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Data Report

Estimation of daily cadmium intake in the United States through food analysis in 1980

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ABSTRACT — Cadmium is an environmental contaminant that accumulates in the human kidneys. Non-rice grains, vegetables, and potatoes are major sources of cadmium in the U.S. However, the daily cadmium intake levels reported in studies conducted in the U.S. vary widely, ranging from 4.63 to 51.3 µg/day/person. Most studies in the U.S. have not measured the actual cadmium concentrations in the collected foods but utilized the database of the average cadmium concentrations provided by the U.S. Department of Agriculture (USDA). In the present study, food and beverages were collected extensively in the U.S. from 1979 to 1980, and the actual cadmium concentrations were determined. Individual food intake data were obtained from a total diet study conducted in 1980 by the USDA. In accordance with previous reports, vegetables, grains, and potatoes were the primary sources of dietary cadmium intake. The estimated total cadmium intake was approximately 15 µg/day/person. We also found a 17-fold difference in cadmium concentrations in carrots between the production areas, which was greater than the variation reported for carrots in Japan. The factors affecting the variation in cadmium concentrations in vegetables, including carrots, need to be clarified. As the relative importance of vegetables as a source of cadmium intake is increasing in Japan due to decreased rice consumption, more attention should be paid to variations in cadmium concentrations in vegetables in Japan.

Key words: Cadmium, Daily intake, Food analysis, United States

INTRODUCTION

Chronic exposure to cadmium results in its preferential accumulation in the kidneys of both humans and animals. The kidney is a critical organ involved in cadmium toxicity. The biological half-life of cadmium in humans has been reported to be up to 40 years (Yokohashi *et al.*, 1973), 13 years (Tsuchiya *et al.*, 1976), and 10–33 years (Ellis *et al.*, 1979). The primary sources of cadmium among nonsmokers are foods and beverages (Lewis *et al.*, 1972; Shuman *et al.*, 1974). Cadmium-containing grains

and vegetables are the predominant determinants of daily cadmium intake worldwide.

Human renal cadmium concentrations vary among countries (Kjellström, 1979; Perry et al., 1961; Tipton et al., 1969). An early study reported that renal cadmium concentrations in humans were the highest in Japan, followed by those in Asian countries such as Thailand, Hong Kong, and Taiwan (Perry et al., 1961). By contrast, renal cadmium concentrations were low in the United States (U.S.), Switzerland, India, Nigeria, and Ruanda Urundi (Perry et al., 1961; Kjellström, 1979). Several studies

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conducted in the 1990s in Japan also reported high cadmium concentrations in the kidneys of the general Japanese population (Noda *et al.*, 1993; Yoshida *et al.*, 1998; Yoshinaga *et al.*, 1990). The primary source of cadmium in Japanese foods is rice, which contributed to more than 50% of the total dietary cadmium intake in the 1980s (Aoshima and Horiguchi, 2019). However, the dietary cadmium intake of Japanese people, which was 30–40 µg/day in the 1980s, has since declined gradually, reaching less than 20 µg/day in the 2010s due to decreased rice consumption (Aoshima and Horiguchi, 2019; Watanabe *et al.*, 2022).

In contrast, the major sources of cadmium in food are vegetables and non-rice grains in Western countries, including the U.S. However, inconsistent values have been reported for dietary cadmium intake in the U.S. population. Average levels of daily cadmium intake have been reported to be 51.3 µg/person (Mahaffey *et al.*, 1975), 17.4 µg/person for females, 18.2 µg/person for males (MacIntosh *et al.*, 1996), and 4.63 µg/person (Kim *et al.*, 2018). The reason for the 10-fold difference in the reported daily cadmium intake in the U.S. remains unclear.

Most U.S. studies on dietary cadmium intake have not collected food samples or determined the actual cadmium concentrations in the collected samples. Instead, they used a database of average food cadmium concentrations provided by the U.S. Department of Agriculture (USDA). The author of this article (S. S.) collected various foods and beverages in the U.S. from 1979 to 1980 and analyzed the cadmium concentrations in the collected samples. Here, we show the results of this rare study that estimated the daily cadmium intake levels of the U.S. population depending on the actual cadmium concentrations in the collected food samples and compared the results with the reported values.

MATERIALS AND METHODS

Sampling of foods and beverages

Food and beverages were purchased from supermarkets in Houston, Boston, and Los Angeles in 1979 and 1980. Raw foods such as vegetables and fruits and processed foods such as canned, dried, smoked, and cooked foods were collected with the help of nutritionists. Prepared foods such as hamburger steak, dessert cake, and boiled vegetables were also collected from several restaurants. Carrot samples produced at nine locations in the U.S. and Canada were collected for detailed analysis.

Analysis of cadmium

First, wet weights of the collected food and beverage samples were determined. Subsequently, the dry weights were determined after the samples were dried in an oven at 105°C for two days. Samples were added with 1.0 mL of concentrated nitric acid, left standing overnight, and heated on a hot plate at 300°C for one to three days. After ashing, the samples were dissolved in 1.0 mL of nitric acid (1:5). Twenty microliters of the solution were applied to a graphite furnace (HGA-2100, Perkin-Elmer Corp., Connecticut, U.S.) of the atomic absorption spectrophotometer (AAS) (Model 603, Perkin-Elmer Corp., U.S.) under the following conditions: drying at 110°C for 20 sec, charring at 400°C for 15 sec, and atomizing at 1,750°C for 5 sec. Background correction was automatically performed using a deuterium lamp.

A calibration curve was constructed from the standard solution, and the internal standard method was used to check for the matrix effect. To ensure measurement accuracy, certified reference materials (CRMs) prepared by the U.S. National Bureau of Standards for rice flour (No. 1567), spinach flour (No. 1570), and bovine liver powder (No. 1577) were processed in the same manner as the collected samples and applied to the AAS.

The detection limit for cadmium by the adopted graphite-furnace AAS was 0.5 ng in 100 mg of dried food samples or 1.0 mL of solution samples. Recovery tests were conducted by spiking two levels of cadmium: one was similar to and the other was twice the rice cadmium levels. The recovery percentages were 108% and 105%, respectively. The coefficient of variation for ten analyses of the same sample was 18%.

Calculation of daily intake of cadmium

The concentrations of cadmium in foods and beverages are presented as either ng/g wet weight or ng/g dry weight. As cadmium concentrations in foods have a lognormal distribution (Shacklette *et al.*, 1978), the geometric mean (GM) concentration and geometric deviation (GD) were calculated for each food group. For samples below the detection limit, one-third of the detection limit was used to calculate the average values (Shacklette *et al.*, 1978).

The average intake of each food group was obtained from the Total Diet Study (TDS), also known as the Market Basket Survey, conducted by the USDA (USDA, 1978; USDA, 1983). The average (GM) cadmium concentration in each food group determined in this study was multiplied by the average intake of the corresponding food group obtained from the TDS data.

RESULTS AND DISCUSSION

Daily cadmium intake in the U.S.

Since vegetables are an important source of Cd intake in the U.S. population, extensive vegetable items were collected (Table 1). As reported previously (Kim et al., 2018; Monteiro et al., 2009; Nawrot et al., 2010), high concentrations of Cd (>500 ng/g dry weight) were detected in leafy vegetables such as lettuce, celery, cabbage, and spinach. To assess the contribution of each food group to daily cadmium intake, the average (GM) cadmium concentration of each food group (ng/g) was multiplied by the average intake of each food group (g) reported by the USDA TDS conducted in 1980. As shown in Fig. 1, vegetables, grains (mainly flour and wheat products), and potatoes were the primary sources of cadmium. Although the average cadmium concentrations in potatoes and grains were lower than those in leafy vegetables, a higher intake of these foods contributed to a higher daily cadmium intake. The total daily cadmium intake, the sum of the cadmium intakes from all food groups, was 14.7 µg/person in 1980 (Table 2). We also calculated the daily cadmium intakes based on the USDA TDS data for 1960, 1965, 1970, and 1975, in addition to 1980, but they were approximately the same at 15 μ g/person from 1960 to 1980.

Comparison with other studies

Most U.S. studies have not measured actual cadmium concentrations in foods and beverages; they utilized a database of average cadmium concentrations in individual foods listed in the TDS report conducted periodically by the USDA. To estimate daily cadmium intake, these studies multiplied the average cadmium concentrations in the TDS database by the intake of each food item; the latter was obtained from their own studies using a food frequency questionnaire or other methods (Adams et al., 2012; Gunderson, 1988; Gunderson, 1995; Kim et al., 2018; Mahaffey et al., 1975). However, the daily cadmium intake reported in these studies varied widely. Mahaffey et al. estimated the cadmium intake as 51.3 µg/day/person based on the USDA's TDS data from 1973 (Mahaffey et al., 1975). Adams et al. reported the arithmetic mean of cadmium intake being 10.9 µg/day/ person using the USDA TDS data from 1991 to 2008 (Adams et al., 2012). Kim et al. estimated the cadmium

Table 1. List of foods collected for cadmium concentration measurements.

Food group	Food item					
Milk and dairy products	Milk, Cheese (natural, processed)					
Eggs	Egg (white, yolk)					
Meat and poultries	Beef (hamburger, stew, rib, kidney, and liver), Chicken, Chicken liver, Pork, Sausage, Luncheon meat, Bacon,					
Fish	Perch, Sardine (canned), Shrimp (dried), Tuna (canned), Red snapper (fried), Clams (canned), Herring (smo and canned), Oyster (canned)					
Flour and cereal products	Wheat flour, Bread, Waffles, Cake, Biscuit, Cookie, Cracker, Ice cream cones, Spaghetti (dry and cooked), Macaroni, Noodles, Wheat germ, Cereal (wheat, oats, rice, and corn), Sweet corn (raw, canned, and frozen), Cornbread, Popcorn, Corn meal, Rice (polished)					
Potatoes	Potato, Sweet potato, Potato chip					
Vegetables						
Leafy Vegetables	Lettuce, Celery, Broccoli (raw and boiled), Cabbage, Spinach, Parsley, Cauliflower, Asparagus					
Root Vegetables	Onion, Radish, Turnip, Ginger, Carrot					
Garden Vegetables	Tomatoes (raw, canned, catsup, and paste), Pumpkin (canned), Squash, Cucumber (raw and pickled), Strawberry, Okra, Sweet pepper, Vegetable soup (canned), Vegetable juice cocktail (canned)					
Beans, peas, and nuts	Snap beans (raw and frozen), Peanuts (roasted and buttered), Green peas (canned and frozen), Sunflower seed kernels, Lentils (dried), Common beans (dried), Lima beans (frozen), Cowpeas (frozen), Walnut (dried), Almonds (dried), Pecans					
Fruits	Prune, Raisin, Pear, Apricot (dried), Grape, Applesauce, Olive (pickled), Tangerine, Orange, Grapefruit, Lemon, Apple, Melon, Cherry (canned), Banana, Avocado, Pineapple, Orange juice (canned), Peach (dried)					
Fats and Oil	Salad Oil, Salad dressing, Margarine, Butter					
Sugar and sweeteners	Sugar, Syrup					
Beverages	Tea (instant), Coffee (instant), Cocoa mix, Lemonade (concentrated), Coke, 7-up, Root beer, Wine, Whisky					
Others	Mustard, Mushroom (bottled), Bouillon (powder), Baking powder, Mayonnaise, Tartar sauce, Yeast, Vinega Onion soup, Mushroom soup, Tomato soup, Chicken soup, Table salt, Cinnamon sugar, Pepper, Pimento, C powder, Vermicelli, Taco shell					

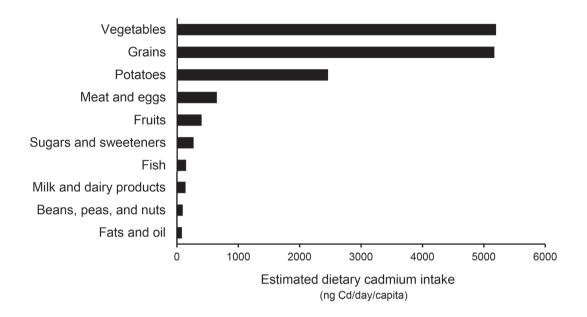


Fig. 1. Estimated dietary cadmium intake. The estimated dietary cadmium intake by food group in 1980, shown in Table 2, was calculated and expressed as a bar chart.

intake to be 4.63 µg/day/person using the USDA's TDS data from 2006 to 2012 (Kim et al., 2018). Thus, there is a 10-fold difference in reported daily cadmium intake values in the U.S. In 2011, Joint FAO/WHO Expert Committee on Food Additives (JECFA) summarized the daily cadmium intakes reported from several countries, and that of the U.S. was 4.6 µg/kg/month (JECFA 2011), corresponding to 10.7 µg/day/person. The daily cadmium intake of $\sim 15 \mu g/person$ obtained in the present study, which examined actual cadmium concentrations in foods and beverages, appears to be an intermediate value among values reported thus far. In line with our results, Kowal et al. estimated the daily cadmium intake to be 13–15 μg by analyzing the daily cadmium intake through cadmium excretion in the stool collected in Chicago and Dallas in 1974–1976 (Kowal et al., 1979).

Contrary to the considerable variation in cadmium intake levels reported by the U.S. studies, Swedish studies have shown relatively stable levels of cadmium intake, ranging from 10 to 18 µg/day/person between 1974 and 1983 (Järup *et al.*, 1998). It is possible that the differences in daily cadmium intake reflect the decline in cadmium concentrations in U.S. foods or the change in the intake of some food groups. However, based on our food analysis, the daily cadmium intake in the U.S. pop-

ulation remained at about 15 μg/person even when the TDS data of the USDA from 1960 to 1980 were used (Table 2). Since infant cadmium intake has been a concern in the U.S., the cadmium intake among infants has been periodically monitored, but it remained at approximately 10 µg/person from the 1980s to the 2020s in the U.S. (Hoffman-Pennesi et al., 2024; Spungen 2019). In contrast, daily lead intake decreased drastically during the same period, suggesting that food contamination by cadmium in the U.S. did not change as markedly as that by lead (Hoffman-Pennesi et al., 2024; Spungen 2019). These authors also suggested that the difference in daily cadmium intake between their studies (Hoffman-Pennesi et al., 2024; Spungen 2019), and the study by Kim et al. (Kim et al., 2018) could be attributed to the selection of food items for cadmium intake estimation. Differences in the frequency with which study subjects consumed cadmium-containing vegetables, such as spinach, have also been shown to cause significant differences in estimated cadmium intakes in the U.S. (Pokharel and Wu, 2023).

JECFA showed that the ranges of cadmium concentrations worldwide for wheat and rice were 9–40 μ g/kg wet weight and 4–20 μ g/kg wet weight, respectively, whereas that for vegetables was as wide as 6–100 μ g/kg wet weight (JECFA, 2011). JECFA also showed that vege-

Table 2. Estimated per capita dietary cadmium intake by food groups calculated with GM concentrations of cadmium and the food consumption data by the USDA from 1960 to 1980.

Food groups	Estimated dietary cadmium intake (ng/day/capita)					GM concentration	Food consumption by food groups (g/day/capita)				
Year	1960	1965	1970	1975	1980	of cadmium (ng/g)	1960	1965	1970	1975	1980
Meat	273	277	307	296	298	1.5	183	185	205	197	199
Poultry	172	206	243	245	303	4.0	42.9	51.4	60.8	61.3	75.8
Fish	113	122	136	142	145	6.9	16.4	17.7	19.9	20.5	21.0
Eggs	66	62	61	55	54	1.3	52.9	49.5	49.1	44.0	43.0
Cheese	21	23	27	31	36	1.3	16.2	17.7	20.7	23.8	27.5
Milk	134	130	125	123	105	0.30	448	435	416	409	350
Fats and oil	66	70	77	78	81	1.1	60.0	63.3	69.9	70.4	73.3
Fruit (fresh)	366	326	328	333	351	3.3	111	98.8	99.3	101	107
Fruit (processed)	51	49	57	57	56	0.82	62.4	59.8	69.2	69.4	68.9
Vegetables	4,980	4,850	5,020	5,110	5,200	20	249	243	251	256	260
Potato	3,140	3,010	2,870	2,870	2,420	24	131	125	120	120	101
Sweet potato	46	45	43	43	36	9.6	4.80	4.70	4.50	4.50	3.80
Beans, peas, and nuts	111	108	104	120	95	5.3	20.5	20.4	19.6	22.6	17.9
Wheat	4,350	4,180	4,070	3,960	4,320	29	150	144	140	137	149
Maize	125	132	137	140	137	7.0	17.8	18.8	19.6	20.0	19.6
Other cereals	592	648	612	648	716	40	14.8	16.2	15.3	16.2	17.9
Sugar	31	32	34	35	38	0.30	105	108	114	115	128
Sweetener	185	190	205	207	229	6.0	30.8	31.7	34.1	34.5	38.1
Coffee	43	42	40	35	31	2.5	17.1	16.9	16.0	13.9	12.4
Tea	5	5	5	4	4	2.5	1.9	1.9	1.8	1.5	1.4
Total	14,870	14,507	14,501	14,532	14,655						

Table 3. Cadmium concentrations in carrots produced in nine different places in the U.S. and Canada.

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Places of production	Cadmium concentration (ng/g)	Coefficient of variation (%)	
Ontario, Canada	20.8 ± 18.1 ^a	$(5)^{b}$	87.0
Saugus, California	51.2 ± 13.8	(5)	27.0
Westfield, Massachusetts	84.8 ± 32.0	(4)	37.7
Saugers, California	88.8 ± 27.4	(5)	30.9
Byron Center, Michigan	184.4 ± 75.4	(5)	40.9
Rochester, New York	187.4 ± 73.3	(5)	39.1
Salina, California	215.3 ± 32.7	(4)	15.1
Hartville, Ohio	237.2 ± 77.3	(5)	32.6
Boston, Massachusetts	351.3 ± 69.3	(3)	19.7

^a, Arithmetic mean ± standard deviation

tarians' cadmium intake was 23 μ g/kg/month, approximately 2-fold that of the Japanese population (12 μ g/kg/month), the highest-exposed country at that time (JECFA 2011). Thus, food intake frequencies of particular vegetables containing different cadmium concentrations might be one of the reasons for the considerable variation in the estimated cadmium intake in the U.S. population.

Variation in cadmium concentrations in carrots

Vegetables were also a key contributor to the daily cadmium intake in the present study (Fig. 1). Carrots were selected for a more detailed analysis because they are eaten worldwide, and some studies have shown a high accumulation of cadmium in carrots (Collado-López et al., 2022; Yang et al., 2009). As shown in Table 3, cadmium concentrations in the carrots produced in different areas of the U.S. and Canada varied from 20.8 to 351 ng/g dry weight, a 17-fold difference, with an average (GM) concentration of 120 ng/g dry weight. These values were lower than the reported carrot cadmium concentrations of 150 ± 270 ng/g dry weight in China (Yang et al., 2009) and 490 ± 180 ng/g dry weight in Croatia (Stančić et al., 2016) but higher than those in Poland (41 \pm 15 ng/g dry weight) (Rusin et al., 2021). Intriguingly, the average (AM) cadmium concentrations in the carrot samples produced in the same geographical areas showed a wide variation: 51.2, 88.8, and 215 ng/g dry weight in California, and 84.8, and 351 ng/g dry weight in Massachusetts (Table 3). These results suggest that factors other than geographical area are involved in the variations in carrot cadmium concentrations, although the soil map for metals in the U.S. showed substantial regional differ-

b, Number of samples

ences in cadmium concentrations (Schaefer et al., 2020). In contrast to the U.S., cadmium concentrations in carrots in Japan fell within a smaller range. Lee et al. collected 373 carrot samples from 46 prefectures in Japan from 1996 to 1998 and showed that the average (GM) cadmium concentration in the carrots was 24.4 ng/g wet weight (GD 2.2) (Lee et al., 1999). When the prefectures were grouped into nine districts, the carrots produced in the Hokuriku district showed the lowest (15.8) and those in the Chugoku district showed the highest (36.7) cadmium concentrations; however, the difference was only 2-fold (Lee et al., 1999). In addition to soil cadmium concentrations, the amounts and types of phosphate fertilizers in which a part of the calcium phosphate is replaced by cadmium phosphate may serve as a source of cadmium accumulation in plants, although their actual contribution has not been fully quantified (Dharma-Wardana, 2018; Jiao et al., 2012; Schroeder and Balassa, 1963). Because carrots have been shown to accumulate soil cadmium more efficiently than other vegetables (Yang et al., 2009), a slight difference in the amounts of environmental cadmium supplied from soil or fertilizers could have caused considerable variation in cadmium concentrations in carrots.

In conclusion, the present study estimated the daily cadmium intake level in the U.S. population to be about 15 μ g/person by measuring the actual cadmium concentrations in foods and beverages collected in 1979–1980. This value fell at an intermediate level among previously reported values, possibly because of the extensive collection of various vegetables. This study also showed a marked variation in carrot cadmium concentrations in the U.S., suggesting that variations in other vegetables produced in different regions or under different conditions should be more carefully monitored. As cadmium intake from rice has been decreasing in Japan, the same attention should be paid to cadmium concentrations in vegetables in Japan.

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Conflict of interest--- The authors declare that there is no conflict of interest.

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