Journal of the Korean Nuclear Society
Volume 8, Number 4, December, 1976

≪Original≫

Study on the Concentration of Some Trace Elements in the Diet in Korea*

Chul Lee, Nak Bae Kim and Ihn Chong Lee

Radioanalytical Division, Korea Atomic Energy Research Institute

(Received August 30, 1976)

Abstract

The present levels for the contents of mercury, arsenic, bromine, copper, manganese and potassium in various groups of foodstuffs are evaluated and compared with data given previously by others. On the basis of the data for the levels, the average daily intakes have been estimated to be $2.0\times10^{1}\mu g$ for Hg, $3.2\times10^{2}\mu g$ for As, $2.3\times10^{3}\mu g$ for Br, $2.5\times10^{3}\mu g$ for Cu, $6.8\times10^{3}\mu g$ for Mn and $2.1\times10^{6}\mu g$ for K. For this purpose, the data reported as a result of the 1974 National Nutrition Survey have been used.

요 약

여러 식품류에 함유된 수은, 비소, 브롬, 구리, 망간 및 칼륨의 현재 준위를 제시하였으며 이미 발표된 다른 저자들의 결과와 비교 하였다. 여기서 얻은 현재의 함량 준위를 근거로하여 각 원소들의 매일 섭취량을 산출 하였더니 수은이 $2.0\times10^{1}\mu g$, 비소가 $3.2\times10^{2}\mu g$, 브롬이 $2.3\times10^{3}\mu g$, 구리가 $2.5\times10^{3}\mu g$, 망간이 $6.8\times10^{3}\mu g$, 칼륨이 $2.1\times10^{6}\mu g$ 이었다. 이들 값을 산출하기 위하여 1974년 수행된 국민영양 조사의 결과를 이용하였다.

1. Introduction

A considerable interest has been taken in the residues that can occur in foodstuffs as a result of the use of various chemical compounds in agriculture and industry. Special problems of food and environmental contamination by mercury were discussed by many experts.[1] The ubiquity and acute toxicity of arsenic has been reported in some detail by Schroeder and Balassa[2] as well as by Heydorn[3]. The tolerance doses for bromine as a pesticide residue in foods were discussed by Staerk and Suess[4]. Besides these, a need for further studies was dis-

*Most of data were collected during the work supported in part by IAEA under the contract number 1136.

cussed by Hamilton and Minski[9] in connection with interrelations between elements and with human morbidity due to deficiency of some elements.

In this study, an attempt has been made in order to evaluate the present levels for the contents of some elements such as mercury, cadmium, arsenic, bromine, copper, manganese and potassium in various groups of foodstuffs. The results are summarized and compared with data given by previous authors. For the analyses of the elements, a neutron activation method involving chemical procedures[5,6] has been used. The result of this study provided an apportunity to assess the daily intakes of mercury, arsenic, bromine, copper, manganese and potassium as follows.

2. Materials and Methods

2.1. Collection and pretreatment of samples

Samples of rice and fresh-water fish were collected directly from the different sites of the country with a few exceptions. The samples of marine fish, fruits and farm products were collected through the Agricultural and Marine Products Cooperative Union in Seoul.

Samples of cereals such as rice, barleycorn, bean etc. were washed with the distilled water and dried in a desicator until they gave a constant weight. These samples were subjected to neutron irradiation without further treatment. Fresh-water fish and marine fish were cut into pieces with a plastic knife. Axial muscles were taken and used for the investigation. Skin and bones were removed as completely as possible, but small muscular bones could not always be avoided. The pieces of muscles were washed several times with distilled water. They were transferred into polyethylene bags and dehydrated by drying for 3 days in a thermovac freeze unit, Virtis Model 10-45 MR-TR. Fruits were skinned. Inner portions were transferred into polyethylene bags and dehydrated as described above. Edible portions of vegetables and sea weeds were also taken as samples. They were washed and dehydrated in the same way.

Table 1. The gamma-ray energies selected

Isotope	Half-life	Selected energy
^{197}Hg	65 hr.	0.077 and 0.068 MeV
^{76}As	26.5 hr.	0.56 MeV
^{82}Br	35.5 hr.	0.78 MeV
^{64}Cu	12.9 hr.	0.511 MeV
^{56}Mn	2.58 hr.	0.845 MeV
^{42}K	12.4 hr.	1.52 MeV

2.2. Neutron Irradiation

All neutron irradiations have been carried out by using the pneumatic transfer systems of rotary specimen racks of the TRICA mark II and mark III reactors. Polyethylene and silica ampoules have been used for the irradiation of samples in order to determine bromine, arsenic, copper and manganese, whereas only silica ampoules have been used for the determination of mercury.

Iron wire has been used for the single comparator or the flux monitor when the samples were irradiated for 1 hours[5]. When irradiated for a longer time, all samples were sealed in silica ampoules before irradiation. In the longer irradiations, the cobalt monitor prepared as described below has been used as the flux monitor.

An accurately weighed piece of cobalt metal was dissolved in diluted nitric acid.

Table 2. The Summary of Analytical Results (ppm, wet basis)

Food	No. of samples	Water content (%)	Hg			As	
			This study ($\pm 1\sigma$)	Other (range)	Refs.	This study ($\pm 1\sigma$)	Other (range)
Rice	44	0	0.040±0.059	0.02~1 (Japan/Korea)	14, 15, 16, 17	0.22±0.3	0.1~0.5
				0.004~0.015 (other)	10, 19		
Barley-corn	1	0	0.013±0.006			0.019±0.004	
Pulse	2	0	0.029±0.010			0.021±0.002	
Vegetable	7	94	0.0038±0.002	0.001~0.13	12, 22, 29	0.0040±0.0038	0.1~1.3
Fruit	5	89	0.0052±0.0028	0.001~0.13	12, 22, 29	0.19±0.40	0.1~1.3
Milk powder	1	0	0.013±0.00323	0.0003~0.01	10, 13, 20, 22, 29	0.010±0.002	<0.1
Beef	1	81	0.013±0.0006	0.001~0.06	10, 13, 21	0.032±0.0019	0.1~1.4
Marine fish	40	81	0.059±0.053	0.04~4.8	10, 13, 23, 24, 25, 29	1.4±1.3	0.5~16
Mollusca	4	81	0.019±0.011	0.001~0.6	13, 26, 27, 28	1.3±1.3	0.5~80
Crustacea	8	81	0.075±0.070	0.02~2.5	13, 29	4.6±3.4	1~72
Shell	12	81	0.034±0.040			1.3±0.7	
Sea-weed	3	0	0.044±0.019			40±19	
Fresh-water fish	27	81	0.11±0.13			0.081±0.28	

After diluting to a finite volume with the distilled water, a 100λ volume of the solution (5μg per ml) was pipetted onto a polyethylene sheet and dried under an infra-red lamp. After folding, the sheet was used an the flux monitor.

2.3. Nuclear Data

The nuclear data pertinent to the present work are shown in Table 1.

2.4. Determination of Mercury, arsenic, bromine, copper, manganese and potassium.

When the elements other than mercury were determined simultaneously, the procedures reported previously[5] were generally adopted after irradiating samples in polyethylene vials. When mercury was determined simultaneously with the other elements, the samples were irradiated in silica ampoules and separated as follows.

Mercury and bromine were distilled into the reservoir of a Bethge apparatus by using the modified Sjöstrand method described previously.[6] Bromine activities were first measured as described[5] in the previous report after transferring the distillate into a 50 ml volumetric flask. Mercury was then determined by using the procedures which were described previously.[5,6] The remaining residues in the distillation flask of the Bethge apparatus were transferred into a 100 ml volumetric flask and diluted to the volume with the distilled water. A 25 ml protion of the solution was loaded on a cation exchange resin column which was then separated using the procedures in the previous report.[5] Gamma-ray spectrometry was then applied for the determination of arsenic, copper, manganese and potassium as described in the report.[5] When these procedures were applied to the reference kale of Bowen, the results appeared to agree with the data reported previously.[5,6]

	Br			Cu	Mn	K
Refs.	This study (±1σ)	Other(range)	Refs.	This study (±1σ)	This study (±1σ)	This study (±1σ)
2	0.43±0.20	4~11	47	2.9±2.8	11±3	690±130
	4.4±0.8			3.5±0.5	8.9±0.5	1500±50
	2.2±1.2			8.1±1.8	24±9	13000±4800
2	0.47±0.27	0.3~45	34	0.37±0.41	0.66±0.53	780±470
2	0.028±0.052	0.3~45	34	2.3±3.4	2.9±5.6	760±330
2	7.2±0.4	1	34	0.13±0.006	0.17±0.02	2000±600
2	0.36±0.076	0.7~5	34	22±1	0.23±0.02	1000± 19
2, 30, 32, 33	3.8±2.5	100	12	1.8±2.7	0.48±0.32	970±340
2, 26, 30, 31, 32, 33	25±38	250	34	2.5±1.5	0.87±0.97	720±550
2, 30, 31, 32, 33	25±25	100	12	6.3±5.7	3.0±6.5	530±270
	17±8.0			6.3±15	4.6±6.5	570±360
	57±8			3.3±1.2	56±20	9800±1000
	0.48±0.43			0.38±0.52	0.21±0.19	660±340

3. Results and Discussion

The data in the previous report[7], which were given on dry basis, converted to fresh wet basis by using water contents given in Table 2, which were obtained from weight difference between the fresh weight and freeze-dried weight. The analytical data are summarized into the following groups: rice, barley-corn, pulse, vegetable, fruit, milk, beef, marine fish, mollusca, crustacea, shell, sea-weed and fresh-water fish as shown in Table 2. In this table, other data reported by previous authors are also given for the comparison.

On the basis of the 1974 National Nutrition Survey[8], in couple with the analytical data given in Table 2 and in the previous report[7], the average daily intakes of the elements have been estimated as shown in Table 3. In this work, food items which have been analyzed do not cover fully the range of foodstuffs in the National Nutrition Survey. By using the available analytical data only, the daily intakes of the elements were estimated approximately as follows.

By assuming that the processed fish and vegetables are same as the fresh ones, the available data of the fresh were substituted for those of the processed. When the data of some species given on the National Nutrition Survey were not available, the average value of the group or the analytical data of other species in the same group were arbitrarily used. The details are given as follows.

The analytical data of barleycorn were used for those of wheat flour, corn and etc., because these are not available. Similarly, the data of soy-bean were used instead of those of some samples such as fermented soybean paste. The average values of marine fish given in Table 2 were used for the evaluation of the normal daily contribution by the fish and shell group given in Table 3. Additionally, the average value of the vegetable group on dry basis

Table 3. Summary of the Daily Contribution of Elements by Various Food Components

Food	Consumption (g/head/day)	Daily Contribution of Elements (μg/day$\pm 1\sigma$)					
		Hg	As	Br	Cu	Mn	K
Cereal	500	12 ± 13	53 ± 66	1300 ± 190	1600 ± 420	4900 ± 420	$5.7\times10^5\pm3.0\times10^4$
Pulse	27	1.0 ± 0.04	0.52 ± 0.02	29 ± 1.5	260 ± 50	880 ± 18	$4.8\times10^5\pm1.5\times10^4$
Potato	53	0.58 ± 0.09	0.29 ± 0.03	14 ± 0.1	10 ± 0.1	49 ± 6	$1.9\times10^5\pm7.5\times10^3$
Vegetable	289	1.1 ± 0.3	1.0 ± 0.4	160 ± 30	84 ± 46	230 ± 60	$2.1\times10^5\pm5.4\times10^4$
Fruit	28	0.14 ± 0.08	5.3 ± 11	0.78 ± 1.0	64 ± 95	81 ± 150	$2.1\times10^4\pm9.1\times10^3$
Sea-weed	4.8	0.21 ± 0.09	190 ± 91	270 ± 38	16 ± 6	270 ± 96	$4.7\times10^4\pm4.8\times10^3$
Seasoning	37	2.4 ± 1.4	2.5 ± 2.3	290 ± 170	230 ± 250	400 ± 330	$4.8\times10^5\pm2.9\times10^5$
Meat, Egg and Milk	10	0.13 ± 0.007	0.29 ± 0.01	15 ± 0.9	190 ± 13	2.2 ± 0.1	$1.2\times10^4\pm1.2\times10^3$
Fish and Shell	49	2.9 ± 1.2	68 ± 32	190 ± 58	86 ± 63	23 ± 8	$4.7\times10^4\pm8.1\times10^3$
Total	998	20 ± 13	320 ± 120	2300 ± 270	2500 ± 510	6800 ± 5800	$2.1\times10^6\pm3.0\times10^5$

Table 4. Intakes (μg/day) for the Elements in Diet with Some Data for the Comparison

Element	This study ($\pm1\sigma$)	Hamilton et al (9)	ICRP II (11)	U. S. A.	W. Europe	Japan	Russia
Hg	$2.0\times10^1\pm1.3\times10^1$	$<1.6\times10^1$	2.0×10^1	$4.3\times10^1\sim1.1\times10^2$ (35)	$7.0\times10^0\sim1\times10^1$ (9, 10, 20, 29)		
As	$3.2\times10^2\pm1.2\times10^2$	3.4×10^3		$4\times10^2\sim9\times10^2$ (2)	1×10^2 (9)	$7\times10^1\sim1.7\times10^2$ (36)	
Br	$2.3\times10^3\pm2.7\times10^2$	8.4×10^3	1.7×10^4		8×10^3 (9)		8×10^2 (37)
Cu	$2.5\times10^3\pm5.1\times10^2$	3.1×10^3	3.0×10^3				
Mn	$6.8\times10^3\pm5.8\times10^2$	2.7×10^3	3.1×10^3				
K	$2.1\times10^6\pm3.0\times10^5$	2.8×10^3	3.0×10^6				

was substituted for that of seasonings. Furthermore, the data of beef were used instead of those of egg in order to evaluate the daily contribution by the group of meat, egg and milk.

As shown in Table 3, cereals are the main contributor of mercury and bromine and these samples contribute about 60% of the total dietary intake of both elements. Based on the mercury data (20 μg/day) given in Table 3, the average total intake of mercury is estimated to be 140 μg per week. This value is about the half of the amount of 300μg of mercury regarded by the World Health Organization (WHO) and Food and Agriculture Organization (FAO) as the "acceptable weekly dose".[10] The average value ($0.11\pm0.13\mu g$) for the content of mercury in fresh-water fish is about twice that found from marine fish, which has been used for the evaluation of the normal daily contribution in Table 3. If the former value is used, the daily intake of mercury is found to be about $23\pm15\mu g$.

There are relatively much arsenic and bromine in seaweeds as shown in Table 2. The replacement of green vegetables by seaweeds may cause the daily intakes to be $1000\pm340\mu g$ for arsenic and $3100\pm300\mu g$ for bromine.

It is noted that the analytical results of the foodstuffs and the daily intakes of the elements, obtained in this work, are comparable with the corresponding values of other countries given by others as shown in Table 2 and 4, respectively. This suggests a similarity in the mass of elements present in diet although some differences exist between countries.

In this study, the contents of the elements in water and salt used in cooking the foods were not analyzed. Further, no consideration was given to the loss of elements arising from wastage of prepared foods which, for some foods, is likely to amount to 15%.[7] Moreover, the loss of some elements during cooking was not considered in this study.

Acknowledgements

The authors wish to thank Dr. H. J. M. Bowen, Reading University, England, for the presentation of his compiled data and references.

References

1. Mercury Contamination in Man and His Environment, IAEA Technical Report Series No. 137, IAEA, Vienna, 1972.
2. H. A. Schroeder and J. J. Balassa, J. Chron, Dis., **19**, 85-106 (1966)
3. K. Heydorn, Clinica Chimica Acta, **28**, 349-357 (1970)
4. H. Staerk and A. Suess, Proc. of the FAO, IAEA, WHO Sympos. on Comparative Studies of Food and Environmental Contamination (Otaniemi, 1973) p. 417, IAEA, 1974.
5. C. Lee, N. B. Kim and E. B. Park, J. Korean Nucl. Soc., **5**, 279 (1973)
6. C. Lee, N. B. Kim and I. C. Lee, J. Korean Chem. Soc., **19**, 163-168 (1975)
7. C. Lee, N. B. Kim and I. C. Lee, 1974 KAERI Technical Report, Vol. 2, No. 2, KAERI, 1975.
8. National Nutrition Survey, the Ministry of Health and Social Affairs, 1974.
9. E. H. Hamilton and M. J. Minski, The Science of the Total Environment, **1**, 375-394 (1973)
10. R. Schelenz and J. F Diehl, Z. Lebensm. Unters.-Forsh., **151**, 369-375 (1973)
11. ICRP Publication 2, Report of Committee II, Recommendations of the International Commission on Radiological Protection, Pergamon Press, 1959.
12. H. J. M. Bowen, Trace Elements in Biochemistry, Acad. Press (1966)
13. J. J. M. de Goeij and J. P. W. Hontman, Reactor Inst. Delft Rept., 133-71-08 (1971)
14. M. K. Kim, K. D. Woo, S. S. Han, S. H. Lee and U. H. Do, Nongsa Sihom Yongu Pogo, **12**, 55 (1969)
15. T. Fujita, Shiga Kenritsu Eisei Kenkyushoho, **7**, 14 (1968)
16. Y. Takawa, Hokkaidoritsu Eisei Kenkyushoho, No. 19, 117 (1969)
17. C. Tomizawa et al., Shokuhin Eiseigaku Zasshi **7**, 33 (1966)
18. T. Goto and A. Sato, J. Japan Soc. Food Nutrit., **49**, 793 (1966)
19. N. A. Smart and A. R. C. Hill, J. Sci. Food Agric., **19**, 315 (1968)
20. D. C. Abbott and J. O'G. Tatton, Pestic. Sci., **1**, 99 (1970)
21. G. Westoo, Var. Foeda, **18**, 88 (1966)
22. J. Lener, and B. Bibr, J. Agr. Food Chem., **19**, 1011 (1971)
23. G. E. Miller et al., Science, **175**, 1121 (1972)
24. J. B. Rivers, J. E. Pearson and C. D. Schultz, Bull. Envir. Contam. Toxicol., **8**, 257 (1972)
25. L. R. Kamps, R. Carr and H. Miller, Bull. Envir. Contam. Toxicol., **8**, 273 (1972)
26. J. D. Peden, J. H. Crothers, C. E. Waterfall and J. Beasley, Marine Pollut. Bull., **4**, 7 (1973)
27. M. Hussain and E. L. Bleiter, Marine Pollut. Bull., **4**, 44 (1973)
28. A. M. Jones, Y. Jones and W. D. P. Stewart, Nature, **238**, 164 (1972)
29. Ministry of Agriculture, Fisheries and Food,

Survey of Mercury in Food, H. M. S. O. (1971)
30. A. Shapman, Analyst, **51**, 548 (1926)
31. M. R. M. Costa and M. I. C. Da Fonseca, Rev. Portug Farm., **17**, 1 (1967)
32. G. Lunde, J. Sci. Food Agric., **21**, 242 (1970)
33. P. J. LeBlanc and A. L. Jackson, Marine Pollut. Bull., **4**, 88 (1973)
34. A. H. Neufeld, Canad. J. Res. **14B**, 160 (1936)
35. F. M. D'Itri, The Environmental Mercury Problem, Chem. Rubber Co. (1972)
36. M. Nakao, Osaka Shiritsu Daigaku Igaku Zasshi, **9**, 541 (1960)
37. M. G. Kolomiitseva, Gig. Pitan, 121 (1966)