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Background Exposure of General Population to Cadmium and Lead in Tainan City, Taiwan

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Abstract. Venous blood samples, 24-h total food duplicate samples, and rice samples were collected from 52 adult nonsmoking women in the city of Tainan, southern Taiwan, in 1994, and analyzed for cadmium (Cd) and lead (Pb) by wet-digestion followed by graphite furnace atomic absorption spectrophotometry. Daily dietary intake was 10 μg for Cd and 22 μg for Pb as geometric means, of which Cd and Pb in rice accounted for 34% and 1.4% of daily Cd and Pb intakes, respectively. The counterpart values for blood were 1.11 ng/ml and 44.5 ng/ml for Cd and Pb, respectively. International comparison with recently published data suggests that the exposure to Cd in Tainan should be among the lowest in the world.

Both cadmium (Cd) and lead (Pb) are not only ubiquitous in the environment, but have been extensively used in industries. They are persistent in the environment once discharged, and they stay in the human body with long half-lives when absorbed. These behavioral characteristics make them good long-term markers of environmental pollution. Further, they are insidious intoxicants to humans.

Although a number of studies have been conducted to measure or estimate the intensity of exposure of the general population to these metals in several areas (Buchet *et al.* 1983; Knutti and Zimmerli 1985; Watanabe *et al.* 1985a, 1994; Becker and Kumpulainen 1991; Barberá *et al.* 1993; Conacher and Mes 1993; Chen and Gao 1993; Sung *et al.* 1993; Müller and Anke 1994; Yang *et al.* 1994), very few have ever been done in the rapidly industrializing island of Taiwan. This study was initiated to examine the present situation in comparison with the situations in other areas in the world, and to draw a baseline for expected changes in the near future.

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Materials and Methods

Participants and Sample Collection

The study was conducted in autumn, 1994, in the city of Tainan (with a population of about 700 × 10^3 inhabitants) in southern Taiwan. Fifty-two female clinical and laboratory staff in a large hospital in the city volunteered to participate in the study. They were 22 to 66 years-old, and were all nonsmokers.

On day 1, they were fully informed of the study design, and given orientation on how to collect duplicate food samples. On day 2, the participants collected 24-h food duplicates by themselves as described (Watanabe *et al.* 1985a; Ikeda *et al.* 1989). On day 3, the participants brought in the food duplicate samples to an examination room, where they had a medical interview on clinical history and drinking/smoking habits as well as anthropological measurements, venous blood sampling, blood pressure measurement, and spot urine collection. A portion of each blood sample was immediately heparinized and kept frozen for metal analysis, and the remainder was used for hematology and serum biochemistry tests.

In addition to samples from the participants, 20 uncooked rice samples were purchased from rice stores in the cities of Tainan (10 samples) and Taipei (10 samples).

Determination of Cadmium and Lead in Biological Samples

A portion (6 g) of each food duplicate after weighing and homogenization, a portion of boiled (6 g) or uncooked (6 g) rice, and an aliquot (1 ml) of each heparinized blood sample were wet-ashed, and Cd and Pb in the digests [*i.e.*, Cd in food (Cd-F) and blood (Cd-B), and Pb in food (Pb-F) and blood (Pb-B)] were measured with a graphite furnace atomic absorption spectrometer (Hitachi Z-8100) connected with an automatic liquid sampler (Hitachi SSC-200) (Watanabe *et al.* 1994) by the standard addition method.

Statistical Analysis

The distribution of Cd-F, Cd-B, Pb-F, and Pb-B was not symmetrical around a mode but showed a tailing toward higher values. Accord-

Table 1. Cadmium and lead in 24-h food duplicates and in human blood

Age range (years)	No. of subjects	Metals in food[a] (μg/day)		Metals in blood (ng/ml)	
		Cadmium	Lead	Cadmium	Lead
<30	18	8.5 (1.45)[b]	22.0 (1.72)	0.96 (1.31)	38.5 (1.28)
30–39	22	10.4 (1.90)	22.8 (2.02)	1.23 (1.27)	46.4 (1.19)
⩾40	12	12.6 (1.51)	22.4 (1.24)	1.13 (1.15)	51.2 (1.08)
Total	52	10.1 (1.70)	22.4 (1.93)	1.11 (1.39)	44.5 (1.28)

The differences among age groups are not significant as assayed by ANOVA ($P > 0.05$)
[a] In 24-h duplicates of food
[b] Geometric mean (geometric standard deviation)

Table 2. Contribution of boiled rice as a source of cadmium and lead

Item	Weight (g/day)	Amount of metal (μg/day for GM)	
		Cadmium	Lead
24-food duplicates[a]	1,990 ± 576[c]	10.1 (1.70)[d]	22.4 (1.92)
Boiled rice[a]	243 ± 114	3.44 (2.02)	0.32 (2.58)
(%[b])	(12.2%)	(34.1%)	(1.4%)

[a] Boiled rice samples, separate from 24-h food duplicates, were available from 48 subjects. Accordingly, GM (GSD) for Cd and Pb in 24-h food duplicates were calculated for the 48 samples
[b] (Amount in boiled rice/Amount in food duplicate) × 100(%)
[c] Arithmetic mean ± arithmetic standard deviation
[d] Geometric mean (geometric standard deviation)

ingly, a log-normal distribution was assumed for Cd-F, Cd-B, Pb-F, and Pb-B in samples as previously described (Watanabe *et al.* 1983, 1985a,b; Ikeda *et al.* 1989). Cd-B, Cd-F, Pb-B, and Pb-F concentrations were expressed in terms of geometric means (GMs) and geometric standard deviations (GSDs). ANOVA, multiple comparison test, and Student's *t*-test were employed to detect possible differences in distribution after logarithmic conversion of the measured values.

Results

Dietary Intake and Cd and Pb Levels in Blood

The results of analyses of 24-h food duplicates and venous blood for cadmium and lead (CD-F, Pb-F, Cd-B, and Pb-B) are summarized in Table 1 by decade of ages of the participants. The GM for all ages in combination was 10.1 μg/day for Cd-F, 22.4 μg/day for Pb-F, 1.11 ng/ml for Cd-B, and 44.5 ng/ml for Pb-B. ANOVA followed by the multiple comparison tests showed that the difference among age groups were essentially insignificant ($P > 0.05$). There was no age-related change in Pb-B.

Significance of Rice as the Source of Cd and Pb Burden

Because it is known that rice is the major source of burden of metals, especially cadmium (Ikeda 1993), the concentrations of cadmium and lead in the boiled rice that the participants consumed were measured in addition to the concentrations in the food duplicate homogenates. The intake of the two metals via rice was calculated from the concentration and the weight of boiled rice in the food duplicates. The results are shown in Table 2, in comparison with the weights and concentrations of cadmium and lead in the food duplicates. Both average weights of food duplicates and rice were calculated for the 48 participants who offered boiled rice samples separate from the whole day food duplicate samples. Calculation shows that 34.1% of cadmium in total food came from rice, whereas the amount was 1.4% in the case of lead. Boiled rice accounted for 12.2% (by weight) of whole day food. Thus, it is clear that Cd was more concentrated in boiled rice among various food items, whereas the reverse was the case for Pb. Cd and Pb contents in uncooked rice samples were determined in 39 samples offered by the participants and another 20 samples purchased from rice

Table 3. Metal concentrations in uncooked rice

Item	Amount of metal (ng/g dry weight)	
	Cadmium	Lead
Present study (whole samples)[a]	40.15 (1.74)[e]	8.9 (2.62)
Present study (from 39 participants)[b]	40.57 (1.71)	6.8 (2.17)
1989 study (11 samples)[c]	74.66 (2.15)*	13.2 (1.94)
Flour (from 9 participants)[d]	8.43 (3.77)†	14.4 (2.59)°

* Significantly ($P < 0.01$) higher than both of the present study results
† Significantly($P < 0.05$) lower than the present study results on whole rice samples
° Significantly ($P < 0.05$) higher than the present values for rice from 39 donors [insignificantly ($P > 0.05$) different from the present values for whole rice samples]
[a] A total of 59 samples collected both in Tainan (including 39 from the food duplicate donors) and in Taipei
[b] A total of 39 samples from food duplicate donors
[c] Cited from Watanabe *et al.* 1989
[d] A total of 9 samples from food duplicate donors
[e] Geometric mean (geometric standard deviation)

Table 4. Daily dietary intake of cadmium and lead by food duplicate studies or by estimation

Metal	Study area (time)	Daily dietary intake (μg/day)[a]	GM or AM[b]	Reference
Cadmium				
	Tainan	10 (W)	CM*	The present study
Asia	Japan (1980s)	44 (M), 37 (W)	GM*	Watanabe *et al.* 1985a
	Japan (1990s)	22 (W)	GM*	Watanabe *et al.* 1994
	China (Continent)	30	AM?†	Yang *et al.* 1994
	China (Continent)	14	AM†	Chen and Gao 1993
Others	Belgium	15	Median*	Buchet *et al.* 1983
	Germany	10 (M), 9 (W)	AM†	Müller and Anke 1994
	Spain	11	AM†	Barberá *et al.* 1993
	Sweden (1987)	12	AM?†	Becker and Kumpulainen 1991
	Switzerland	25	AM*	Knutti and Zimmerli 1985
	Canada (1969–72)	69	AM?*	Conacher and Mes 1993
Lead				
	Tainan	22 (W)	GM*	The present study
Asia	Japan (1980s)	38 (M), 33 (W)	GM*	Ikeda *et al.* 1989
	Japan (1990s)	11 (W)	GM*	Watanabe *et al.* 1994
	China (Continent)	29–86 (C)	GM†	Sung *et al.* 1993
	China (Continent)	86	AM†	Chen and Gao 1993
Others	Belgium	96	Median*	Buchet *et al.* 1983
	Spain	120	AM†	Barberá *et al.* 1993
	Sweden (1987)	17	AM?†	Becker and Kumpulainen 1991
	Switzerland	12	AM*	Knutti and Zimmerli 1985
	Canada (1969–72)	125	AM?†	Conacher and Mes 1993

[a]M for adult men and W for adult women. C stands for children

[b]Geometric mean or arithmetic mean. A question mark (?) indicates possibility. An asterisk (*) indicates the measures by the food duplicate collection, whereas a dagger (†) shows the estimates, e.g., by the market basket method

stores in the cities (Table 3). There were no significant changes in means between the 39 samples offered by the participants and a combination of the 39 given and 20 purchased samples. Since people may eat flour-based foods such as steamed bread and 'Yutiao' (or fried unleavened bread in a rod shape), the contents in flour were also determined. Although only nine flour samples were available, the comparison showed that Cd in flour [8.4 ng/g (3.77) as GM (GSD)] was much lower than that in rice (40.15 ng/g as GM; Table 3), whereas Pb in flour [14.4 ng/g (2.59)] might be higher ($P < 0.35$) than that in rice (8.90 ng/g as GM).

Discussion

The island of Taiwan is one of the most active foci of rapid industrialization in the world since the 1970s. Accordingly, a number of studies on possible environmental pollution have been conducted. For example, dust-fall, suspended particulates, and SO_2 and CO concentrations in urban air have been monitored as indicators of air quality (Sung and Chuang 1973). Prevalence of bronchial asthma, a classic health indicator of SO_2-type air pollution, has been studied among schoolchildren in the largest city of Taipei, and an increase from 1.30% in 1974 to 5.07% in 1985 was reported (Hsieh and Shen 1988). In the same city, 40% of the outdoor air samples contained suspended particulates at levels in excess of 150 μg/m^3, and the levels in indoor air also paralleled with the outdoor air concentrations at the levels of about 60% of the outdoor concentrations (Li 1994). In addition, people such as students who commute by motorcycle or by bus were exposed to volatile organic carbons in street air; they spent 45 to 95 min/day for commuting (Chan *et al.* 1993).

Lung cancer mortality might be higher in extensively urbanized areas (Chen *et al.* 1990). Comparative evaluation by the Ames' test of air-borne particulates showed that mutagenic activities were higher in samples from large cities of Taipei and Kaohsiung than in those from Hsinchu and Tainan (Chou and Lee 1990). Mobile sources rather than stationary ones were considered to be more important as the main pollution source of mutagenicity (Lee *et al.* 1994).

Data are scarce on secular trends of the pollution levels. Nevertheless, the latest report (Bureau of Environmental Protection, Taipei 1993) states that the average of SO_2 concentration in the atmospheric air in Taipei is less than 20 ppb in 1990–1993, in contrast to 46 ppb reported some 20 years ago (Sung and Chuang 1973). The report (Bureau of Environmental Protection, Taipei 1993) further shows reduction in lead in air from 0.34 μg/m^3 in 1988 to 0.18 μg/m^3 in 1993. It may be plausible to conclude, therefore, that air quality in Taipei City and probably also in other cities including Tainan has improved in recent years.

The risk of contamination of humans with long-persisting pollutants such as Cd and Pb has apparently been attracting less attention until now, despite the fact that heavy metals enter the human body primarily with food, especially rice (Ikeda 1993), and that people in Taiwan depend on rice as the major nutritional source for daily life. The present study in Tainan, a middle-sized city in southern Taiwan, made it clear that the daily dietary intake (as GM) among nonsmoking adult women

Table 5. Cadmium and lead levels in blood of nonexposed populations

Metal	Study area (year)	Metal level in blood (ng/ml)[a]	GM or AM[b]	Reference
Cadmium				
	Tainan	1.1 (W)	GM	The present study
Asia	China (Continent)	0.94 (M), 0.83 (W)	GM	Qu *et al.* 1993
	Japan (1990s)	1.85 (W)	GM	Watanabe *et al.* 1994
Others	Belgium	0.73–1.12 (M), 0.70–1.25 (W)	GM	Sartor *et al.* 1992
	Belgium	0.84	AM	Roels *et al.* 1993
	Czechoslovakia	1.1 (M), 1.0 (W)	AM	Cikrt *et al.* 1992
	Germany	1.0	Median	Jung *et al.* 1993
	Italy	0.36	GM	Alessio *et al.* 1993
	Sweden	0.45 (W)	GM	Lagerkvist *et al.* 1993
	U.S.A.	0.41	GM	Hovinga *et al.* 1993
Lead				
	Tainan	45 (W)	GM	The present study
Asia	China (Continent)	92 (M), 72 (W)	GM	Qu *et al.* 1993
	China (Continent)	29–86 (C)	GM	Sung *et al.* 1993
	China (Taiwan)	201 (M)	AM	Chiang and Chang 1989
	China (Taiwan)	89.9	AM	Liou *et al.* 1994
	Japan (1990s)	38 (W)	GM	Watanabe *et al.* 1994a
Others	Belgium	12.6	AM	Roels *et al.* 1993
	Czechoslovakia	96.9 (M), 83.4 (W)	AM	Cikrt *et al.* 1993
	Sweden	22.8 (W)	GM	Lagerkvist *et al.* 1993
	U.S.A.	38	GM	Hovinga *et al.* 1993

[a]M for men and W for women. C stands for children
[b]Geometric mean (GM) or arithmetic mean (AM), unless otherwise specified

was 10.1 μg/day for Cd and 22.4 μg/day for Pb, respectively (Table 1). The counterpart concentrations in blood were 1.11 ng/ml for Cd and 44.5 ng/ml for Pb (Table 1).

A number of studies have been conducted on dietary intake of environment-polluting heavy metals such as Cd and Pb, of which only results recently reported are summarized in Table 4 for comparison with the results from the present study. As rice-eating habits are more common in Asian countries than other areas in the world, the results from Asia are shown separately from other areas.

In making comparisons, factors that need to be considered include differences in sampling methods such as the 24-h food duplicate collection vs the market basket method (Acheson *et al.* 1980), as well as possible effects of gender on food intake and mineral absorption (Berglund *et al.* 1994). The effect of gender should not be ignored in the case of rice-eating people, especially in Asia, as men will take more rice (and therefore more Cd) because of a larger body size as well as heavier muscle work. In addition, some authors took AM rather than GM as the statistical representative of the measures despite the fact that the pollutant concentrations distribute log-normally rather than normally (Watanabe *et al.* 1983, 1985a,b; Ikeda *et al.* 1989). Theoretically, AM should be larger than GM when the same sample population is calculated both ways. It should further be considered that the intake may vary in a flow of time, possibly reflecting the changes in lifestyle as well as efforts for a better environment (Watanabe *et al.* 1994).

Nevertheless, simple comparison of reported values indicates the Cd burden via foods in Tainan should be among the lowest group, being comparable to many European countries (the top half in Table 4). Dietary Pb burden in contrast is not among the lowest group, although it appears much lower than the values for Spain (Barberá *et al.* 1993) and Canada (Conacher and Mes 1993). Such dietary intake should be reflected in the levels in blood. In addition to the confounders already discussed for dietary intake, social habits, especially smoking, may result in elevated metal levels in blood (Watanabe *et al.* 1983, 1985b) and general air pollution will be another source of burden (*i.e.*, via respiration), especially in the case of lead (Ikeda 1989). International comparison of blood cadmium and lead (Table 5) suggests that Cd-B in Tainan is among the lowest, as is the case of Cd-F, whereas Pb-B is not lower than others.

Recently, a large-scale survey was launched covering nearly 3,000 people in Taiwan to find the Pb-B in the general population (Liou *et al.* 1994). The nominal AM was 82.9 ng/ml, and the best estimate after adjustment for various factors was 89.9 ng/ml (Liou *et al.* 1994). Taking the factors into consideration that AM tends to be larger than GM and that the coefficient of variation was in excess of 70% in the study by Liou *et al.* (1994) as expected, it is reasonable to conclude that two study results—89.9 ng/ml as AM by Liou *et al.* (1994) and 57 ng/ml as GM in the present study—are essentially the same.

From the present results combined with assumptions of several physiological parameters [*i.e.*, the respiration volume is 15 m^3/day, and the absorption rates of Cd and Pb are 50% when inhaled (Heard and Chamberlain 1983) and 5–10% when ingested (Kitamura 1972)], it is possible to calculate (Ikeda 1993) for the present participants (*i.e.*, nonsmoking and nondrinking women in Tainan) that they will absorb 3.4 μg Pb/day through the respiration of the atmospheric air conditioning 0.32 μg Pb/m^3 [the concentration reported for Tainan (Environment Protection Administration 1991)] and 1.7 μg Pb/day via ingestion of 22 μg Pb/day in food (Table 1), or 5.1 μg Pb daily through both routes. A counterpart value for nonsmoking, nondrinking women in Japan is 0.6 μg Pb/day via inhalation, 2.5 μg Pb/day via ingestion, and 3.1 μg Pb/day via both routes

(Ikeda *et al.* 1989). It should be noted that the Pb burden via inhalation accounts for about 67% of the total in Tainan women, whereas it is about 19% for Japanese women. The rate in total burden between the two populations, 1.65:1 (*i.e.*, 5.1 μg/day for Tainan women vs 3.1 μg/day for Japanese women) is similar to the ratio of Pb-B, 1.41:1 [*i.e.*, 45 ng Pb/ml in women in Tainan (Table 1) vs 32 ng Pb/ml in Japanese women (Watanabe *et al.* 1985b)].

In the case of Cd, dietary intake of 10 and 37 μg Cd/day by women in Tainan (Table 1) and their counterparts in Japan (Watanabe *et al.* 1985a) will result in the absorption of 0.8 and 2.8 μg Cd/day, respectively. The counterpart values for Cd-B were 1.1 (Table 1) and 3.6 ng/ml (Watanabe *et al.* 1985b). The rate in Cd-F between Tainan and Japanese women (1:3.7) is close to that in Cd-B (1:3.3) because Cd burden via respiration is essentially negligible when compared with that via ingestion (Ikeda *et al.* 1989).

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