaged 1.2 ppm (range 0.2–5.5 ppm) and the pH 6.0 (range 4.6–7.4). This was

TABLE 1
CHARACTERISTICS OF THE SUBJECTS

	Men	Women
Number	83	147
Age (years)	46 ± 16	46 ± 16
Body mass index (kg/m^2)	25.2 ± 3.3	25.9 ± 5.1
Blood cadmium (nmole/liter)	10.9 (1.8–40.0)	12.0 (1.8–40.9)
Serum ferritin (μg/liter)	186 (38–860)	81 (9–880)
γ-glutamyl transpeptidase (U/liter)	15 (5–151)	9 (2–117)
Urinary cadmium (nmole/24 hr)	9.3 (1.3–33.9)	8.4 (1.5–47.1)

Note. Values are mean ± standard deviation or geometric means with range between parentheses.

significantly ($P < 0.001$) less than in the polluted district (93 homes), where these values were 7.4 ppm (range 1.3–44.0 ppm) and 6.5 (range 5.5–7.7), respectively.

Cadmium in the Soil as a Determinant of Blood Cadmium

Blood cadmium was lower in the polluted than in the nonpolluted district (9.5 versus 15.4 nmole/liter; $P < 0.001$). In the 230 subjects combined, blood cadmium was negatively associated with the cadmium concentration in the soil. The single correlation coefficient was -0.33 ($P = 0.004$) in men and -0.34 ($P < 0.001$) in women. The partial correlations after adjustment for age were -0.32 ($P < 0.01$) and -0.30 ($P < 0.001$), respectively (Fig. 1).

Stepwise multiple regression analysis showed that blood cadmium in men was positively correlated with smoking (partial $R^2 = 0.21$; $P < 0.001$) and with log γ-glutamyl transpeptidase ($R^2 = 0.05$; $P < 0.04$). With these variables in the equation, the *t*-to-enter for the cadmium concentration in the soil was -1.71 ($R^2 = 0.03$; $P = 0.09$). In women blood cadmium increased with smoking ($R^2 = 0.16$; $P < 0.001$), age ($R^2 = 0.14$; $P < 0.001$), and γ-glutamyl transpeptidase ($R^2 = 0.03$; $P = 0.02$), but there was a negative correlation with the cadmium concentration in the soil ($R^2 = 0.06$; $P < 0.01$) and with serum ferritin ($R^2 = 0.03$; $P = 0.01$).

The regression equation in men was: log blood cadmium (nmole/liter) = 0.7711 + 0.2019 smoking (coded 0 for nonsmokers and 1 for smokers) + 0.2040 log γ-glutamyl transpeptidase (U/liter) − 0.0881 log cadmium concentration in the soil (ppm) ($R^2 = 0.29$). In women the equation was: log blood cadmium (nmole/

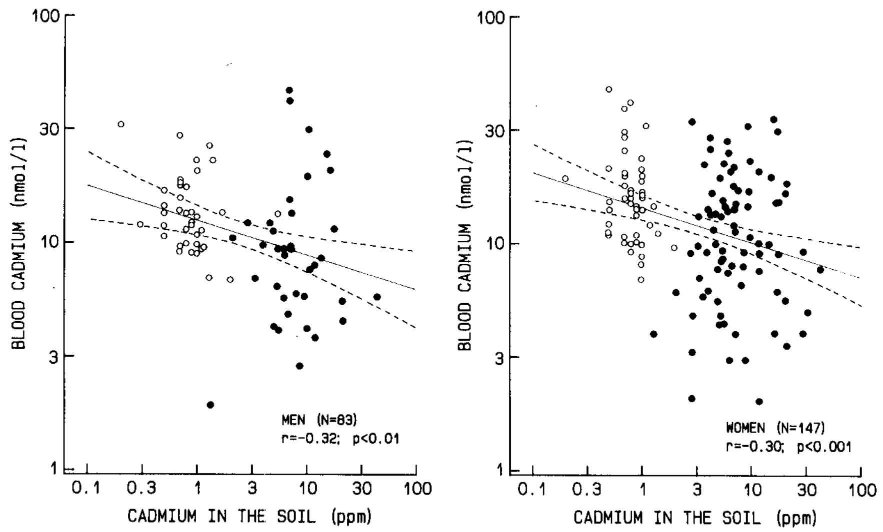

FIG. 1. Scatterplot of blood cadmium on the cadmium concentration in the soil in 83 men (left) and 147 women (right) living in a nonpolluted (open symbols) and polluted (solid symbols) area. Blood cadmium was standardized to the mean age of the population (46 years). The regression lines with 95% confidence interval are shown.

liter) = 0.8800 + 0.2860 smoking (0,1) + 0.0056 age (years) + 0.2017 log γ-glutamyl transpeptidase (U/liter) − 0.1510 log cadmium in the soil (ppm) − 0.1323 log serum ferritin (μg/liter) (R^2 = 0.42).

Cadmium in the Soil as a Determinant of Urinary Cadmium

Urinary cadmium tended to be higher in the polluted than in the nonpolluted district (9.3 versus 8.1 nmole/24 hr; P = 0.15). The single correlation coefficients between urinary cadmium and the cadmium concentration in the soil were not significant (+0.06 in men and +0.07 in women). However, after adjustment for age (linear and quadratic terms), a significant and positive association became apparent: the partial correlation coefficient between urinary cadmium and the cadmium concentration in the soil was +0.22 (P = 0.04) in men and +0.16 (P = 0.05) in women (Fig. 2).

Stepwise multiple regression showed that age and the cadmium level in the soil were the main determinants of urinary cadmium. The multiple partial R^2 for the linear and quadratic terms of age combined was 0.33 ($P < 0.001$) in men and 0.46 ($P < 0.001$) in women; for the cadmium concentration in the soil, the partial R^2 was 0.05 (P = 0.04) in men and 0.02 (P = 0.05) in women.

The regression equation in men was: log urinary cadmium (nmole/24 hr) = −0.4068 + 0.0532 age (years) − 0.00047 age^2 (years^2) + 0.1012 log cadmium concentration in the soil (ppm) (R^2 = 0.37). In women it was: log urinary cadmium (nmole/24 hr) = −0.5403 + 0.0558 age (years) − 0.00048 age^2 (years^2) + 0.0626 log cadmium in the soil (ppm) (R^2 = 0.48). With both age (linear and quadratic

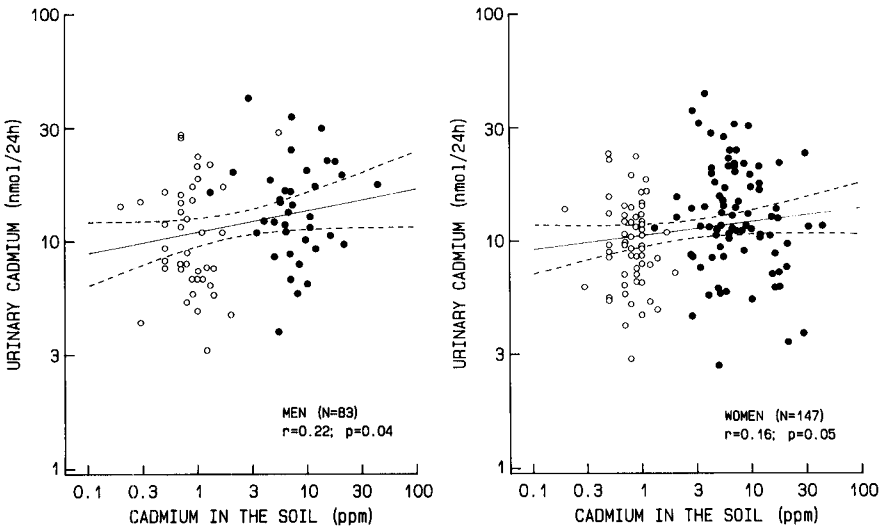

FIG. 2. Scatterplot of urinary cadmium on the cadmium concentration in the soil in 83 men (left) and 147 women (right) living in a nonpolluted (open symbols) and polluted (solid symbols) area. Urinary cadmium was standardized to the mean age of the population (46 years). The regression lines with 95% confidence interval are shown.

terms) and the cadmium concentration of the soil in the regression equation, the *t*-to-enter for smoking was 1.32 (slope = 0.0738; $P = 0.19$) in men and 1.17 (slope = 0.0507; $P = 0.24$) in women; for serum ferritin the *t*-to-enter was 0.27 ($P = 0.78$) in men and −1.35 (slope = −0.0711; $P = 0.18$) in women.

DISCUSSION

The 24-hr urinary excretion of cadmium is thought to provide an indirect measure of lifetime integrated exposure to this metal (Lauwerys, 1983). The main finding of the present study was that 2 to 4% of the variance of the body burden of cadmium was explained by the cadmium concentration in the soil. The partial regression coefficients indicated that for all other factors being constant a twofold increase in the cadmium concentration of the soil was accompanied by a 7% rise in the body burden of cadmium in men and by a 4% rise in women.

Several mechanisms may be invoked to explain the positive correlation between the cadmium body burden and the amount of cadmium in the soil. First, it cannot be excluded that the cadmium concentration in the soil does not represent more than a general marker of the overall environmental exposure to cadmium, without hinting to any specific mechanism of transfer. On the other hand, vegetables grown on contaminated soils potentially constitute an important source of dietary cadmium. An elevated level of cadmium has been demonstrated in vegetables and grains grown on polluted soils in several industrialized countries worldwide (Elinder, 1985), including the present polluted rural area (De Temmerman *et al.*, 1982; Lauwerys *et al.*, 1990). In a duplicate meal study (Buchet *et al.*, 1983), whereby foodstuffs were collected in hospital kitchens and in selected families living in a nonpolluted region of Belgium, the median daily intake of cadmium amounted to 133 nmole (15 μg). How much the intake of contaminated vegetables would increase the oral ingestion of cadmium in the present population remains presently unknown. Furthermore, cadmium may also, at least partially, be transferred via routes other than the food chain, such as via the inhalation of dust during gardening or via licking fingers contaminated by the polluted soil during childhood (Buchet *et al.*, 1980; Roels *et al.*, 1978).

The most important factor in the determination of the cadmium body burden, as measured by the 24-hr urinary cadmium excretion, was the duration of the exposure. Indeed, age explained 33% of the variance of urinary cadmium in men and 46% in women. Smoking is also an important determinant of the body burden of cadmium in subjects not exposed at work, because smoking 20 cigarettes per day results in the inhalation of 18 to 36 nmole (2 to 4 μg) cadmium (Elinder, 1985). Smoking explained less than 1 to 2% of the variation in urinary cadmium in the present subjects, when the effects of age and the cadmium content of the soil had been accounted for. It remains possible however that the effect of smoking was partially removed by the adjustment for the cadmium levels in the soil. Indeed, in workers it has been demonstrated that smoking materials, when carried in the pocket, may become heavily contaminated by cadmium, leading to the ingestion of cadmium dust via the mouth (Piscator *et al.*, 1976). A similar mechanism may be operative during gardening, especially when the cadmium level in the soil is considerably elevated.

Depleted iron stores facilitate the gastrointestinal absorption of cadmium (Flanagan *et al.*, 1978), which is estimated to average 3 to 7% of the amount orally ingested, but which can increase up to 20% in individuals with depleted iron stores (Nordberg *et al.*, 1985). In the present study, in which none of the subjects had severely depleted iron stores (Table 1), but the rural areas had been chosen to provide a wide range of environmental cadmium exposure, an inverse relationship between the internal cadmium dose and serum ferritin was only observed for blood cadmium in women; a twofold increase in serum ferritin was associated with an 8% decrease in blood cadmium.

A weak positive correlation between blood cadmium and the serum γ-glutamyl transpeptidase activity was observed in the present study. This relationship may be explained by a decreased intrahepatic storage of cadmium, when the hepatic synthesis of metallothionein is diminished, for instance as a consequence of liver function impairment (Bernard and Lauwerys, 1981) or due to an excessive alcohol intake (Bremner, 1978). Alternatively, cadmium accumulation in the liver may be accompanied by an increased release of γ-glutamyl transpeptidase from the hepatocytes into the circulation (Nordberg *et al.*, 1985). The positive relationship between blood cadmium and γ-glutamyl transpeptidase may also reflect the fact that excessive alcohol consumption and smoking, an important source of cadmium, often coexist in the same subjects.

An unexpected finding in the present study was the negative relationship between blood cadmium and the cadmium concentration in the soil. The slope of the regression equation indicated that a twofold rise in the cadmium level of the soil was accompanied by a 6% decrease of blood cadmium in men and by a 10% decrease in women. Blood cadmium, in contrast to urinary cadmium, reflects more recent exposure (Lauwerys, 1983). For 10 years the inhabitants of the polluted district have been advised by the local and provincial authorities to lime the soil of their kitchen gardens and to cultivate only vegetables that do not readily concentrate cadmium. These preventive measures may explain why the participants living in the polluted district had experienced a lower recent exposure (Lauwerys, 1983), as evidenced by their blood cadmium levels (Fig. 1), and why, as a consequence of liming, the average pH of the soil was slightly higher in the polluted district.

The geometric mean cadmium concentration in the soil was 1.2 ppm in the control area and 7.4 ppm in the polluted district. In the absence of pollution, the cadmium level in the soil is generally less than 1 ppm (Page *et al.*, 1981). This suggests that some sources of cadmium pollution must have been present in the nonpolluted district. The latter is 15 to 30 km distant from the cadmium-emitting industries in the polluted area in a direction opposite from the prevailing winds. Use of waste from the cadmium and zinc-producing plants, which has been made available at no cost to contractors and private persons for building roads and heightening terrains, and the widespread application of phosphate-containing fertilizers (Mulla *et al.*, 1980) may have been potential sources of cadmium contamination even in the area not directly polluted by the past emissions of the cadmium and zinc industry.

Although the cadmium uptake by plants can be reduced by increasing the pH of the soil to above 6.5 (Page *et al.*, 1981), there was in the present study no relationship between the internal cadmium dose and the pH of the soil. Water from private wells in the polluted area contained a considerable amount of cadmium with a median concentration of 38 nmole/liter (4.3 μg/liter) and a 90th percentile of 222 nmole/liter (25 μg/liter) (Lauwerys *et al.*, 1990). Nonetheless, the use of well water for drinking was not significantly associated with the internal cadmium dose in the present subjects, possibly as a consequence of the long-standing recommendation to avoid it for human consumption.

In conclusion, in a rural population consuming vegetables grown in kitchen gardens with a sandy acidic soil, 2 to 4% of the variance of the urinary cadmium excretion, an index of lifetime integrated exposure, can be explained by the cadmium level in the soil. The negative correlation with blood cadmium, a measure of more recent exposure, suggests that the inhabitants of the polluted district followed the guidelines to prevent cadmium uptake.

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