# Dietary Intakes of Some Chlorinated Hydrocarbons and Heavy Metals Estimated on the Experimentally Prepared Diets

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It is established that the residues of persistent enviromental pollutants of one kind or another are detectable in almost all of the food stuffs we consume. The national studies have to be made on the average diet of the general population to assess whether or not the residue levels of these pollutants in the meal can pose any real hazard to human health.

Since such national survy has not yet been carried out in Japan, we have made some attempt to estimate the dietary intakes of the residues of pesticides, PCBs, PCTs, HCB, total mercury, methylmercury, lead and cadmium on the diets for 10 day period experimentally prepared.

## Materials and Methods

Dietary regimen for the total period of 10 days was prepared. The diets for 4 day period in this schedule were provided to meat the nutritional requirements for 20 ~ 29 years old male in the year of 1973 and those for 6 day period for pregnant woman in 1974.

The food materials were collected at the retail and department stores in Tokyo and were weighed out in the amount consumed in a day according to the menus of the diet. These materials weighed out were divided into 4 food groups, such as cereal products(A), vegetal products such as vegetable, vegetal oil, seasoning and seaweed(B), marine animal products(C) and terrestrial animal products including daily products and egg(D) and subjected to the analyses without cooking.

The residue levels of pesticides, PCBs, PCTs, HCB, total mercury, methylmercury, cadmium and lead were determined. No residues of PCTs, HCB and methylmercury were analyzed in the diet samples prepared in 1973, and analysis of total mercury made in 1973 was substituted



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for methylmercury in 1974.

Residues of PCBs and PCTs were extracted with acetone-hexane(2:1) and hexane-ether(3:1). Column chromatography on florisil and silica gel followed precleanup by saponification. The final extract was examined by gas chromatography, using EC detection. The analytical method used for BHC, DDT and HCB was the same as for PCBs and PCTs, except for the precleanup procedure. Decomposition of lipid was made with fuming sulphuric acid instead of alkali saponification. Total mercury and methylmercury were determined by the AOAC method (1975, 12th edition) and modified FDA method (SAITO, et al. 1972), respectively. Cadmium and lead extracted with MIBK after dry-ashing at 450° were analyzed by atomic absorption spectrophotometry.

## Results and Discussion

Chlorinated Hydrocarbons : Significant amount of PCBs was detected in all diet samples examined, except for one, diet No.3, in which no food group(C) was involved. The average dietary intake of PCBs was estimated as 6.3μg/day with a range of trace ~ 17μg. The study made by the Metropolis of Tokyo Bureau of Health on the diets for 10 day period showed the average intake of 3μg/day in 1973 (1974). HAMANO carried out the survies of PCB residues in the diet of nursing mother from 1972 to 1974 (1975). The average daily intake of PCBs was shown to be 3.56μg on the diets for 33 day period in 1973 and 2.80μg on the diets for 45 day in 1974 with respective ranges of 0.33 ~ 20.3μg and 0.15 ~ 40.16μg. Whereas the study on the diets for the total period of 60 days in 1972 showed remarkably higher average intake, 21.18μg, and wider range, 0 ~ 334μg. The higher levels of PCBs found in some diets in 1972 could be attributable to the some kind of fish contained in the diet which were suspected to be heavily contaminated by this substance. The temporary acceptable limit for PCBs, 3 ppm for fish in coastal and inland-water and 0.5 ppm for pelagic fish, was scarcely exceeded in the fish collected at the retail stores and markets in Tokyo from 1974 through 1975 (YAMANOBE, et al. 1974, 1975). It is estimated that the dietary intake of PCBs for the majority of population in Tokyo rarely exceed 20μg/day, so long as heavily polluted fish are not contained in the diet.

PCT residues could not be detected in any such 3 food groups as (A), (C) and (D) of all sample diet examined, but only in the group(B) of some diet. The average concentration of PCTs, 0.05$\mu$g, was so low as to be about 1/100 of PCBs. MINAGAWA (1975) and NISHIMOTO (1973) failed to detect any significant amount of PCTs in such food stuffs as edible oil, vegetable, daily products, meat and fish. It is of interest that, irrespective of extremely lower levels of PCTs in food, PCTs were found in human fat and blood at almost equivalent levels to those of PCBs (FUKANO, et al. 1975).

The dietary intake of DDT and BHC ranged from 1.3 to 12.2$\mu$g/day and 3.0 ~ 6.9$\mu$g/day with the means of 5.3$\mu$g and 4.4$\mu$g respectively. The content of HCB residues in the daily diet was on the average 0.5$\mu$g. This level was thought to be harmless, because daily intake of 0.6$\mu$g/kg was considered below any dosage rate known to be harmful (WHO, 1974).

TABLE 1-1

THE LEVELS OF SOME CHLORINATED HYDROCARBONS AND HEAVY METALS IN EXPERIMENTALLY PREPARED DIETS IN 1973, 74.

| No | Food stuffs group | Consumption of food | Total BHC | | Total DDT | | PCB | | Cadmium | | Lead | | Total Mercury | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | G | PPB | µG | PPB | µG | PPB | µG | PPB | µG | PPB | µG | PPB | µG |
| 1 | A | 420 | 1.0 | 0.4 | 0.1 | <0.05 | 6 | 3 | 42 | 18 | 200 | 84 | 10 | 4 |
| | B | 582 | 4.3 | 2.5 | 0.3 | 0.2 | 4 | 2 | ND | | 60 | 35 | 8 | 4 |
| | C | 196 | 2.5 | 0.5 | 2.3 | 0.5 | 23 | 5 | 19 | 4 | 90 | 18 | 360 | 70 |
| | D | 45 | 1.3 | 0.1 | 13.1 | 0.6 | 20 | 1 | 7 | <0.5 | 120 | 5 | 50 | 2 |
| | | | | 3.5 | | 1.3 | | 11 | | 22 | | 142 | | 80 |
| 2 | A | 390 | 1.3 | 0.5 | 0.5 | 0.2 | 6 | 2 | 27 | 10 | 170 | 68 | 10 | 4 |
| | B | 698 | 1.2 | 0.8 | 0.3 | 0.2 | 5 | 3 | ND | | 80 | 56 | 16 | 11 |
| | C | 67 | 1.7 | 0.1 | 0.3 | <0.05 | 21 | 1 | 13 | 1 | 150 | 10 | 40 | 3 |
| | D | 175 | 11.3 | 2.0 | 48.7 | 8.5 | 63 | 11 | 11 | 2 | 180 | 32 | 30 | 5 |
| | | | | 3.4 | | 8.9 | | 17 | | 13 | | 166 | | 23 |
| 3 | A | 370 | 3.8 | 1.4 | 0.1 | <0.05 | ND | | 97 | 36 | 190 | 72 | 12 | 4 |
| | B | 1090 | 2.3 | 2.5 | ND | | ND | | 8 | 8 | 130 | 142 | 5 | 5 |
| | D | 459 | ND | | 6.1 | 2.7 | <0.5 | <0.5 | 6 | 3 | 160 | 72 | 10 | 5 |
| | | | | 3.9 | | 2.7 | | <0.5 | | 47 | | 286 | | 14 |
| 4 | A | 370 | 3.8 | 1.4 | 0.1 | <0.05 | ND | | 97 | 36 | 190 | 72 | 12 | 4 |
| | B | 983 | 3.4 | 0.4 | 0.4 | 0.4 | ND | | 5 | 5 | 80 | 79 | 6 | 6 |
| | C | 300 | 2.6 | 0.8 | 16.3 | 4.9 | 9 | 3 | 95 | 29 | 220 | 66 | 43 | 13 |
| | D | 40 | 8.9 | 0.4 | 7.4 | 0.3 | 2 | <0.5 | 5 | <0.5 | 230 | 9 | ND | |
| | | | | 3.0 | | 5.6 | | 3 | | 70 | | 226 | | 23 |

A : CEREAL PRODUCTS. B : VEGETAL PRODUCTS INCLUDING VEGETAL OIL, SEASONING AND SEAWEED.
C : MARINE ANIMAL PRODUCTS.
D : TERRESTRIAL ANIMAL PRODUCTS INCLUDING MILK AND EGG.

TABLE 1-2

| No | Food stuffs group | Consumption of food | Total BHC | | Total DDT | | PCB | | Cadmium | | Lead | | Methyl Mercury | | HCB | | PCT | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | G | PPB | µG | PPB | µG | PPB | µG | PPB | µG | PPB | µG | PPB | µG | PPB | µG | PPB | µG |
| 5 | A | 400 | 1.7 | 0.7 | 0.5 | 0.2 | 4 | 2 | 124 | 49 | 571 | 228 | ND | | <0.05 | <0.05 | ND | |
| | B | 637 | 2.8 | 1.8 | 0.1 | 0.1 | 1 | 1 | 15 | 10 | 42 | 26 | ND | | 0.1 | 0.1 | 0.1 | 0.1 |
| | C | 81 | 13.9 | 1.1 | 15.0 | 1.2 | 21 | 2 | 22 | 2 | 228 | 18 | 38 | 3.0 | 3.5 | 0.3 | ND | |
| | D | 235 | 5.2 | 1.2 | 3.3 | 0.8 | 3 | 1 | 12 | 3 | 46 | 11 | 4 | 0.8 | 1.1 | 0.3 | ND | |
| | | | | 4.8 | | 2.3 | | 6 | | 64 | | 283 | | 3.8 | | 0.7 | | 0.1 |
| 6 | A | 410 | 0.7 | 0.3 | 0.2 | 0.1 | 2 | 1 | 95 | 39 | 133 | 54 | ND | | <0.05 | <0.05 | ND | |
| | B | 883 | 1.5 | 1.3 | 0.2 | 0.2 | 2 | 2 | 19 | 17 | 201 | 177 | ND | | 0.5 | 0.4 | 0.1 | 0.1 |
| | C | 56 | 8.0 | 0.4 | 12.2 | 0.7 | 11 | 1 | 11 | 1 | 151 | 8 | 55 | 3.1 | 0.6 | <0.05 | ND | |
| | D | 330 | 5.2 | 1.7 | 4.1 | 1.4 | 4 | 1 | 5 | 2 | 72 | 24 | ND | | 1.1 | 0.4 | ND | |
| | | | | 3.7 | | 2.4 | | 5 | | 59 | | 263 | | 3.1 | | 0.8 | | 0.1 |
| 7 | A | 415 | 2.5 | 1.0 | 0.5 | 0.2 | 4 | 2 | 66 | 27 | 167 | 69 | ND | | 0.1 | <0.05 | ND | |
| | B | 690 | 2.7 | 1.9 | 0.3 | 0.2 | 1 | 1 | 20 | 14 | 36 | 25 | ND | | <0.05 | <0.05 | 0.1 | 0.1 |
| | C | 91 | 5.0 | 0.5 | 5.4 | 0.5 | 14 | 1 | 15 | 1 | 291 | 26 | 29 | 2.6 | 0.1 | <0.05 | ND | |
| | D | 297 | 2.9 | 0.9 | 10.3 | 3.1 | 2 | 1 | 4 | 1 | 85 | 25 | 3 | 0.8 | 1.5 | 0.3 | ND | |
| | | | | 4.3 | | 4.0 | | 5 | | 43 | | 145 | | 3.4 | | 0.3 | | 0.1 |
| 8 | A | 360 | 4.8 | 1.7 | 2.7 | 1.0 | 2 | 1 | 103 | 37 | 155 | 56 | ND | | 0.1 | <0.05 | ND | |
| | B | 718 | 2.7 | 1.9 | 0.1 | 0.1 | 2 | 2 | 26 | 19 | 39 | 28 | ND | | <0.05 | <0.05 | ND | |
| | C | 126 | 13.2 | 1.7 | 13.2 | 1.7 | 20 | 3 | 17 | 2 | 43 | 5 | 93 | 11.6 | 0.1 | <0.05 | ND | |
| | D | 205 | 7.9 | 1.6 | 12.9 | 2.6 | 2 | <0.5 | 3 | 1 | 52 | 11 | ND | | 1.5 | 0.3 | ND | |
| | | | | 6.9 | | 5.4 | | 6 | | 59 | | 100 | | 11.6 | | 0.3 | | |
| 9 | A | 442 | 1.6 | 0.7 | 0.4 | 0.2 | 2 | 1 | 97 | 42 | 313 | 138 | ND | | <0.05 | <0.05 | ND | |
| | B | 682 | 2.4 | 1.6 | 0.1 | <0.05 | <0.5 | <0.5 | 23 | 16 | 22 | 15 | ND | | <0.05 | <0.05 | ND | |
| | C | 21 | 43.9 | 0.9 | 407.0 | 8.5 | 77 | 2 | 26 | 1 | 122 | 3 | 15 | 0.4 | 2.9 | <0.05 | ND | |
| | D | 350 | 3.7 | 1.3 | 10.0 | 3.5 | 2 | 1 | 12 | 4 | 29 | 10 | 1 | 0.4 | 0.9 | 0.3 | ND | |
| | | | | 4.5 | | 12.2 | | 4 | | 64 | | 166 | | 0.8 | | 0.3 | | |
| 10 | A | 440 | 1.5 | 0.7 | 0.4 | 0.2 | 3 | 1 | 72 | 32 | 127 | 56 | ND | | <0.05 | <0.05 | ND | |
| | B | 668 | 5.1 | 3.4 | 0.8 | 0.5 | 1 | 1 | 22 | 15 | 49 | 33 | ND | | 0.1 | 0.1 | <0.05 | <0.05 |
| | C | 91 | 1.9 | 0.2 | 22.3 | 2.0 | 20 | 2 | 39 | 4 | 51 | 5 | 630 | 57.3 | 0.6 | 0.1 | ND | |
| | D | 280 | 5.9 | 1.7 | 21.1 | 5.9 | 8 | 2 | 8 | 2 | 91 | 25 | 13 | 3.5 | 1.6 | 0.4 | ND | |
| | | | | 6.0 | | 8.6 | | 6 | | 53 | | 119 | | 60.8 | | 0.6 | | <0.05 |

Heavy Metals : The average intakes of cadmium and lead were estimated as 49µg/day and 190µg/day with the ranges of 13 ~ 70µg and 100 ~ 286µg which corresponded to about 1/2 of each provisional tolerable weekly intakes for cadmium, 400 ~ 500µg, and for lead, 3mg/person, propossed by WHO (1972). More than 50% of cadmium was encountered in group(A).

The highest concentration of total mercury, 80µg, was shown in the diet No.1 in which tuna was involved as a ingredient, and that of methylmercury, 60.8µg, was found in the diet No.10 which involved Sebastes matsubarai, a kind of fish in a deep sea. The level of methylmercury in the group(C) consisted of marine animal products of the diet No.10 was 0.6 ppm which exceeded the temporary acceptable limit for methylmercury in fish, 0.3 ppm. The average contents of total mercury and methylmercury were 35$\mu$g and 13.9$\mu$g which did not exceed the levels of provisional tolerable weekly intake for total mercury, 0.3mg/person, and for methylmercury, 0.2mg (WHO, 1972).

The levels of total mercury, methylmercury and PCBs in fish seem to be principal factores to control the dietary intake of these substnces.

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