

Content of Cadmium in Carrots Compared with Rice in Japan

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Cadmium (Cd) in the human diet still constitutes a potential chronic hazard to health. Cd accumulates in the kidneys and harmful effects are likely to occur when its concentration in the renal cortex exceeds 200 $\mu\text{g/g}$ fresh weight (Fassett, 1980). Suzuki et al. (1988) and Kawada et al. (1998) reviewed the Cd concentrations in the human renal cortex which ranges from 10 to 30 $\mu\text{g/g}$ wet weight for Europeans and Americans, while it is 65 to 115 $\mu\text{g/g}$ wet weight for Japanese who have always had the highest renal Cd levels in the world. The amount of Cd accumulated in the kidneys is a function of age and/or daily Cd intake, the latter of which comes mostly from foods and beverages. Cd in drinking water and in the atmosphere contribute very little to such accumulation (Yokohashi 1973). Consequently, the monitoring of levels of Cd in various foods is essential for the better understanding of Cd accumulations in the human body. As the pollution levels of Cd have decreased as a result of measures and regulations against hazardous substances in almost every country, the background survey of Cd has become increasingly important.

Much data on Cd content of rice in Japan have been reported (Masironi et al., 1977; Morishita 1981; Nakatsuka et al., 1988; Rivai 1990) and reviewed by Kawada and Suzuki (1998). Rice is not a dominant staple in the world, despite the numerous rice-eating ethnic groups such as Chinese, Indonesians and Japanese, etc. Moreover, rice consumption in Japan has decreased about 33% in the last twenty years. Per capita rice consumption was 248.3 gm/day in 1975 and it was 167.9 gm/day in 1997 (Ministry of Health and Welfare, 1997). Now rice consumption represents only 11.6% of the average person's daily food intake and it is still decreasing (Ministry of Health and Welfare, 1997). On the other hand, green and yellow vegetables (carrot, spinach, pimento, tomato et al.) consumption in Japan has increased considerably since the 1970s: In 1975, green and yellow vegetable consumption per person was about 48.2 gm/day and in 1995 it rose to 94.0 gm/day (Ministry of Health and Welfare, 1997). Thus, our study was designed to assess Cd content in the edible portion of a popular root crop namely carrots which are consumed all over the world. In order to confirm whether or not carrots can be used as an indicator food of environmental monitoring of Cd, the present study aimed: 1) to assess Cd concentration in carrots in Japan, 2) to know the distribution of Cd content in carrots by geography, village/city, prefecture, regions, and by soil type, and 3) to compare the relation of Cd content in carrot and rice.

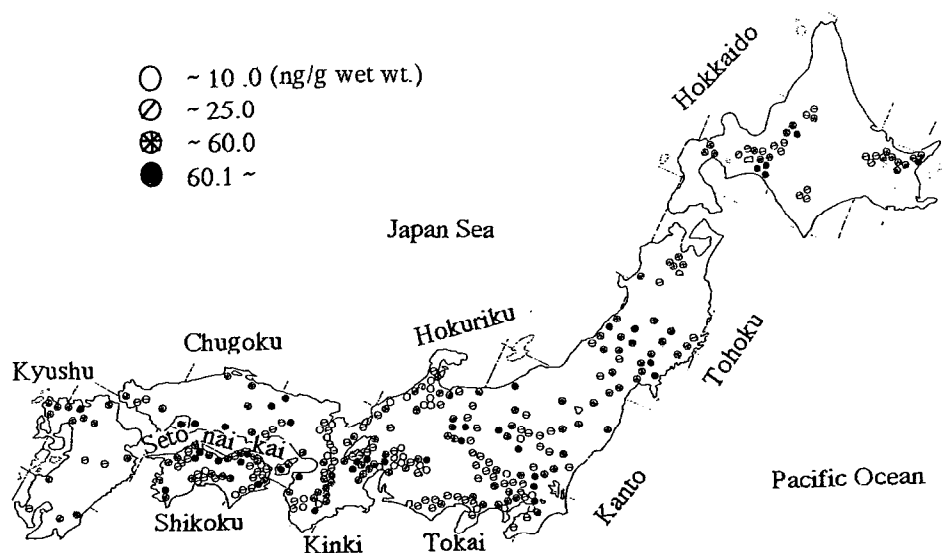


Figure 1. Distribution of cadmium concentrations of carrot samples in Japan (ng/g wet weight)

MATERIALS AND METHODS

In the harvesting seasons of 1996-98, 373 carrot samples were collected from 232 villages, towns and cities of 46 prefectures except Okinawa. According to geographical features and vegetation, 46 prefectures were divided into 9 regions: Hokkaido, Tohoku, Kanto, Hokuriku, Tokai, Kinki, Shikoku, Chugoku and Kyushu. The sampling site locations are shown in Figure 1. The carrot samples were delivered in the following three ways: 1. local greengrocers; 2. carrot farmers; 3. JAs (Japan Agricultural Cooperative Association). All of the carrot samples were confirmed to be locally grown products.

The fresh carrot samples were first thoroughly washed with tap water, then rinsed with distilled water and left to drip dry. Approximately 1 gram of each fresh carrot sample was put into a metal-free test tube for pretreatment and weighing. Subsequently, samples were dried in an oven at 105°C for 48 hours and weighed again to assess the water content. The completely dried carrot samples were ashed on a hot plate (80°C) with 1.0 ml concentrated metal-free nitric acid until dryness. 1.0 ml of 14% nitric acid was then added to dissolve each dried sample. Cd in the solutions was measured by a flameless atomic absorption spectrophotometer (AAS) (Hitachi Z-9000) at a wavelength of 228.8 nm.

Under the same conditions, rice flour (No. 10-b) as standard materials which were distributed by Japan National Institute for Environmental Studies (NIES) was used for reference to confirm the accuracy. The reliability of Cd determination was 7.4%, in terms of the coefficient of variation of 10 repeated analyses of the same sample lot. A recovery test was determined by adding 0.04 µg of Cd to each ten carrot samples (0.1 gram). The recovery rate was 103% on average, ranging from 95% to 114%.

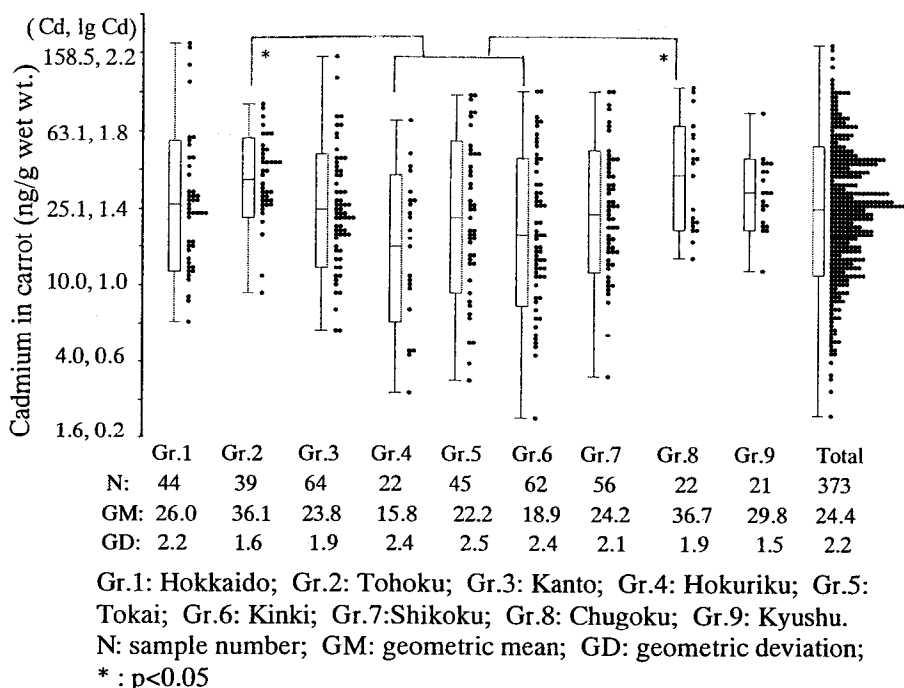


Figure 2. Distribution of cadmium contents of carrot samples from 46 prefectures in 9 regions in Japan

Rice samples were collected in 1988, and were measured by a few of the present authors by the same methods of analysis. Cd contents in carrot and in rice are compared in the present paper. Identification of soil type was based on the "Soil Map of Japan" (National Land Agency 1996, scale: 1:50,000). Out of 373 carrot samples, 268 had the places of cultivation confirmed. The rest of the samples which fell on a borderline between two soil types were excluded.

Statistical software, NAP, was used to make all the statistical calculations (Aoki, 1989)

RESULTS AND DISCUSSION

Since the frequency distribution of the 373 carrot samples was skewed to the left, the geometric mean (GM) and the geometric deviation (GD) were calculated. GM and GD of the total 373 carrot samples were 24.4 $\mu\text{g/g}$ wet weight and 2.2, respectively, and the range was from 2.1 to 179.1 $\mu\text{g/g}$ wet weight. For our reference, Arithmetic mean and standard deviation of the total 373 carrot samples were 32.2 ± 25.4 ng/g wet weight.

The sampling site locations and Cd levels of carrot samples in 9 regions in Japan are shown in Figure 1. The four classes of Cd concentration (Figure 1) and the sample distributions are: ~ 10.0 ng/g wet wt., 43 samples; 10.1~25.0, 137 samples; 25.1~60.0, 143 samples; and 60.1~, 50 samples. A few samples were excluded in

Table 1. Cadmium contents in carrot (ng/g wet wt.) reported by other investigators (including the present study).

Mean	N	Range	Country	Reference
32	373	2 ~179	Japan	Present study
11	11	1 ~ 18	Finland	Tahvonen and Kumpulainen 1991
30	4	20 ~ 40	Finland	Varo <i>et al.</i> 1980
56	--	0 ~145	United States	Gunderson 1989
35	--	14 ~ 75	Germany	Schelenz and Bobbel 1983
35	--	104	Germany	Kampe 1983
40	100	--	Germany	König 1989
41	--	3 ~ 160	Sweden	Jorhem <i>et al.</i> 1984
40	--	5 ~ 160	The Netherlands	Wiersma <i>et al.</i> 1986
28	207	2 ~ 130	United States	Wolnik <i>et al.</i> 1985
12,18	--	--	Canada	Dabeka and McKenzie 1992
15	41	1 ~ 41	United States	Suzuki 1980
24	--	2 ~ 74	Finland	Tahvonen and Kumpulainen 1995

Mean: arithmetic mean; N: sample number; --: none

Figure 1 whose places of production are not known on the village level. The Cd concentrations in carrot samples appear higher in north Kyushu, Seto-naikai (inland sea) side of Shikoku and Chugoku, and the middle of Kinki.

The distribution of Cd concentration in carrot samples is shown by 9 regions as in Figure 2. As the distribution of 373 carrot samples was skewed, the Cd content was calculated on a logarithmic scale. The highest geometric mean of Cd in a region was Chugoku (36.7 ng/g), followed by Tohoku (36.1 ng/g). The lowest was Hokuriku (15.8 ng/g) then Kinki (18.9 ng/g). Geometric mean of Cd concentration in carrot samples were significantly higher in Chugoku and Tohoku than in Hokuriku and Kinki($p<0.05$) by Multiple comparisons of Ryan’s method.

Our data of Cd content in carrots were compared with those of other investigators (Table 1). The averages ranged from 11 to 56 ng/g wet weight. Compared to other studies, the carrot Cd levels found in our study were general. The sources of low level Cd contamination of agricultural and horticultural soils are usually phosphate fertilizers, atmospheric deposition from industrial sources, and sewage sludge (Hutton and Symon, 1987). Low Cd-content phosphate fertilizers used in Japan may be a contributing factor. The Cd concentrations of phosphate fertilizers in Japan are controlled to be less than 10 $\mu\text{g/g}$ (Shibuya 1975).

Table 2 shows the difference in Cd content in the carrot and rice samples classified by soil type. Seven soil types were identified from the collection sites of carrot samples as: *Cambisols*, *Andosols*, *Fluvic Gleysols*, *Gleysols*, *Acisols-Luvisols*, *Fluvisols* and *Histosols*. The soil type *Histosols* was attributed to the highest amount of Cd in carrot (41.1 ng/g), and *Fluvisols* to the lowest (22.3 ng/g). The Cd content of carrot grown in *Histosols* soil type was significantly higher than that in *Fluvisols* soil type by t-test ($p<0.05$). A survey on the trace elements in soils by

Table 2. Geometric means and geometric deviations of cadmium in carrot and in rice samples by soil types in Japan (ng/g wet wt.)

soil type	cadmium in carrot			cadmium in rice		
	N	GM	GD	N	GM	GD
1. <i>Cambisols</i>	41	27.2	2.6	81	50.1	2.7
2. <i>Andosols</i>	80	24.4	1.9	8	70.0	3.2
3. <i>Fluvic Gleysols</i>	65	29.8	2.5	--	--	--
4. <i>Gleysols</i>	48	25.2	2.2	3	81.8	2.4
5. <i>Acrisols and Luvisols</i>	12	23.6	2.2	--	--	--
6. <i>Fluvisols</i>	16	22.3	2.1	11	171.9*	2.2
7. <i>Histosols</i>	6	41.1*	1.4	--	--	--

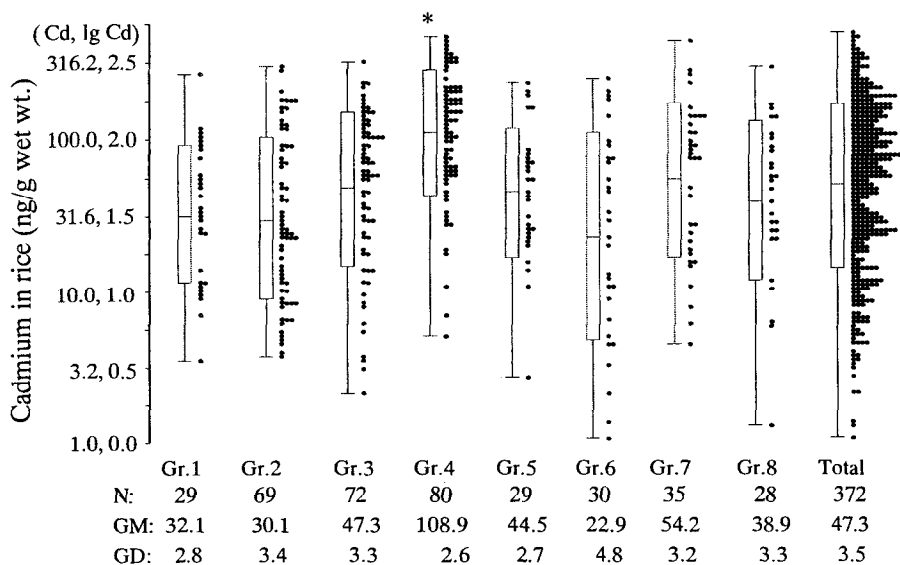
N, GM, GD: see the footnote of Figure 2; *: $p < 0.05$, by t test.

Pendias AK (1992) revealed *Histosols* had the highest content of Cd (780 ng/g) among all the soil types. Harrison and Chirgawi (1989) reported that soil uptake accounted for 69-94% of the Cd in plants. Substantial differences have been found in heavy metal uptakes among plant species, and the capacity of carrot to accumulate Cd is moderate to strong (König 1989).

The Cd contents in rice by soil types are also shown in Table 2. The highest Cd level was in the soil type *Fluvisols* (171.9 ng/g), and the lowest (50.1 ng/g) was in *Cambisols*. The Cd contents of carrot grown in *Fluvisols* soil type were significantly higher than the other soil types (t-test $p < 0.05$). Possibly, Cd in *Fluvisols* is absorbed most easily by rice plants than from other soil types (Rivai 1990). The Cd concentration of rice samples from Hokuriku were significantly higher than those of other provinces (Figure 3). *Fluvisols* in Japan mainly appear along the sea coasts (Matsuzaka 1982). The high Cd content in rice of Hokuriku is thought to be due to the high Cd level of *Fluvisols* soils there.

The distribution of Cd concentrations of rice samples in 8 regions are shown in Figure 3. The number of rice sample in Kinki region was only two which were excluded from statistical analysis. Since the distribution of the rice samples was skewed, the logarithm of Cd content was calculated. The highest geometric mean of Cd was that of Hokuriku (108.9 ng/g), followed by Chugoku (54.2 ng/g). The Lowest ranking province was Shikoku (22.9 ng/g). Multiple comparisons by Ryan's method showed that geometric mean of Cd concentration of rice samples in Hokuriku was significantly higher than those in the other provinces ($p < 0.05$).

Since production places of most of rice sample are known only on prefectural level, the relationship was not significant between geometric mean of Cd concentration of rice and carrot samples ($r = -0.163$). In rice data, there were 20 production places



Gr.1: Hokkaido; Gr.2: Tohoku; Gr.3: Kanto; Gr.4: Hokuriku; Gr.5: Tokai;
 Gr.6:Shikoku; Gr.7: Chugoku; Gr.8: Kyushu.
 N, GM, GD, *: See the footnote of Fig.2.

Figure 3. Distribution of cadmium contents of rice samples in 8 regions in Japan.

known on city or village level. The Cd concentration of 20 pairs of carrot and rice samples showed significant relationship ($r=0.492$, $p<0.05$) which is shown in Fig.4.

Although the correlation coefficient between the mean Cd concentration in rice and carrot samples both from prefecture and region level was negative, that from cities or villages was positive. In the same prefecture, the soil type, irrigation water, and air pollution level differ by cities, towns or villages as Japan is a mountainous country with various soil types. The Cd concentrations in crops of different villages in the same prefecture should be different too. Therefore it is difficult to compare the relationship between mean Cd concentrations of rice and carrot samples at prefectural or regional levels. On the other hand, the soil types, irrigation waters, and air pollution levels in the same city or village are more similar than over a whole prefecture, so the relationship between Cd concentration of rice and carrot was significant at city or village level.

In 1968 the Ministry of Health & Welfare in Japan, officially admitted that “Itai-itai disease” was caused primarily by Cd, and three years later “Agricultural Land Soil Pollution Prevention and Others Law” was enacted. Since 1971, 6,110 ha in 56 Cd polluted areas in Japan were designated and counterplans for soil restoration established. Untill 11 Nov. 1994, out of 56 Cd polluted area, soil restoration in 52 areas was carry out and completed except 39 areas which were cancelled of polluted soil (Environmental Agency Water Preservation Bureau 1994). In the present study,

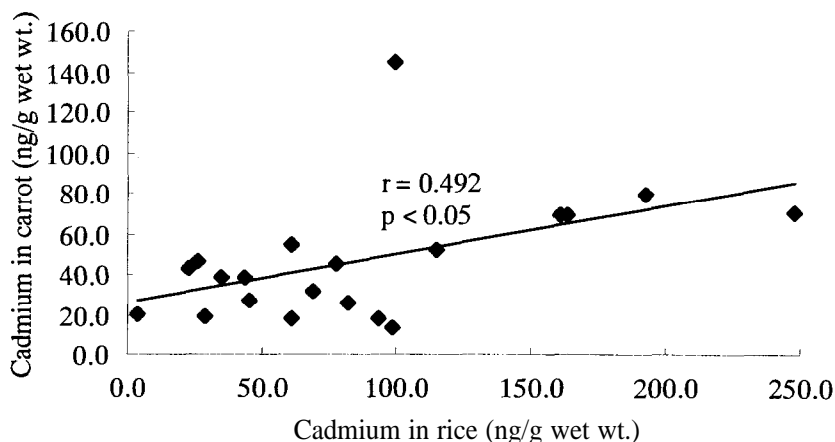


Figure 4. Relationship between cadmium concentrations of rice and carrot samples from 20 cities and villages.

the carrot samples were collected at random from 46 prefectures out of 47 prefectures in Japan, the sampling sites include both Cd polluted and non-polluted areas. The acceptable limit of Cd in carrot was proposed to be 100 ng/g (Tahvonen 1995). In our present study, only in 5 cities and towns, the mean Cd content of carrot samples were higher than the acceptable standard. Out of the 5 areas, three are Cd-polluted areas, the Cd concentrations of carrot samples were 225.2, 213.3 and 179.6 ng/g (wet wt.), respectively. These three cities were excluded from the statistical analysis in this paper. Another city (145.4 ng/g) is near the Cd-polluted area and the other one is “non-polluted” city (160 ng/g) in Hokkaido. In these three high Cd areas, most of the soil restoration programs were already completed. The following problems still remain: 1) designation of policy area covers only limited part of the whole polluted farms; 2) Selection of cheaper engineering methods for restoration has brought insufficient results far different from returning to an original non contaminated state; 3) Fear of repeated pollution is considerable due to imperfect prevention measures of pollution resource (Morishita 1998).

Until now, to estimate daily Cd intake, determination of Cd in rice was recommended for rice-eating countries. But in other nations, rice consumption is very low. Therefore as the relationship between Cd concentration of rice and carrot samples indicated a positive significance in the present study, and carrot being a common root crop, consumed all over the world with most of the crop consumed locally, carrots are an ideal indicator food for the monitoring of Cd pollution and for the comparison of baseline level of Cd exposure in other countries of the world as opposed to rice.

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REFERENCES

- Aoki S (1989) Reference Manual of Medical Statistics for NAP. Igakusyoin, Tokyo
- Dabeka RW, McKenzie AD (1992) Total diet study of lead and cadmium in food composites: preliminary investigations. *Journal of AOAC International* 75: 386-394
- Environmental Agency Water Preservation Bureau (1994) The outline of the agriculture soil pollution prevention countermeasure
- Fassett DW (1980) Cadmium. In: Waldron HA (ed) *Metals in the Environment*, Academic Press, London, p 61-110
- Gunderson EL (1989) Total Diet Study, April 1982-April 1984, Dietary Intakes of Pesticides, Selected Elements, and other Chemicals. Chemical Contaminants Monitoring. Data Tables. AOAC Source Document No. 1 (Arlington, VA AOAC)
- Harrison RM Chirgawi MB (1989) The assessment of air and soil as contributors of some trace metals to vegetable plants III. Experiments with field-grown plants. *Science of the Total Environment* 83:47-62
- Hutton M, Symon C (1987) Sources of cadmium in the UK environment. In: P.J. Coughtrey, Martin MH, Unsworth MN (eds) *Pollution Transport and Fate in Ecosystems*, Blackwell Scientific, Oxford, p 223-237
- Jorhem L, Mattsson P, Slorach S (1984) Lead, cadmium, zinc and certain other metals in foods on the Swedish market. *Vår Föda*, 36, suppl. 3
- Kampe W (1993) Blei und Cadmium in der Nahrung-eine aktuelle Gefährdung? *Verbraucherdienst* 28:58-61, 75-78
- Kawada T, Suzuki S (1998) A review on the cadmium content of rice, daily cadmium intake, and accumulation in the kidneys. *J Occup Health* 40 (4): 264-269
- König W (1989) Schwermetallbelastung von Böden und Pflanzen in Haus- und Kleingarten des Ruhrgebietes, *Verhandlungen der Gesellschaft für Ökologie* (Esse 1988) 18:325-331
- Masironi R, Koirtzyhann SR, Pierce JO (1977) Zinc, copper, cadmium and chromium in polished and unpolished rice. *Sci., Total Env.* 7:27-43
- Matuuzaka Y (1982): Major soil groups in Japan. In: Finkl, C.W. Jr. (ed.) *Soil Classification* Hutchinson Ross, Stroudsburg, Pennsylvania, p 270-276
- Ministry of Health and Welfare, the Government of Japan (1997) Nutritional Status in Japan, based on National Nutrition Survey in 1995. Dai-Ichi Shuppan, Tokyo
- Morishita T (1981) The Jinzu River Basin: Contamination of soil and paddy rice with cadmium discharged from Kamioka mine. In: Kitagishi K (ed) *Heavy Metal Pollution in Soil of Japan*. Japan Sci, Soc. Press, Tokyo, p 107-124
- Morishita T (1998) Problems on cadmium polluted rice and restoration of polluted soils in Japan. In: *International Symposium on Itai-itai Disease, Environmental Cadmium Pollution and Countermeasures, Abstracts of Communications*, Toyama, Japan, p 73-74
- Nakatsuka H, Watanabe T, Ikeda M (1988) Cadmium content in rice from various countries. *Jpn. J. Hygiene* 43, 1, 126 (in Japanese)
- National Land Agency Land Bureau National Land Investigation Section (1996) *Soil map of Japan*
- Pendias AK, Pendias H (1992) *Trace elements in soils and plants*. CRC press,

- London, 2nd ed., p 3-43
- Rivai IF, Koyama H, Suzuki S (1990) Cadmium content in rice and its daily intake in various countries. *Bull Environ Contam Toxicol* 44:910-916
- Schelenz R, Bobbel B (1983) Veränderungen der Gehalte von Arsen, Blei, Cadmium und Quecksilber bei der haushaltsüblichen Zubereitung. *Landwirtschaftliche Forschung, -Sonderheft* 39:342-352
- Shibuya M, Yamazoue F (1975) Environmental contamination and agriculture. Hakuyusha, Tokyo
- Suzuki S (1980) unpublished
- Suzuki S, Koyama H, Hattori T, Kawada T, Rivai If (1988) Daily Intake of Cadmium: An Ecological View. In: Sumino (ed) *Environmental and Occupational Chemical Hazards*, No 8. Kishimoto Publ & Print p 205-217
- Tahvonen R, Kumpulainen J (1991) Lead and cadmium in berries and vegetables on the Finnish market 1987-1989. *Fresenius Journal of Analytical Chemistry* 340: 242-244
- Tahvonen R, Kumpulainen J (1995) Lead and cadmium in some berries and vegetables on the Finnish market in 1991-1993. *Food Additives and Contaminations* 12 (2):263-279
- Varo P, Lähelmä O, Nuortamo M, Saari E, Koivistoinen P (1980) Mineral element composition of Finnish foods. VII. Potato, vegetables, fruits, berries, nuts and mushrooms. *Acta Agriculturae Scandinavica* 22:89-113
- Wiersma D, Goor BJ, Veen N (1986) Cadmium, lead, mercury and arsenic concentrations in crops and corresponding soils in The Netherlands. *J Agric Food Chem* 34:1067-1074
- Wolnik KA, Fricke FL, Capar SG, Meyer MW, Satzger RD, Bonnin E, Gaston CM (1985) Elements in major raw agricultural crops in the United States. 3. Cadmium, lead, and eleven other elements in carrots, field corn, onions, rice, spinach, and tomatoes. *Journal of Agricultural and Food Chemistry* 33: 807-811
- Yokohashi G, Suzuki S, Matsubara J (1973) Kinetics of cadmium in mammals in the environment. In: Sasa M, Yamamoto T (eds) *Environment and Human Survival*, Tokyo University Press, Tokyo, 120