Estimate of toxic element intake in diets of pre-school children and elderly collected by duplicate portion sampling

V. A. Maihara†, M. B. A. Vasconcellos†, M. B. Cordeiro‡ and S. M. F. Cozzolino‡‡

† Radiochemistry Division, IPEN-CNEN/SP, São Paulo SP, Brazil; ‡ ‡ University of São Paulo, Department of Food and Experimental Nutrition-FCF-USP-São Paulo, Brazil

(Received 25 July 1997, revised 27 April 1998, accepted 28 April 1998)

A study was carried out with a group of pre-school children staying at the central nursery of the University of São Paulo and with a group of elderly living in private institutions in São Paulo, Brazil, with the aim of evaluating the contents of toxic elements present in the diets of these groups. For sampling, the duplicate portion technique was used, which consisted of collecting all the foods and beverages consumed during three consecutive days. A radiochemical separation procedure was developed and applied to the determination of As, Cd, Sb, W, Th and U, by means of retention of these elements in the resin Chelex 100 and inorganic exchanger tin dioxide in an appropriate medium. The elements analysed presented levels of ingestion below the maximum levels recommended by the World Health Organization (WHO), and thus can be considered as not presenting any health hazard to the individuals studied in the present work.

Keywords: duplication portion diet, toxic element, neutron activation analysis

Introduction

The dietary intake of toxic elements by humans is influenced by different conditions, such as the geochemical environment, dietary habits, availability of foods, the impact of food processing and the bioavailability of dietary constituents in the foods consumed (Bro *et al.* 1990).

The whole world population is continuously exposed to a variety of contaminants from the environment. Some of these are considered harmful, such as the elements arsenic, cadmium, antimony, thorium, tungsten and uranium and their biological function is not yet clearly known. The study of toxic element intake is of paramount important since the presence of these elements often interferes with the normal biological processes. Their deleterious effects can also alter the metabolism and function of some essential trace elements by competition for ligands in the biological system (Abdulla and Chmielnicka 1990).

The main source of the elements for humans is the food and diet, however, the intake data of these elements by different Brazilian groups is rather scarce.

One of the problems involved in measuring trace element intakes is the difficulty in obtaining reliable information on food consumption. Exact data on dietary intake of trace elements can only be obtained by direct chemical analysis of food and/or diets. It is important to ensure representative sampling and accurate analysis so that comparisons along time and across nations will be meaningful (Bro *et al.* 1990).

There is no simple method that can be employed to estimate the dietary intakes of trace elements in food. Three different methods are useful in estimation of food consumption: the total diet method; selective studies of individual foodstuffs; and the duplicate portion method (FAO/WHO 1985).

This study is intended to provide information on the dietary intakes of As, Cd, Sb, U, Th and W as a means of detecting potential health hazards in two population groups. Little information is available on these elements in Brazilian diets. Certain areas of Brazil have high concentrations of the elements thorium and uranium in soil, and their determination is important in relation to both ratio and chemical toxicity to the human system.

The duplicate portion method has been used to study the trace element intakes for institutional groups or for small surveys, due to the cost and time involved.

0265-203X/98 $12·00

In this kind of survey, which provides the most exact data of trace element intakes, a group of people is asked to provide for analysis an exact duplicate of everything that is eaten and drunk during a period that can vary from 1 to 7 days. The plate with the meal is weighed before consumption, and weighed again at the end of the meal. In this way, the amount of inedible and unconsumed portions is not included. The portions collected are mixed and stored in polyethylene containers and kept in the refrigerator or freezer.

In this study, this sampling method was applied to determine the toxic elements in 19 diets of pre-school children collected for 3 days at the central nursery of the University of São Paulo and in 23 diets of elderly individuals who live in provate institutions in São Paulo. These population groups are considered to be most sensitive to deleterious effects of the toxic elements and were also chosen due to the ease of sampling.

Growing children may be at risk from toxic elements exposure since they consume more food in proportion to body weight than adults. This risk can be evaluated by comparing the actual intake with the provisional tolerable weekly intake (PTWI) or the acceptable daily intake values (ADI), set by the World Health Organization (WHO 1996).

Neutron activation analysis (NAA) can be successfully applied to the determination of toxic trace elements in food samples, due to its sensitivity, precision and accuracy. In the Radiochemistry Division of IPEN/CNEN-SP, it has been used for several years for this procedure after irradiation (RNAA) (Maihara and Vasconcellos, 1988, 1989, Vasconcellos *et al.* 1991, Fávaro *et al.* 1994).

The concentrations of As, Cd, Sb, Th, U and W could be determined through their radionuclides: ^{57}As (half life, $t_{1/2} = 2.63$ h), ^{115}Cd-^{115m} In($t_{1/2} = 53.5$ h+4.5 h); ^{122}Sb ($t_{1/2} = 2.7$ days), ^{233}Pa ($t_{1/2} = 27$ days); ^{239}Np ($t_{1/2} = 2.36$ days and ^{187}W ($t_{1/2} = 23.9$ h), respectively. However, their gamma-ray photopeaks are masked by the activities of the more abundant radionuclides, e.g. ^{24}Na, ^{32}P, ^{82}Br. A radiochemical separation based on the retention of Cd, Sb, Th, U and W in the resin Chelex 100, in 0.1 M HAc–NH_4Ac medium, followed by retention of arsenic in tin dioxide (TDO), in HCl 6 M medium was applied. The procedure has been described in detail elsewhere (Maihara *et al.* 1995).

Experimental

Groups selected

Pre-school children and elderly groups. The group of 19 pre-school children consisted of 12 boys and 7 girls, mean age 5.1 years (range 3.4–6.9 years), weight 19 (13.7–29.7) kg and height 110 (94.5–125) cm. Their parents were employees of the University and were medium income.

Twenty-three healthy elderly (7 men and 16 women) were selected in three private institutions of the urban region of São Paulo. Their mean age was about 84 (range 70–95) years, weight 55 (33–91) kg and height 157 (135–172) cm. These diet samples of the elderly were part of another study whose objective was to verify the dietary adequacy and nutritional status in relation to zinc (Cordeiro 1994).

Duplicate portion technique

Collection and preparation of children's diets. The children were divided into four groups to collect the duplicate portion. Most of them remained for the whole day in the nursery, eating two complete meals (lunch and dinner) and snacks. All samples were collected in the period of September/November 1992. Before the sample collection a meeting was organized with the children's parents that were interested in participating in the study. The purpose of the investigation was explained and the necessary instructions for collection were given. The importance of maintaining the usual eating habits was stressed.

Different polyethylene containers precleaned with 0.1 N HNO_3 and also distilled water were supplied for each family to collect the meals and beverages eaten outside the nursery. Samples were collected for three consecutive days, from the morning of the first day to the night of the third day for each child. During the meal, the duplicate amount of food served for each child was placed on a second plate. The inedible portions (like bone, peel of fruits) were discarded and the equivalent of the food consumed was weighed and put in the polyethylene containers and stored in the refrigerator. Food and beverages were stored separately in polyethylene containers.

After the period of collection for each child, the duplicate portions collected both in the nursery and the house were mixed and homogenized in a domestic blender which was coated with Teflon and equipped with titanium blades. This was necessary to avoid any contamination from metallic parts during this step.

The diets were freeze-dried at 0.1–0.3 mmHg for 24 h and, thereafter, were again homogenized in the blender.

To verify the homogeneity of the samples, the procedure proposed by Kucera and Soukal (1989) for new reference materials was adopted. Instrumental neutron activation analysis was applied to determine the concentration of Na, K and Mn in several aliquots of the same diet sample and the values of the relative standard deviation for these elements were below 3%.

Collection and preparation of diets of the elderly. The diets of the elderly were also collected by the duplicate portion technique, where all food and beverages supplied by the institution were collected during 3 days. The duplicate portions were put onto plates and weighed. Only the edible portions were packed in plastic bags and preserved in a freezer.

The diet samples of the elderly were placed in stainless steel trays and dried in a ventilated oven at 60°C for 12 h. Afterwards the samples were pulverized and homogenized in a knife mill and kept at 4°C before analysis in the Nutrition Laboratory of the FCF-USP.

Radiochemical separation procedure

Samples and standard irradiation. A multielemental standard solution of As (1.6 μg), Cd (8 μg), Sb (0.4 μg), Th (0.78 μg), and W (1.6 μg) from primary standard solutions from British Drug House (UK) was prepared.

The diet samples (150 mg) and the standard solution were sealed in clean quartz ampoules for irradiation for 8 h under a thermal neutron flux of 10^{13} n cm^{-2}s^{-1} in the IEA-R1 nuclear research reactor.

Analytical procedure. After a cooling time of 2 days, the diet samples were transferred to Teflon digestion bombs (Parr bombs) and dissolved in HNO_3/HF (5:1) at 150°C for 10 h. An aliquot of 0.5 ml of the non-irradiated multielemental standard solution was used as carrier. After dissolution, the resulting solutions were brought to dryness by evaporation at 60–70°C to avoid possible losses by volatilization and the residues were redissolved in 10 ml buffer solution (0.1 M ammonium acetate at pH 4.8). Thereafter the solutions were percolated through the Chelex and TDO columns. The Chelex 100 resin was previously conditioned with 0.1 M ammonium acetate buffer and the TDO inorganic exchanger was conditioned with 6 M HCl. During the percolation through the Chelex columns the TDO was maintained at a strong acidity by adding dropwise 6 M HCl directly onto the TDO columns. The Chelex columns were then washed with 100 ml of buffer solution using the same operating procedure. In these conditions the elements Cd, Sb, Th, U and W were retained in the Chelex resin while As was absorbed on the TDO column.

The gamma-ray spectra were obtained in an ORTEC EG&G high resolution solid state Ge detector, type POP TOP, Model 20190 with resolution of 1.9 keV for the 1332 keV γ-ray peak of ^{60}Co. This detector was coupled to an EG & G ORTEC ACE8K card and associated electronics. The spectra were analysed using the VISPECT2 software.

Results and discussion

Quality assurance

The analytical method applied in this work was tested by analysis of certified reference materials SRM 1515 Apple Leaves and SRM 1547 Peach Leaves. The recovery for the elements As, Cd, Sb, Th, U and W obtained by radioactive tracers was high (above 95%, except for arsenium) and published elsewhere (Maihara *et al.* 1995). The results obtained showed good agreement with the certified values (table 1). For uranium and thorium the reference materials present only informative values.

The detection and determination limit values for these elements were determined by the Currie criterion (1968) in experimental conditions and are presented in table 2.

Table 1. Results of determination of toxic elements in certified reference materials (ng/g)

Element	Peach Leaves SRM 1547			Apple Leaves SRM 1515		
	This work ($n = 3$)	Becker (1992)	Certified value	This work ($n = 3$)	Becker (1992)	Certified value
As	67 ± 7	66.6 ± 2.0	60 ± 18	40 ± 5	35.9 ± 1.7	(38)
Cd	28 ± 5	27.3 ± 0.3	(30)	14 ± 2	13.8 ± 0.9	(14)
Sb	14.6 ± 1.4	24.6 ± 0.5	(20)	nd	12.6 ± 0.9	(13)
Th	50.3 ± 6.7	na	(50)	34.0 ± 4.6	na	(30)
U	15.7 ± 1.8	na	(15)	9.9 ± 0.6	na	(6)
W	23.8 ± 3.3	na	–	8.0 ± 2.3	na	—

n: number of determinations.
na: not analysed.
nd: not determined.
( ): informative values.

Table 2. Limits of detection and determination for the toxic elements analysed by radiochemical neutron activation analysis.

Element	Detection limit (ng/g)	Determination limit (ng/g)
As	1.8	6.1
Cd	4.7	25
Sb	0.28	1.7
U	1.8	4.1
Th	1.6	5.0
W	2.0	6.3

Dietary intake estimates

The diet samples analysed consisted of all foods which an individual ate during three consecutive days. The duplicate diets included solid food and beverages but did not include drinking water. In the diets of the elderly the consumption of foodstuffs of vegetal origin was high, while the milk and derivatives comprised the main group of the children's diets.

The concentrations of toxic elements in these samples were determined (table 3) and the daily intakes were obtained by multiplying the concentrations in individual diet samples by the total weight of food consumed by each participant. The average amount of food consumed daily was about 800 g. The mean daily intakes (in µg per person) of each element were obtained by adding together the daily intake calculated for each individual and dividing by the number of participants of the study (19 children and 23 elderly persons).

The range of dietary intakes, the arithmetic mean and median values with the standard deviation (SD) of the arithmetic mean for the diets of the children and elderly are reported in table 4.

The elements antimony, arsenic, thorium, uranium and tungsten do not play any essential role in the human system and cadmium is considered toxic even at low concentration. The data obtained are discussed briefly for each element individually.

Antimony. Antimony is emitted in the environment by various industrial processes and has no known functions in living organisms. It has a low inherent toxicity. The FDA set 2 µg/g as the maximum tolerable limit in foods (Iffland 1988).

In this study the daily dietary intake of antimony was of the order of 1.1–2.3 µg. The data are slightly less than those reported by Fávaro *et al.* (1997) in the four regional Brazilian diets, the values of which ranged between 3.3 and 6.3 µg/day. Mannan *et al.* (1992) determined 1.29 and 1.61 µg/person in Pakistan diets.

Arsenic. The carcinogenic effects of inorganic arsenic compounds in human beings has long been known. In recent years, however, some studies have shown evidence of the essentiality of arsenic compounds for growth of several animals (Nielsen 1990).

Arsenic contamination in foods is caused mainly by indiscriminate use of arsenic compounds as insecti-

Table 3. Results of analysis of diets of children and the elderly by radiochemical separation—RNAA (dry weight).

Element	Children's diet (ng/g)			Diet of the elderly (ng/g)		
	Average ± SD (*n*)	Range	Median	Average ± SD (*n*)	Range	Median
As	61.6. ± 41.4 (19)	9.3–161	64.7	31.3 ± 37.4 (15)	2.3–128	17.5
Cd	20.8 ± 18.8 (18)	6.1–8.26	14.5	11.2 ± 8.5 (15)	5.8–27.0	7.0
Sb	5.3 ± 5.1 (19)	1.2–19.4	3.7	11.0 ± 10.2 (15)	2.4–37.0	7.0
U	6.7 ± 5.0 (18)	1.8–21.7	5.4	4.1 ± 3.9 (15)	1.1–16.5	3.5
Th	14.0 ± 22.3 (14)	1.6–87.8	6.1	6.0 ± 4.5 (15)	1.3–13.9	4.5
W	34.6 ± 37.1 (19)	3.4–166	29.7	48.2 ± 35.6 (15)	5.6–139	41.3

n: number of analysed samples.

Table 4. Daily intakes of As, Cd, Sb, U, Th and W of the diets of children and the elderly.

Element	Children's diet (μg/day)			Diet of the elderly (μg/day)		
	Average ± SD	Range	Median	Average ± SD	Range	Median
As	12.4. ± 9.0	1.6–37	12.1	6.9 ± 8.6	0.62–26.5	3.6
Cd	4.1 ± 3.9	1.4–17	3.0	2.7 ± 2.2	1.2–6.9	1.9
Sb	1.1 ± 1.1	0.2–4.5	0.73	2.3 ± 2.0	0.49–7.4	1.7
U	1.4 ± 1.1	0.4–4.5	1.1	0.81 ± 0.58	0.22–2.3	0.73
Th	2.6 ± 3.7	0.3–14	1.1	1.2 ± 0.9	0.3–3.0	0.88
W	6.7 ± 6.7	0.7–29	5.4	9.9 ± 7.0	1.5–26	7.1

cides, herbicides, fungicides and food additives. The content of seafood in the diet also influences greatly the amount of arsenic ingested daily by humans.

There are few available data about daily intake of arsenic in Brazilian diets. The dietary intakes of arsenic determined in this study are below the PTWI value set by WHO, which is 15 μg/kg body weight. For the children's diets in the present work the values varied from 1.6 to 37 μg/day, with a mean daily intake of 12.4 μg/day. The levels in the diets of the elderly were lower, and the values varied from 0.6 to 26.5 μg/day, with a median value of 3.6 μg/day. These values were lower than reported by Fávaro *et al.* (1997) in another Brazilian diet. The daily intake range determined was from 18.7 to 159.3 μg/day in four regional Brazilian diets. The highest level was obtained in diets with elevated consumption of fish, meat and poultry. Buchet *et al.* (1983) also reported a large variation in the diets of Belgian women (from 0.1 to 720 g As/day).

Cadmium. Anaemia is a common manifestation of chronic cadmium toxicity in all species, due at least in part to its metabolic antagonism to copper and iron. Cadmium metabolism is strongly influenced by the dietary intakes of other elements, such as zinc, copper, iron and selenium.

In this study the daily dietary intakes of cadmium determined were much lower than the PTWI value set by WHO/FAO (7 μg/kg body weight). About 70% of the values obtained in the diets of children and the diets of the elderly were lower than 5 μg/day. These values reported in this study are lower than those determined by Fávaro *et al.* (1997), which ranged from 7.8 to 31.5 μg/day.

The daily dietary intake of cadmium is variable in different regions. It has been estimated at 43 g/day in China (Chen *et al.* 1994), 3–102 μg/day in Denmark (Bro *et al.* 1990) and 5.3–17.1 g/day in Spain (Dabeka and McKenzie 1992).

Thorium. Thorium absorption by the human organism is from soil inhalation or from foods. The ^{232}Th isotope is one of the most important natural radionuclides which is used in the production of nuclear energy. The daily exposure of this element was estimated by the International Commission on Radiological Protection (ICRP) at 3 μg (ICRP 1975). Higher values were obtained by Dang *et al.* (1992) in Indian diets from different

regions (range from 8.6 to 62.4 µg/day). Shiraishi and Yamamoto (1995) determined the value of 0.55 µg/person in basic foods in Japan. The average values obtained in this study (2.6 µg/day and 0.88 µg/day) agree with the estimate given by ICRP.

Tungsten. Little is known about the toxicity of tungsten compounds, although the LD_{50} of soluble salts in the rat is relatively high. Its metabolism is related to molybdenum which is closely similar in its chemical properties (Kazantzis 1986). There is practically no information about the dietary intake of tungsten. The estimated daily intakes for this element varied from 0.7 to 29 µg and from 1.6 to 26 µg, respectively, in the diets of children and the elderly.

Uranium. Uranium is one of the primordial radioactive elements distributed widely in the earth's crust. As all uranium isotopes in nature are radioactive, the harmful effects of the high intake of this element are increased. The hazards of a high intake of uranium are two-fold—chemical toxicity and radiological damage (Berlin and Rudell 1986).

The ICRP stated that the kidney is the critical organ for uranium exposure, and that the maximum permissible intakes should be limited to 25 mg/day by inhalation and 150 mg in two consecutive days by ingestion. The total dietary intake of uranium has been estimated at 1.9 µg/day by ICRP (1975).

Daily intake of uranium in Japanese urban areas has been estimated at 1.25 µg/day by Shiraishi and Yamamoto (1995). According to Yamamoto *et al.*(1994), the dietary intake levels of U in normal background areas ranged widely from 0.56 to 7.9 µg/day. Daily dietary intakes of both the children and the elderly were determined to be in this range, whose average values were 1.4 µg/day and 0.81 µg/day.

Conclusion

This paper reports on a sensitive and reliable method for the analysis of As, Cd, Sb, Th, U and W in diet samples. The radiochemical separation procedure allowed the simultaneous determination of these elements in the range of parts per billion in the diets of children and the elderly.

From the results presented it can be concluded that the daily dietary intakes of arsenic (range from 0.62 to 37 µg/day) and cadmium (from 1.2 to 17 µg/day) are below the tolerable limits set by WHO/FAO. For the natural radionuclides thorium and uranium the estimated intakes (median of 1.1 µg U/day and 1.1 µg Th/day) are similar to those reported in other normal background regions. Daily dietary intakes from 0.2 to 7.4 µg/day for antimony and from 0.7 to 29 g/day for tungsten were found in this study.

References

Abdulla, M., and Chmielnicka, J., 1990, New aspects on the distribution and metabolism of essential trace elements after dietary exposure to toxic metals. *Biological Trace Element Research*, **23**, 25–53.

Becker, D. A., Greenberg, R. R., and Stone, S. F., 1992, The use of high accuracy NAA for the certification of NIST botanical standard reference materials. *Journal of Radioanalytical and Nuclear Chemistry*, **160**(1), 41–53.

Berlin, M., and Rudell, M., 1986, Uranium. *Handbook on the Toxicology of Metals*, edited by L. Friberg, G. F. Dorberg and V. B. Vouk (New York: Elsevier), volume 2, pp. 623–637.

Bro, S., Sandstrom, B., and Heydorn, K., 1990, Intake of essential and toxic trace elements in a random sample of Danish men as determined by the duplicate portion sampling technique. *Journal of Trace Elemental of Electrolytes Health Disease*, **4**, 147–155.

Buchet, J. P., and Lauwerys, R., 1993, Oral daily intake of cadmium, lead, manganese, copper, chromium, mercury, calcium, zinc and arsenic in Belgium: a duplicate meal study. *Food and Chemical Toxicology*, **21**, 19–24.

Chen, F., Cole, P., Wen, L., Mi, Z., and Trapido, E. J., 1994, Estimates of trace element intakes in Chinese farmers. *Journal of Nutrition*, **124**, 196–201.

Cordeiro, M. B., 1994, Adequação alimentar e avaliação do estado nutricional em relação zinco em grupos de idosos institucionalizados. Dissertação de Mestrado. Universidade de São Paulo.

Currie, L. A., 1968, Limits for qualitative detection and quantitative determination. *Analytical Chemistry*, **40**, 586–592.

Dabeka, R. W., and McKenzie, A. D., 1992, Diet study of lead and cadmium in food composites: Preliminary investigation. *Journal of AOAC International*, **75**, 386–395.

Dang, H. S., Pullat, V. R., and Pillai, K. C., 1992, Simultaneous determination of ^{232}Th and ^{238}U in biological samples. Application to the estimation of their daily intake through diet. *Journal of Radioanalytical and Nuclear Chemistry*, **162**, 163–169.

Fávaro, D. I. T., Maihara, V. A., Armelin, M. J. A., Vasconcellos, M. B. A., and Cozzolino, S. M., 1994, Determination of As, Cd, Cr, Cu, V, Hg, Sb and Se concentrations by radiochemical neutron activation analysis in different Brazilian region diets. *Journal of Radioanalytical and Nuclear Chemistry, Articles*, **18**, 385–394.

Fávaro, D. I. T., Hui, M. L. T., Cozzolino, S. M. F., Maihara, V. A., Armelin, M. J. A., Vasconcellos, M. B. A., Yuyama, L. K., Boaventura, G. T., and Tramonte, V. L., 1997, Determination of various nutrients and toxic-elements in dif-

ferent Brazilian regional diets by neutron activation analysis. *Journal of Trace Elements in Medicine and Biology*, **11**, 129–136.

FAO/WHO, 1985, *Guidelines for the Study of Dietary Intakes of Chemical Contaminants* (Geneva: WHO), Offset Publication no. 87.

KAZANTZIS, G., 1986, Tungsten. *Handbook on the Toxicology of metals*, edited by L. Friberg, G. F. Dordberg and V. B. Vouk (New York: Elsevier), Volume 2, pp. 610–622.

KUCERA, J., and SOUKAL, L., 1989, Homogeneity tests and certification analyses of the Irant coal fly ash reference material ECO by instrumental neutron activation analysis *Journal of Radioanalytical and Nuclear Chemistry*, **134**, 209–219.

ICRP, 1975, *Report of the Task Group on Reference Man*, Report 23, International Commission on Radiological Protection (Oxford: Pergamon).

IFFLAND, R., 1988, Antimony. *Handbook on Toxicity of Inorganic Compounds*, edited by H. G. Seller and H. Siegel (New York: Marcel Dekker, Inc), pp. 67–75.

MAIHARA, V. A., and VASCONCELLOS, M. B. A., 1988, Multielement analysis f Brazilian milk powder and bread samples by neutron activation. *Journal of Radioanalytical and Nuclear Chemistry*, **122**, 161–173.

MAIHARA, V. A., and VASCONCELLOS, M. B. A., 1989, Determination of trace elements in Brazilian rice grains and biological materials by neutron activation. *Journal of Radioanalytical and Nuclear Chemistry*, **132**, 329–337.

MAIHARA, V. A., GALLORINI, M., and VASCONCELLOS, M. B. A., 1995, Radiochemical separation for the certification of some trace elements in biological reference materials by neutron activation analysis. *Journal of Radioanalytical and Nuclear Chemistry*, **198**, 343–348.

MANNAN, A., WAHUD, S., AHMAD, S., and QUERSHI, I. H., 1992, Dietary evaluation of toxic elements through integrated diets. *Journal of Radioanalytical and Nuclear Chemistry*, **162**, 11–123.

NIELSEN, F. H., 1990, New essential trace elements for the life sciences. *Nuclear Analytical Methods in the Life Sciences*, edited by R. Zeisler and V. P. Guinn (New Jersey: Human Press), pp. 599–611.

SHIRAISHI, K., and YAMAMOTO, M., 1995, Dietary ^{232}Th and ^{238}U intakes for Japanese as obtained in a market basket study and contributions of imported foods to internal doses. *Journal of Radioanalytical and Nuclear Chemistry, Articles*, **196**, 89–96.

VASCONCELLOS, M. B. A., MAIHARA, V. A., FÁVARO, D. I. T., ARMELIN, M. J. A., CORTES-TORO, E., and ORGRIS, R., 1991, Radiochemical separation methods for the determination of some toxic elements in biological reference materials. *Journal of Radioanalytical and Nuclear Chemistry, Letters*, **153**, 185–199.

WORLD HEALTH ORGANIZATION (WHO), 1996, *Codex General Standard for Contaminants and Toxins in Foods* (The Netherlands: Codex Alimentarius Commission (CX/FAC 96/17).

YAMAMOTO, M., SHIRAISHI, K., KOMURA, K., and UENO, K., 1994, Measurement of uranium in total diet samples: daily intake for Japanese. *Journal of Radioanalytical and Nuclear Chemistry, Articles*, **185**, 183–192.