Chapter 31
Total Diet Studies in Japan

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Introduction

Two methods exist for directly assessing exposure to chemicals in food as consumed, namely by the total diet method and duplicate diet method. A total diet study (TDS) is suitable for assessment of contaminants and other chemical in populations, especially where individual food consumption data are available. Such data are useful for identifying high-risk populations and for risk management and surveillance planning. On the other hand, duplicate diet methods are less expensive and yield quicker results. While they include factors of real cooking and more precise portion size of the meals, the results of duplicate diet studies are often limited to the local area and cannot be used to estimate exposure of sex and age groups. The TDS based on market

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basket samples is greatly dependent both on surveillance data of contaminants in foods and food consumption data. In Japan, food consumption data used in the TDS have been obtained from the annual National Nutrition Survey (NNS).

National Nutrition Survey

After World War II, food shortages in Japan were very serious. General Headquarters of the Allied Powers conducted the first NNS in Japan in 1945 to assess the need for food aid from overseas. Food consumption of 6,000 families in Tokyo was monitored in December 1945. The purpose of the survey was to improve the management of the food supply to solve food shortages in the upheaval of the postwar period. In 1946, the survey was expanded to include 9 cities, 27 prefectures, and 4 mining areas. In 1948, a nationwide survey was conducted with study areas selected on the basis of a population-weighted random sampling method. The survey was modified in 1952 based on the Nutrition Improvement Law. Due to the rapid recovery of the Japanese economy, the new survey parameters, such as smoking, drinking and exercise habits, were added to the questionnaire.

Current NSS in Japan is conducted in November each year. First, 300 study areas are selected randomly and then 6,000 families or 20,000 individuals in the areas are asked to participate the survey. Participants of NSS are asked how much food was consumed on a certain day in November by the family and are requested to fill out an allocation table of food items consumed by each family member. A food composition table is used to calculate the intake amounts of energy and nutrients recorded on the sheets.

Method of Total Diet Study in Japan

The Ministry of Health, Labor, and Welfare (MHLW) has conducted total diet studies in Japan annually since 1977. Based on the methods of the Global Environmental Monitoring System/Food Contamination Monitoring and Assessment Programme (GEMS/Food), the TDS is used to estimate average Japanese dietary exposure to heavy metals and various chemical substances, such as pesticides, dioxins, food additives, and other potentially hazardous chemicals. The Division of Foods under the National Institute of Health Sciences (NIHS) in Tokyo orchestrates TDS in Japan. Twelve survey districts for the TDS are selected based on a population-size-weighed random sampling method. Food items are categorized into 14 groups (see Table 31.1). Samples of between 100 and 120 different food items are purchased at local markets in each sampling site and prepared as for consumption, i.e. 'as consumed'. Composite samples for each food category are prepared in proportion to regional food frequency questionnaire data collected during the NNS. The composite samples are sent to NIHS where concentrations of contaminants in the samples are measured.

Table 31.1 Food categories for Japanese total diet study

Category	Food items
1	Rice and processed rice products
2	Wheat, barley, rye, buckwheat and other cereals
3	Sugar and sweets, confectionary products
4	Oil and fats
5	Beans and bean products
6	Fruit
7	Green and yellow vegetables
8	Other vegetables and seaweed
9	Seasoning and condiments
10	Fish
11	Meat and eggs
12	Milk and dairy products
13	Processed foods
14	Water

The chemicals analyzed at NIHS are pesticides, including three isomers of hexachlorohexanes, four DDT analogues, three organophosphates, three other organochlorines and certain polychlorinated biphenyls (PCBs), and seven metals, namely cadmium, mercury, arsenic, lead, copper, manganese, and zinc. These are measured in 13 composite food samples from each sampling location.

Strengths and Limitations of the Current Total Diet Study

Food consumption data derived from the NNS conducted on one day in November annually consists of food items (1,194 food codes) and weights of each food ingredients collected from approximately 5,000 families, randomly sampled in all over the nation. The number of individuals included in the NNS is approximately 13,000 annually. Individual physical data, such as height and weight, physical activities and ratio of food consumption relative to the family members have been collected since 1995. This modification of the NNS enabled food consumption estimates to be made for 10 sex-age groups. Note that data on intakes by pregnant women were excluded to improve the precision of these estimates.

There are several limitations in using the current NNS data for TDS. Because the survey is based on 1-day study in November, it is impossible to evaluate seasonal changes of food items and individual daily variations of food intake. It is also impossible to evaluate intake assessment of sensitive subpopulations or high-risk groups. The family-based NNS has only limited information on dining-out as well as on composite and processed foods.

To partially address these limitations, NNS data covering several years are compiled for use of TDS exposure assessments of contaminants, such as cadmium, dioxins and dioxin-like PCBs and pesticide residues. Epidemiological studies among high-risk subpopulations have been conducted in the several areas of Japan. At the same time, 150 pesticides were monitored in food for preparation before adopting a positive list system of residual pesticides in food in 2006.

Heavy and Other Metal Exposures from the Nationwide TDS

The average exposure to heavy metals, such as mercury, cadmium, lead and arsenic, was calculated based on the analysis of stored composite samples from the 2000, 2002, and 2004 TDSs [1]. The average intake of total mercury in the three TDSs was 8.9 μg/day, ranging 7.3–10.5 μg/day, which corresponds approximately to 38–55 % of PTWI. Exposure to total mercury was derived mainly from fish (Group 10) and rice (Group 1), but it should be noted that rice contains mainly inorganic mercury, as reported in early GEMS/Food data.

Average cadmium exposure ranged from 32.4 to 48.2 μg/day; the average is 42.0 μg/day; this corresponded to 65–96 % of PTWI. Cadmium exposure was derived from almost all the food categories. Cadmium intakes from rice (Group 1) and Groups 8 and 10 were fairly high. Average exposure from Group 1 was 0.033 mg/ day.

Average lead exposure ranged from 33.8 to 50.3 μg/day, which were 19–28 % of former PTWI (now withdrawn), and the average was 42.5 μg/day, lead from rice consist 50–60 % of total intake. Arsenic exposure ranged from 181 up to 350 μg/ day, equivalent to 7–15 % of former PTWI (now withdrawn), with an average of 243 μg/day. Arsenic is found in seafood, including seaweeds and fish and shellfish, with the major sources of arsenic were mainly from Groups 8 and 10.

Judging from longitudinal study of total diet study [2], cadmium exposures calculated from the data showed no changes in the last decade. In 2004, average cadmium exposure was 21.4 μg/day or 46 % of PTWI. Total mercury exposure is 8.5 μg/day is more than 50 % of the current PTWI of 1.6 μg/kg bw/week for methyl mercury. Arsenic exposure is high among the Japanese due to their higher intakes of marine products. However, there are some technical issues in extraction and measurement of chemical forms of organic arsenic in various food matrixes.

Arsenic Intake from Algae

Arsenic is ubiquitous in soil and sea sediments. It accumulates in food in varied concentrations and in several chemical forms. The most important from a toxicological point of view is inorganic arsenic compounds, such as arsenic trioxide, sodium arsenite, arsenic trichloride (i.e. trivalent forms), arsenic pentoxide, arsenic acid and arsenites (i.e. pentavalent forms). But marine organisms are well adapted to tolerate the metal and to accumulate arsenic as organic compounds. The major organic arsenic compounds are arsenobetaine, a water-soluble arsenical found in most of seafood, such as fish and shrimps. In contrast, the major organic arsenic compounds in algae are arsenosugars. These organic arsenic compounds are less toxic than inorganic arsenic compounds. For risk assessment purposes, it is very important to collect concentration data on both organic and inorganic forms. Total diet studies provide very important information on total arsenic and inorganic arsenic exposures for risk assessment.

The Japanese people consume several kinds of edible species of algae such as "nori" or *Porphyra*, including most notably *P. yezoensis* and *P. tenera*, "kombu" or *Saccharina japonica* (*Laminaria japonica*), "wakame" or *Undaria pinnatifida*, "hijiki" or *Hizikia fusiforme,* and "mozuku" or *Cladosiphon okamuranus*. Most of these species contain arsenosugars, which are recognized as essentially nontoxic. But hijiki contains inorganic arsenic and is consumed regularly in Japanese dishes. This raised a question about whether dietary intake of hijiki is safe or not.

Matsuda and Watanabe [2, 3] reported the monitoring data of arsenic contents in three food groups as major sources of arsenic exposure in the Japanese diet. They are rice (Group 1) and composite samples of vegetables and seaweed (Group 8) and fish, cephalopods, and shellfish (Group 10). The 10 rice samples, collected from 10 districts all over Japan, were cooked and homogenized by food processors. The composite samples from the 10 different districts were used for determination of arsenic contents. Trivalent and pentavalent inorganic arsenic concentrations in the samples were determined by high performance liquid chromatography with inductively coupled plasma mass spectrometry (HPLC-ICPMS) by the method of Hamano-Nagaoka et al. [3, 4] Total arsenic contents were measured by atomic absorption spectrophotometry. Total and inorganic arsenic exposure was assessed by multiplication of the concentrations and food consumption data from NNS.

The results of the study revealed that total arsenic exposure was 245.7 μg/day, when zero was applied for non-detected (ND) samples, or 248.2 μg/day, when half the limit of quantification (LOQ) was used for ND samples. Inorganic arsenic intake was 15.7 μg/day (ND=0), and 30.4 μg/day (ND=½ LOQ). When units are converted using 50 kg for the average body weight, these values for inorganic arsenic were 14.7 % or 28.4 % of withdrawn PTWI. Contributions to total arsenic exposure from Groups 1, 8, and 10 were 6 %, 31 % and 59 %, respectively. On the other hand, inorganic arsenic exposure from Group 1, 8, and 10 were 42 %, 58 %, and 0 % (when ND=0), or 37 %, 47 %, and 16 % (when ND=½ LOQ). Even though the exposure is lower than the withdrawn PTWI, inorganic arsenic exposure from rice, as a staple food, is unexpectedly high, and dietary habit of eating certain algae elevates inorganic arsenic exposure in the Japanese diet.

Probabilistic Exposure Assessment on Cadmium Among Japanese by Monte Carlo Simulation

In 2005, the MHLW commissioned a probabilistic assessment of cadmium intake among the Japanese population. Datasets of cadmium concentrations in food were converted from surveillance data of cadmium in agricultural and fishery products

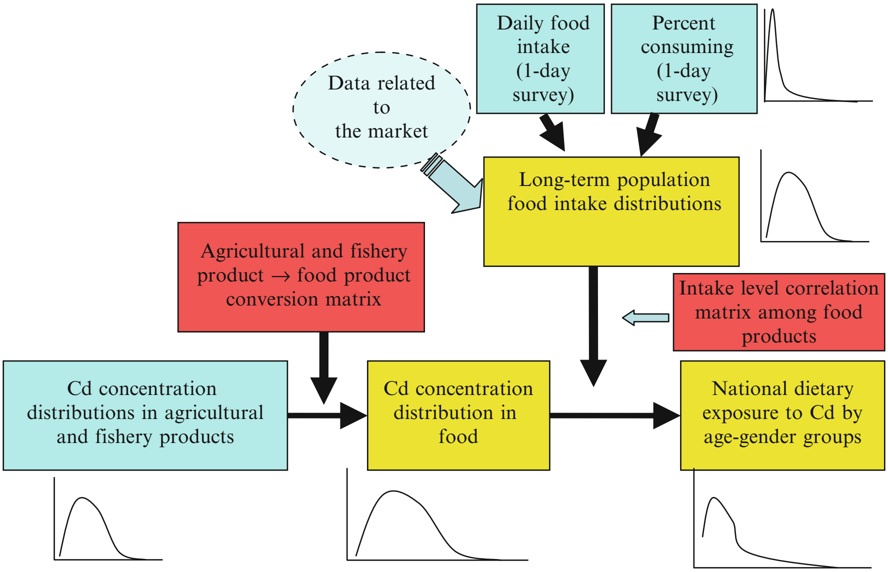

Fig. 31.1 Outline of the procedure for estimating exposure to cadmium in Japan

conducted by the Ministry of Agriculture, Forestry, and Fisheries (MAFF). As most of soybean and wheat consumed in Japan is imported, cadmium concentration data in soybean and wheat imported from the USA was used. Japanese Food Balance Sheets were used to estimate the consumption ratios of domestically produced and imported soybeans and wheat.

Average food intakes for the various food groups were obtained from the NNS database for the years 1995–2000. Age, sex, and body weight data was also obtained, but such data excluded individuals younger than 20 years and pregnant women in order to improve the precision of data. Data for approximately 53,000 adults were used for this analysis.

The outline of the procedure for estimating exposure to cadmium by age-sex groups in Japan is shown in Fig. 31.1. Although it is preferable to determine the mean long-term intake of food, 1-day data from the NNS were used without further treatment in the analyses. As food groups are interrelated in ordinary dietary habits, the following rank correlations were assumed: –0.32 for rice-wheat, 0.22 for rice-soybean, and –0.1 for wheat-soybean.

Approximately 36,000 cadmium concentration values in 130 agricultural and fishery products were obtained from the surveillance of cadmium concentrations performed by MAFF. However, as there was no one-to-one correspondence between these 130 surveillance items by MAFF and 1,000 food items included in NSS database by MHLW, cadmium concentrations of agricultural products were applied to grouped food categories for these calculations. In addition, previously determined coefficients for the retention of residual chemicals in food during cooking and food processing were used [5]. A conversion coefficient of 1 was applied for all fishery products not indicated here on the assumption that cadmium remains in the fishery products after cooking and processing. The intake amounts of food groups were calculated by summing up values obtained through multiplication by the conversion coefficients.

The estimation of cadmium exposure distributions and factors involved in the estimations were examined by Monte Carlo simulations using the Japanese version of *Crystal Ball 2000*. The intake distributions of about 90 food groups were multiplied by the distributions of the cadmium concentrations in the agricultural products contained in the food groups. In practice, random numbers satisfying the distributions of the presence or absence of consumption, the amount of consumption (eaters only) and cadmium concentrations were generated; the product of the three random numbers is the cadmium exposure. The operation was repeated many times to estimate cadmium intake distribution. A binary distribution with 1 and 0 was assumed for the presence and absence of consumption; the expectation of the ratio of those whose consumption of a food group was not a zero in the NNS was set at 1 in the binary distribution. Lognormal distribution was presumed for the theoretical distribution of intake levels and cadmium concentrations. Distribution properties were determined based on the parameters estimated from mean values as well as standard deviations obtained by the NNS and the surveillance of cadmium in food.

Table 31.2 shows the different cadmium concentrations used in the three scenarios in the Monte Carlo simulation. In the scenarios, it is assumed that the food items containing cadmium concentrations at or exceeding the maximum level would be excluded from the food supply. By selecting the different cadmium concentrations in each scenario, Monte Carlo estimations were made by excluding any random numbers exceeding the maximum levels for the food. For food items fixed at median values due to their limited surveillance data, medians were calculated by excluding samples exceeding the set maximum levels.

Table 31.3 depicts the arithmetic means and values of cadmium intake at 25th, 50th, 75th, 90th, 95th, and 97.5th percentiles of cadmium exposure estimated in setting three different scenarios. By setting different maximum levels of cadmium in food, there were no large differences in distributions and the values at the 97.5th percentile, which in all the scenarios were above 7 μg/kg bw/week.

Contaminant Priorities for Surveillance in Food in Japan

Staple food and traditional cuisine among Japanese people consist of rice and many marine products. These dietary factors are thought to be beneficial for Japanese longevity. However, they also contribute to much higher exposures to cadmium, arsenic, methylmercury, PCBs, dioxins, and dibenzofurans. This may be called the "Japanese paradox". As a consequence, the Ministry of Health, Labor, and Welfare established a priority list for the surveillance for these contaminants, which will be included in future TDSs. These are shown in Table 31.4. Dioxins and dioxin-like PCBs are also prioritized contaminants especially in fish and other seafood as well as in agricultural products and meat and dairy products in Japan. Cadmium is one of the most

Table 31.2 Distribution maximum values for three Monte Carlo scenarios

Item	Scenario (when data higher than or equal to each value were omitted) values in mg/kg		
	1	2	3
Cereals			
Polished rice	0.3	0.4	0.5
Wheat	0.2	0.2	0.2
Cereals other than rice and wheat	0.1	0.1	0.1
Beans (matured)			
Soybean	–	–	–
Beans other than soybean	0.1	0.1	0.1
Stem and root vegetables			
Burdock	0.1	0.1	0.1
Taro	0.1	0.1	0.1
Potato	0.1	0.1	0.1
Celeriac	–	–	–
Other than burdock, taro, and potato	0.1	0.1	0.1
Leafy vegetables			
Spinach	0.2	0.2	0.2
Other than spinach	0.2	0.2	0.2
Bulb vegetables (Alliums)			
Garlic	0.05	0.05	0.05
Other than garlic	0.05	0.05	0.05
Non-cucurbitaceous fruits and vegetables (including mushrooms and sweet corn)			
Eggplant	0.05	0.05	0.05
Okra	0.05	0.05	0.05
Tomato	–	–	–
Mushroom	–	–	–
Other than tomato, eggplant, and okra	0.05	0.05	0.05
Stalk vegetables			
Stalk vegetables	0.1	0.1	0.1
Cress (bulb-forming vegetables)			
Cress	0.05	0.05	0.05
Cucurbitaceous fruits and vegetables			
Cucurbitaceous fruits and vegetables	0.05	0.05	0.05
Beans and peas (immatured)			
Fabaceous vegetables	0.1	0.1	0.1
Peanut			
Peanut	–	–	–
Fruits			
Fruits	–	–	–
Mollusks (including cephalopods)			
Mollusks	1	1	1
Herbs			
Herbs	–	–	–

Table 31.3 Exposure to cadmium under three Monte Carlo scenarios

	Scenario 1	Scenario 2	Scenario 3
Arithmetic mean	3.29	3.33	3.35
25 percentile	2.10	2.10	2.11
50 percentile	2.85	2.86	2.86
75 percentile	3.94	3.97	3.98
90 percentile	5.45	5.54	5.57
95 percentile	6.67	6.85	6.93
97.5 percentile	8.01	8.32	8.46

Unit: μg/kg-bw/week

Table 31.4 Priority list of contaminants in food for total diet studies in Japan

Contaminants	Food items
PCDD/PCDFs and dioxin-like PCBs	Agricultural products
PCDD/PCDFs and dioxin-like PCBs	Meat and dairy products
PCDD/PCDFs and dioxin-like PCBs	Fish and other seafood
PCDD/PCDFs and dioxin-like PCBs	Feed
Deoxynivalenol and nivalenol	Agricultural products
Ochratoxin A	Agricultural products
Zearalenone	Agricultural products
Acrylamide	Processed foods
3-Methylchloropropane-1,2-diol-esters	Processed foods
1,3-Dimethlychloropropane	Processed foods
Cadmium	Rice, feed
Aflatoxin	Feed
Pesticides	16 major agricultural products
Pesticides	Imported foods on positive list
Pesticides	Feed

prioritized heavy metals in rice and feed. As for mycotoxins, aflatoxin, ochratoxin A, and zearalenone have been important contaminants in feed and agricultural products. In addition, mycotoxins from *Fusarium* fungi, such as deoxynivalenol and nivalenol, have been included as monitoring contaminants because of humid weather often occurs during the harvesting of barley and wheat. Contaminants recently evaluated in JECFA meeting, such as chloropropanols and acrylamide, are rather low in food, but these chemicals are also included in monitoring in response to changes of dietary habits of the younger generations, which includes increased consumption of imported processed foods.

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