# Chromium and Manganese in Japanese Diet

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#### ABSTRACT

Measurements of chromium and manganese in the complete diet samples collected throughout Japan were made to know the normal daily intake and percentage contribution from different food-groups. More than 70 per cent of manganese is supplied by plant products, while chromium is more evenly supplied by each food-group as in the case of iron. The daily intake of manganese is  $2.7\sim2.9\,\mathrm{mg}$  in adults. The ratio Mn:Cr in raw food materials on the average was found to be 21.0 (range: $5.4\sim63.6$ ), while that in cooked servings was only  $3.1\sim4.0$ ; this may suggest the introduction of chromium in the course of cooking.

#### INTRODUCTION

The presence of chromium-51 was reported in Columbia River water down the reactor site, marine organisms and agricultural products<sup>1)</sup> in association with the Hanford reactor operation. Manganese-54 was found widely in fresh-water<sup>2)</sup>, marine organisms<sup>3)</sup> and even in bovine and human livers<sup>4)</sup> principally as the result of nuclear weapon tests.

The possible entry of radioisotopes which have been discharged to the surface

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of the earth into man through the food chain should be estimated for the preliminary planning of radiological protection of the public. A study on the distribution of important stable elements in human environment is one of the important projects in radioecological investigation. In this concern, such element as manganese<sup>5</sup>, iron<sup>5</sup>, cobalt<sup>6</sup>, zinc<sup>7</sup> and cesium<sup>8</sup> were analyzed in the Japanese diet and the results were previously reported.

In the present study, the stable chromium and manganese were analyzed in diet samples collected throughout this country and the daily intake and contribution by different food-groups were estimated.

### TOTAL DIET SAMPLES

The total diet samples used for this study are composed of two series. The first series indicated as "well-balanced diets recommended for Japanese people" was collected under the instruction of Prof. Y. Hiyama at the University of Tokyo. The method in which the first type of diet is employed is called the "Standard Recipes Method". It should be noted that the food samples were purchased on the market and analyzed of mixtures of uncooked raw materials. These foodstuffs were classified into seven groups of food category as shown in Table 1.

The second series indicated as "collection of cooked servings" was collected under the instruction of Dr. H. Hayami at the National Institute of Nutrition. This differs from the first series in representing actually consumed cooked servings in chosen families. The first series of diet sample was used to estimate the contribution of the elements by different food sources, while the second one for the purpose of obtaining the daily intake of the elements by people.

An example of the standard recipes after which the first series of diet samples was collected was previously described in detail (N. and T. Yamagata, 1964) and the difference with respect to the nutritional aspects on the diet between the two series was also discussed by one of the present authors (N. Yamagata, 1962).

#### ANALYTICAL METHOD

Sample ash  $(0.3 \sim 0.4 \, \mathrm{g})$  was moistened with several drops of water and the organic matter was completely decomposed by heating with the mixture of nitric and sulfuric acids with occasional addition of nitric acid and the final addition of 30% hydrogen peroxide followed by evaporation to fumes. The residue was dissolved by boiling with  $10 \, \mathrm{m}l$  of water, and the solution was filtered and the residue was washed with three portions of each  $2 \sim 3 \, \mathrm{m}l$  of  $0.1 \, N$  sulfuric acid to make about  $20 \, \mathrm{m}l$  sample solution.

Manganese was determined spectrophotometrically by the permanganate method by use of silver peroxide as an oxidant, in which the coloration of chromate did not interfere<sup>10</sup>. Chromium was determined by a diphenylcarbazide method after decomposition of permanganate with sodium azide<sup>11</sup>)

**Table 1.** Chromium and manganese contents of well-balanced diets recommended for Japanese

No. Time of collection Locality	Classification of food-groups	Daily intake (g)		Daily Cr (µg)	intake of Mn (mg)
6201 K January 1962 Kagoshima	Cereals	453		7.9	2.25
	Pulses, nuts & fruits	204		15.9	0.702
	Leafy vegetables	202		36.8	0.500
	Root vegetables	184		18.0	0.385
	Seaweeds	6.8		23.4	0.240
	Fish & shellfish	98		19.8	0.133
	Meat, eggs & milk products	141		8.2	0.067
			Total	130.0	4.277
6301 H	Cereals	446		70.3	2.00
January 1963 Hokkaido	Pulses, nuts & fruits	123		56.7	0.592
Homaido	Leafy vegetables	164		10.6	0.761
	Root vegetables	236		24.4	0.890
	Seaweeds	7.3		10.3	0.040
	Fish & shellfish	96		37.7	0.181
	Meat, eggs & milk products	95		19.8	0.344
			Total	229.8	4.808
6307 T	Cereals	517		17.7	2.01
July 1963 Tokyo	Pulses, nuts & fruits	226		136	0 993
	Leafy vegetables	120		>1.7	>0.133
	Root vegetables	183		50.7	0.545
	Seaweeds	3.3		3.7	0.031
	Fish & shellfish	98		33.9	0.176
	Meat, eggs & milk products	72		9.8	0.079
	E 2000		Total	253.5	3.967

## CONTRIBUTION BY DIFFERENT FOOD-GROUPS

In Table 1 are shown the contents of chromium and manganese in foodstuffs which were grouped into seven and were collected at three localities. The daily intake of each food-group (g) was calculated by summing up the consumption in four weeks of each food item shown in the standard recipes and dividing by 28. The condition under which foodstuffs are sold on the market differs by item; some are dry and some wet. Each food item was weighed in its raw dry or wet condition. Therefore, concentrations of the elements shown in Table 2 are expressed on the basis of the weight of the mixture of dry and wet foodstuffs in each food-group. On this basis, seaweeds are the richest sources of both elements, however, it should be taken into consideration that most of seaweeds is sold in dried condition. Cereals are the richest in manganese and on the contrary, the

Classification of	Cı	r	Mn		Mn/Cr
food-groups	Range	Average	Range	Average	Average
Cereals	0.017~0.16	0.070	3.89~4.97	4.45	63.6
Pulses, nuts & fruits	0.078~0.60	0.38	3.44~4.81	4.22	11.1
Leafy vegetables	$0.065 \sim 0.182$	0.12	$2.47 \sim 4.64$	3.56	29.7
Root vegetables	$0.098 \sim 0.277$	0.16	$2.09 \sim 3.78$	2.95	18.4
Seaweeds	$1.1 \sim 3.4$	2.0	5.5~35.3	16.6	8.3
Fish & shellfish	$0.202 \sim 0.393$	0.31	$1.36 \sim 1.89$	1.68	5.4
Meat, eggs & milk products	0.058~0.21	0.14	0.48~3.62	1.70	12.1
Weighted average	$0.100{\sim}0.208$	0.17	$3.26{\sim}4.12$	3.57	21.0

Table 2. Average concentrations of chromium and manganese in different food-groups (p. p. m.)

poorest in chromium.

Percentage contribution by each food-group to the total intake is schematically shown in Fig. 1. The main contributor of manganese in Japanese diet is cereals, and the percentage contribution is estimated to be almost 50 per cent, while chromium is more evenly supplied by each food-group.

The results obtained for manganese are quite similar to those previously reported on the diet samples (N. and T. Yamagata, 1964)<sup>5)</sup> which were collected from different localities at different time, showing more than 70 per cent contribution by plant pro-

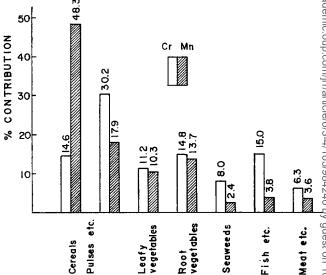


Fig. 1. Contribution of chromium and manganese by different food-group to the total intake

ducts. In contrast, the contribution pattern of chromium reflects lower contents of this element in cereals in spite of the greatest amount of consumption of cereals in daily diet and rather high levels in animal products. The pattern is close to that obtained for iron reported previously<sup>5</sup>).

### DAILY INTAKE OF CHROMIUM AND MANGANESE

As mentioned previously, the daily intake by people was estimated on the basis of the analytical results on the diet samples from family groups. Each datum shown in Table 3 represents the analytical result on a composite of cooked servings for one complete day collected from ten families each.

**Table 3.** The daily intake of chromium and manganese by Japanese people (based on collection of cooked servings, mg/d/person)

Locality	Time of collection	Cr	Mn
	Urban a	lults	
Hokkaido	1962	0.70	03 2.71
Miyagi	1962	0.24	12 3.22
Tokyo	1962	1.71	4.16
Osaka	1962	3.32	?* 1.89
Fukuoka	1962	0.20	2.87
"	1963	0.75	1.39
		Av. 0.72	23 2.71
	Rural a	dults	
Hokkaido	1962	2.05	5.76
Miyagi	1962	>0.18	>1.84
Tokyo	1962	0.59	0 1.07
Osaka	1962	1.19	1.53
Fukuoka	1962	0.70	7 1.59
"	1963	0.94	0 5.82
		Av. 0.94	3 2.93
	Rural chi	ldren	
Hokkaido	1962	0.66	3.53
Miyagi	1962	0.26	3.15
Tokyo	1962	0.31	2 1.88
Aichi	1962	0.26	2.06
Osaka	1962	1.66	3.31
Fukuoka	1962	1.81	5.03
11	1963	0.34	8 2.44
		Av. 0.76	3.06

<sup>\*</sup> Excluded in making average.

The daily intake on the average of manganese by adult people was 2.71 and 2.93 mg, which is in good accordance with the previous work<sup>5)</sup> which showed  $1.5\sim$  2.8 mg for urban and  $2.0\sim3.0$  mg for rural people. However, the daily intake by rural children seems to be rather higher when comparing with the previous estimation which showed values a little larger than a half of those of adults; the reason is not clear.

The difference between the daily intake of manganese estimated on the basis of "well-balanced diet" and "collection of cooked servings", namely,  $3.9\sim4.8$  mg in the former and  $2.7\sim2.9$  mg in the latter, is not significant. In contrast, the difference of daily intake of chromium between the two series is significantly large as much as a factor of four on the average. The ratio Mn: Cr in the raw food materials varied from 5.4 in fish and shellfish to 63.6 in cereals (Tab. 2), while the average ratio in cooked servings was almost constant and it showed a smaller value of only  $3.11\sim4.03$  (Tab. 3).

As to the reason of higher concentrations of chromium in cooked servings comparing with those in raw materials, there is no evidence but it is suspected the introduction of this element in the course of cooking possibly by corrosion of kitchen utencils. This should be confirmed by further experiments in view of both toxicological and radiological importance.

Assuming the ratio Mn: Cr in the raw food materials is 21 on the average (Tab. 2), the mean daily intake of chromium could be calculated to be 0.13~0.14 mg on the basis of the values 2.71 and 2.93 mg for manganese. These values are in good agreement with those cited in the I. C. R. P. Recommendations<sup>12)</sup>, namely, 0.15 and 3.1 mg for chromium and manganese respectively.

#### REFERENCES

- Foster, R. F. (1963) Environmental behavior of chromium and neptunium. p, 569-576 in "Radioecology" ed. by V. Schultz and A. W. Klement, Jr. Reinhold Publ. Corp., New York
- 2. Gaglione, P. and O. Ravera (1964) Manganese-54 concentration in fall-out, water, and Unio mussels of Lake Maggiore. *Nature*, 204: 1215-6.
- 3. Nagaya, Y. and T. R. Folsom (1964) Zinc-65 and other fallout nuclides in marine organisms of the California Coast. J. Rad. Res. 5: 82-89.
- 4. Sax, N. I. and J. J. Gabay (1965) Occurrence of <sup>54</sup>Mn in human and bovine livers. Health Physics, 11: 585.
- 5. Yamagata, N. and T. Yamagata (1964) The daily intakes of manganese and iron by Japanese people with reference to marine environmental contamination with radioisotopes. *Bull. Inst. Publ. Health*, 13 (1): 11-20.
- 6. Yamagata, N., Kurioka, W. and T. Shimizu (1963) Balance of cobalt in Japanese people and diet. J. Rad. Res., 4: 8-15.
- 7. Yamagata, N. and K. Iwashima (1962) Balance of zinc in Japanese people and diet with reference to environmental contamination with radioactive zinc-65. *Bull. Inst. Publ. Health*, 11 (3): 131-138.
- 8. Yamagata, N. (1962) The concentration of common potassium, rubidium and cesium in Japanese diet. J. Rad. Res., 3: 158-169.
- 9. Yamagata, N. and T. Yamagata (1964) Dietary Intakes of fallout radioactivities by the Ryukyuans. Bull. Inst. Publ. Health, 13 (3): 162-169.
- 10. Kimura, K. and Y. Murakami (1950-1951) Application of a new oxidizing reagent —"silver peroxide"—in microanalysis. *Mikrochemie*, 36/37: 727-740.
- 11. Suzuki, Y. and Y. Murakami (1966) Simultaneous colorimetric determination of manganese and chromium with diphenylcarbazide. To be appeared in Bull. Chem. Soc. Japan.
- 12. Report of ICRP. Committee II on Permissible Dose for Internal Radiation (1959) *Health Physics* 3: 164-165.