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Average Daily Intake of Pesticides and Polychlorinated Biphenyls in Total Diet Samples in Osaka, Japan

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Total diet studies (also referred to as Market Basket Programs) have been conducted by the U.S. Food and Drug Administration (FDA) since 1964. The recent studies were introduced by Johnson et al. (1984), Podrebarac (1984) and Gartrell et al. (1985a, b). In Japan, similar studies have been carried out since 1977 by the National Institute of Hygienic Sciences in combination with some Prefectural Institutes of Public Health including ours (Maeda et al. 1979, 1980; Murakami et al. 1981, 1983; Kuwabara et al. 1982; Uchiyama 1983; Obane et al. 1984). This study was done to obtain fundamental data concerning the safety of foods; (1) the chemical contaminants in the foods eaten in selected areas in Japan were analyzed and their content determined, and (2) the problems and trends in the intake of these chemicals and the sources of unusual residues were identified.

We carried out these studies seven times between the fiscal year 1977 and 1985 in Osaka Prefecture (relatively urbanized district in comparison with a rural area in Japan). Daily dietary intake of organic contaminants was obtained and the means of these were calculated from the data.

MATERIALS AND METHODS

In each fiscal year (1977–1982 and 1985), 89 food items were selected and 99–143 foods were collected from local supermarkets for the market basket samples. The choice of the foods and their consumption values were based on food consumption surveys by the Ministry of Health and Welfare in Japan and the Health Department of Osaka Prefecture. People over one year old were studied. The collected foods were classified into 13 groups; drinking water was also analyzed (Table 1). The food was cooked without seasoning (or prepared without cooking) and then homogenized with a minimum of water if neccesary. Analytical methods of pesticides and polychlorinated biphenyls (PCBs) were as described elsewhere (Maeda et al. 1980). Individual organic extracts were obtained from the low fat samples (I, II, III, V, VI, VII, VIII and IX groups) with n-

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Table 1. Average composition of the total diet in Osaka, Japan

Key	Food group	Average wt[a] g/day
I	Rice	219
II	Cereals except I	163
III	Sugar & adjuncts	41
IV	Oils & fats	17
V	Pulses	60
VI	Fruits	172
VII	Vegetables (green and yellow)	58
VIII	Vegetables except VII & seaweed	185
IX	Seasonings & beverages	135
X	Fish & shellfish	84
XI	Meat & eggs	123
XII	Milk & daily products	109
XIII	Prepared foods	12
XIV	Drinking water	600
total		1978

[a] The average in fiscal year 1977-1982 and 1985.

-hexane (pesticide analysis grade; Wako, Japan) and acetone (pesticide analysis grade; Wako) according to the method of Mills et al. (1963). The extraction method of high fat samples (IV, X, XI, XII and XIII groups) was based on that of Johnson (1965). Each fat material was extracted by n-hexane after the addition of ethanol (for pesticide analysis; Wako).

Fractions containing organo-chlorinated and phosphorous pesticides were obtained from a part of the fat (3 g) by acetonitrile partitioning. The former was purified by Florisil (60-100 mesh; Floridin Co) column chromatography and then analyzed by gas chromatography (GC) equipped with an electron capture detector (ECD). The latter was purified by Darco G-60 (activated charcoal; Atlas Powder Co)-Avicel (microcrystalline cellulose; Funakoshi Co, Japan) (1:10) column chromatography and analyzed by GC with an alkali-flame ionization detector. Carbamate residues were allowed to react with trifluoroacetic acid anhydride (extra-pure grade; Wako) and the products were purified with neutral alumina (activity I; Merck) containing 5% water and then analyzed by ECD-GC. Six residual chlordane compounds (α- and γ-chlordane, cis- and trans-nonachlor, oxychlordane and γ-chlordene) were analyzed according to the method of Miyazaki et al. (1980) after slight modification (Maeda et al. 1981). To detect them in samples of fiscal year 1983 and 1985, a selected ion monitoring technique using GC-mass spectrometry (MS; Jeol DX300) connected to a mass data analysis system (Jeol JMA-3500) was carried out instead of ECD-GC (Murakami et al. 1983). Residual PCBs were analyzed by ECD-GC after each sample was decomposed through a reflux (1 hr) with ethanol-KOH (100:5.6, v/w) and then purified by Florisil column chromatography. The values were determined by the calculation method for PCBs (Ugawa et al. 1973).

RESULTS AND DISCUSSION

Table 2 shows the dietary intake of pesticides and PCBs for each fiscal year and the average calculated from them. Each value varied with the fiscal year, but the average values were the same as those for Japan between fiscal year 1977 and 1982 (Uchiyama, 1983). Our values were also similar to those of the U.S. total market studies except for a few chemicals (Gartrell et al. 1985b). In the U.S. diet malathion residues were widely found in food groups containing grain & cereal products and sugar & adjuncts as well as those of meat, fish and poultry, fruits and oils & fats. However, they were found in only the II, III and XII groups in Japan and came from commercial flour (Kawamura et al. 1980). Therefore, U.S. malathion intake was about 7 times higher than ours. Dieldrin and heptachlor epoxide residues in the U.S. diet were 8 and 16 times more than ours, but β-HCH (β-BHC), p,p'-DDD (TDE) and hexachlorbenzene residues in the former were 18, 9 and 10 times less than the latter, respectively: the reason was unclear. These differences between countries are possibly derived from dietary custom and regulations of pesticide residues.

Table 2. Dietary intakes of PCBs and pesticides in total diet samples in Osaka, Japan

	Daily dietary intake (μg)							
Compounds	FY 77	FY 78	FY 79	FY 80	FY 81	FY 82	FY 85	average ± SD
PCBs	3.3	3.2	1.5	2.2	1.1	1.4	4.3	2.43 ± 1.20
α-HCH	0.3	0.95	1.8	2.6	1.3	0.26	0.37	1.08 ± 0.88
β-HCH	1.7	0.90	1.0	0.24	0.42	0.12	nd[a]	0.63 ± 0.61
γ-HCH	tr[b]	0.15	0.84	0.20	0.42	0.01	0.10	0.25 ± 0.30
δ-HCH	nd	0.04	nd	0.02	nd	nd	nd	0.01 ± 0.02
total HCH	2.0	2.04	3.64	3.06	2.14	0.39	0.47	1.96 ± 1.21
p,p'-DDD	0.5	0.43	1.6	0.55	0.44	0.27	0.38	0.60 ± 0.45
p,p'-DDE	0.9	1.9	1.6	2.1	1.1	0.61	1.7	1.42 ± 0.55
p,p'-DDT	0,3	2.1	1.6	2.0	0.14	0.22	1.0	1.05 ± 0.86
total DDT	1.7	4.43	4.8	4.65	1.68	1.10	3.08	3.06 ± 1.58
dieldrin	0.2	0.28	0.15	0.35	0.07	nd	0.05	0.16 ± 0.13
HCE[c]	0.1	0.13	nd	nd	0.01	nd	nd	0.03 ± 0.06
HCB[d]	0.2	0.12	0.90	0.60	0.22	0.18	0.17	0.34 ± 0.29
fenitrothion	nt[e]	0.69	0.54	0.61	1.1	1.1	1.6	0.94 ± 0.40
malathion	nt	1.1	2.5	1.6	2.2	2.6	1.1	1.85 ± 0.68
α-chlordane	nt	nt	nt	nd	nd	nd	0.45	0.11 ± 0.23
trans-nonachlor	nt	nt	nt	nd	nd	nd	0.56	0.14 ± 0.28

[a] Not detected. [b] Trace (present at levels below the limit of quantitation). [c] Heptachlor epoxide. [d] Hexachlorbenzene. [e] Not tested. Captan, captafol, chlorbenzilate, diazinon, dichlorvos, disyston, dimethoate, parathion, phenthoate, chlorpyrifos, EPN, phosalone, carbaryl, γ-chlordane, cis-nonachlor, γ-chlordene and oxychlordane were all under the detection limits.

Residues of metals and polycyclic aromatic hydrocarbons in total diet samples have been determined by our study group (Maeda et al. 1979, 1980; Murakami et al. 1981, 1983; Kuwabara et al. 1982; Obana et al. 1984). Many other contaminants such as chlorpropham and toxaphene should be analyzed even in Japan because of their high U.S. dietary intakes (Podrebarac 1984; Gartrell 1985b). A siginificant change in the U.S. total diet program format occurred in 1974; a part of the adult samples were replaced with those of infants and toddlers. Consequently, we ought to conduct such a study in infant and toddler market basket samples for long periods of time, although infant (0-3 years old) duplicate portion studies have been carried out by our working group (Yoshida et al. 1985).

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