RESIDUES AND TRACE ELEMENTS

Dietary Intake and Residues of Organochlorine Pesticides in Foods from Hsinchu, Taiwan

RUEY-AN DOONG and CHEN-YU LEE
National Tsing Hua University, Department of Nuclear Science, Hsinchu, 30043, Taiwan
YUH-CHANG SUN
National Tsing Hua University, Nuclear Science and Technology Development Center, Hsinchu, 30043, Taiwan

The levels of contamination with various organochlorine pesticides (such as total HCH, heptachlor, heptachlor epoxide, aldrin, dieldrin, endrin, endosulfan, and total DDT) of different foods from 3 traditional markets were determined to estimate Taiwanese daily intake of organochlorine pesticides. Of the 18 organochlorine pesticides investigated, α-HCH, β-HCH, lindane, δ-HCH, heptachlor, heptachlor epoxide, dieldrin, endrin, α-endosulfan, *p,p'*-DDE, and *p,p'*-DDT were detected at concentrations ranging from 0.26 to 10.2 ng/g wet weight. Contamination with organochlorine pesticides followed the order heptachlor > dieldrin > α-endosulfan > HCH isomers > heptachlor epoxide > DDT. Frequencies of detection of organochlorine pesticide residues ranged from 2.0 to 52.3%. α-Endosulfan was the most frequently detected organochlorine pesticide in the foods analyzed, followed by heptachlor epoxide (47.6%) and α-HCH (38.9%). Estimated daily intakes (EDIs) of organochlorine pesticides from foods were 1.137 μg for total HCH, 2.147 μg for heptachlor, 0.702 μg for heptachlor epoxide, 0.624 μg for endosulfan, 0.098 μg for cyclodiene, and 0.541 μg for total DDT. These EDIs were only 0.075% of the acceptable daily intake (ADI) for lindane, 47.5% of ADI for heptachlor and heptachlor epoxide, 0.045% of ADI for total DDT, and 1.01% of ADI for aldrin and dieldrin. Therefore, consumption of the foods analyzed does not pose a risk to consumer health.

Organochlorine pesticides were used extensively worldwide beginning in the early 1950s. Owing to their suspected carcinogenicities and mutagenicities, their use has been officially banned in Taiwan since 1974. However, it is estimated that 2.5×10^7 kg were released into the environment annually from the 1950s to 1970s. Also, the highly lipophilic properties of organochlorine pesticides have led to contamination of the environment and the food chain. The U.S. Food and Drug Administration (FDA) has regulatory responsibility to enforce pesticide tolerances in foods and routinely monitors the food supply for residues of these chemicals (1, 2). Therefore, it is necessary to estimate the dietary intake of organochlorine pesticides from different foods.

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The occurrence of organochlorine pesticides in foods has been reported by different authors (3–7). Most studies find that >80% of the total intake of organochlorine pesticide residues by nonoccupationally exposed humans was accumulated through the food chain. Although residue levels of these compounds have declined significantly in the past 20 years, the agricultural and medicinal uses of some organochlorine pesticides, such as DDT, HCH, and endosulfan, still continue in some Asian countries (8, 9). This continued use results in high exposure to organochlorine pesticides in the areas near the pollution sources. Nakagawa et al. (10) reported that the estimated daily intakes (EDIs) of total DDT in Japan, Canada, and the United States were 1.42, 6.38, and 1.3 μg/day, respectively. These EDIs are only 1/3 to 1/15 of the daily intake in China (20.47 μg/day; 8). Moreover, Sarkar et al. (11) indicated that sediments from the Arabian Sea along the west coast of India are contaminated with organochlorine pesticides and pose a threat to the biota in seawater. However, little information is available on the residues and dietary intake of organochlorine pesticides in Taiwan.

The present study was performed to estimate the amount of organochlorine pesticide residues in representative foods from traditional markets in Hsinchu in Northern Taiwan. EDIs of organochlorine pesticides from foods were evaluated to assess the possible health risks.

Experimental

Reagents and Materials

(a) *Solvents.*—Acetone, acetonitrile, ethyl ether, *n*-hexane, and petroleum ether. Acetone (Tedia, Fairfield, OH) was high performance liquid chromatographic (HPLC)/Spectro grade. *n*-Hexane and ethyl ether (Mallinckrodt, Phillipsburg, NJ) were Nanograde. Petroleum ether (Riedel de Haen, Seelze, Germany) was Pestanal grade, and acetonitrile (Tedia) was HPLC grade.

(b) *Analytical standards.*—Aldrin, α-HCH, β-HCH, lindane, δ-HCH, dieldrin, α-endosulfan, β-endosulfan, endosul-

Table 1. Recoveries (mean ± RSD) and method detection limits (MDLs) of organochlorine pesticides[a]

Organochlorine pesticide	Recovery, % (n = 3)			MDL, ng/g wet weight (n = 7)
	Shrimp	Shark	Pork	
α-HCH	87.8 ± 3.8	99.0 ± 5.2	106 ± 5.7	0.10
β-HCH	82.1 ± 3.9	90.2 ± 8.9	88.0 ± 6.3	0.13
Lindane	95.9 ± 3.7	104 ± 2.6	95.6 ± 6.7	0.24
δ-HCH	93.7 ± 4.9	105 ± 5.4	85.3 ± 4.6	0.10
Heptachlor	86.2 ± 4.1	91.1 ± 2.7	105 ± 5.7	0.12
Heptachlor epoxide	82.8 ± 3.9	92.5 ± 3.4	94.9 ± 7.2	0.36
Aldrin	86.2 ± 4.1	88.6 ± 3.8	110 ± 6.4	0.47
Dieldrin	81.0 ± 3.7	96.3 ± 5.1	102 ± 3.3	0.23
Endrin	87.3 ± 2.3	85.4 ± 2.3	95.4 ± 3.1	0.15
α-Endosulfan	67.9 ± 2.2	88.6 ± 3.8	95.2 ± 4.1	0.09
β-endosulfan	37.4 ± 1.0	92.3 ± 4.9	94.0 ± 2.74	0.14
p,p′-DDE	91.3 ± 3.8	98.8 ± 5.5	107 ± 4.1	0.21
p,p′-DDD	89.3 ± 3.3	98.8 ± 4.6	78.7 ± 2.6	0.18
p,p′-DDT	86.6 ± 4.4	96.6 ± 4.3	100.0 ± 3.2	0.13

[a] Samples were fortified with 25 ng organochlorine pesticides.

fan sulfate, endosulfan aldehyde, endrin, endrin aldehyde, heptachlor, heptachlor epoxide, methoxychlor, *p,p′*-DDE, *p,p′*-DDT, and *p,p′*-DDD were purchased from Supelco (Bellefonte, PA). The concentrations were 2000 μg/mL.

(c) *Sodium sulfate.*—Anhydrous, Pestanal grade from Riedel de Haen.

(d) *SRM 1945 standard.*—α-HCH, lindane, *p,p′*-DDE, *p,p′*-DDT, and *p,p′*-DDD in whale blubber. Certified concentrations were 4.25, 0.87, 116.8, 34.9, and 64.3 μg/kg, respectively.

(e) *Solid-phase extraction (SPE) cartridge.*—Envi-Florisil column, 6 mL, 1.0 g size, 100–120 mesh (Supelco). A Florisil SPE column was conditioned with 6 mL petroleum ether at a flow rate of 5 mL/min. Organochlorine compounds were eluted with 12 mL petroleum ether–ethyl ether (95 + 5).

Apparatus

(a) *Gas chromatograph.*—Hewlett-Packard 5890 equipped with ^{63}Ni electron capture detector (ECD). Operating conditions: column temperature programmed at 140°C, raised to 200°C at 15°C/min, held for 2 min, and then programmed to 250°C at 2°C/min, held for 2 min; injection mode, splitless; injector port temperature, 250°C; detector temperature, 300°C; carrier gas, nitrogen; column flow rate, 1.73 mL/min (linear velocity, 25.2 cm/s); makeup gas, nitrogen at 35.5 mL/min.

(b) *GC columns.*—PTE-5 fused-silica capillary column (30 m length × 0.32 mm id × 0.25 μm film thickness) and SPB-608 fused-silica capillary column (30 m length × 0.53 mm id × 0.5 μm film thickness).

(c) *Nitrogen purge device.*—Supelco.

(d) *Rotary vacuum evaporator.*—Model N-1N, Rikakikai, Tokyo, Japan.

(e) *SPE vacuum manifold.*—Supelco.

Food Sampling

Food samples were collected randomly between June 1996 and April 1997 from 3 traditional markets in Hsinchu, Taiwan. To monitor daily organochlorine pesticide intake from foods, 149 samples of 14 kinds of foods were collected. Three kinds of fruits (10 apples, 10 grapes, and 10 oranges), 3 kinds of meats (14 beef, 15 pork, and 10 chicken), 6 kinds of seafood (10 shrimps, 10 oysters, 12 clams, 3 sharks, 4 roaches, and 4 salmons), and 2 kinds of cereal products (25 rice and 12 wheat flours) were collected to represent the major food groups in the daily diet.

Extraction and Cleanup

The edible portion of each sample was finely chopped with a kitchen knife, wrapped with aluminum foil, and dried with a vacuum drier at –50°C for 48 to 72 h. The dried samples were homogenized and passed through a 325 mesh sieve. One to 5 g of each dried sample, depending on the water content, was placed into a thimble filter, and the organochlorine pesticides were extracted with 250 mL hexane for 16 h at a rate of 20 cycles/h. The extract was preconcentrated to 2–3 mL on a rotary evaporator and cleaned up with a Florisil SPE cartridge. Sodium sulfate (ca 1.0 cm) was added to a Florisil SPE cartridge, and the cartridge was washed with 6 mL petroleum ether–ethyl ether (95 + 5) at 5 mL/min. The organochlorine pesticides were eluted with 12 mL petroleum ether–ethyl ether (95 + 5) at a rate of 2 mL/min. The eluates were concentrated to ca 1–2 mL on a rotary evaporator and then transferred to 10 mL glass tubes with small amounts of hexane. The solvent in the glass tube was evaporated almost completely under a gentle stream of nitrogen, and the residues were redissolved in 1 mL hexane and analyzed by GC–ECD.

Table 2. Residues of organochlorine pesticides in 14 kinds of foods[a]

Analyte	Residue, ng/g wet weight										
	Pork (n = 15)	Beef (n = 14)	Chicken (n = 10)	Shrimp (n = 10)	Clam (n = 12)	Oyster (n = 10)	Roach (n = 4)	Salmon (n = 4)	Shark (n = 3)	Rice (n = 25)	Wheat flour (n = 12)
α-HCH	0.32	1.42	0.27	ND[b]	3.9	ND	ND	ND	ND	ND	ND
β-HCH	2.22	ND	0.45	ND	ND	ND	0.58	ND	ND	'ND	ND
Lindane	2.71	1.22	ND	ND	ND	ND	ND	ND	ND	ND	ND
δ-HCH	1.35	ND	1.59	0.38	2.46	ND	0.85	ND	ND	ND	ND
Total HCH	6.60	2.64	2.31	0.38	6.36	ND	1.43	ND	ND	ND	ND
Heptachlor	10.22	ND	0.37	ND	6.45	6.13	ND	13.9	ND	0.26	0.31
Heptachlor epoxide	2.08	1.65	0.81	0.26	ND	ND	1.97	3.63	0.89	0.29	ND
α-endosulfan	1.33	2.08	1.32	4.53	0.30	6.80	4.50	ND	1.15	ND	0.30
β-endosulfan	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Aldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Dieldrin	ND	ND	ND	ND	5.75	ND	ND	ND	ND	ND	ND
Endrin	ND	ND	0.5	ND	ND	ND	ND	ND	ND	ND	ND
p,p'-DDE	0.52	ND	0.67	ND	ND	ND	0.94	ND	ND	ND	ND
p,p'-DDD	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
p,p'-DDT	0.82	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.66

[a] No organochlorine pesticide was detected in fruit (n = 30), and the data were not shown.
[b] ND, not detected; below method detection limit.

Table 3. Frequencies of detection and average concentrations of organochlorine pesticide residues in 14 kinds of foods

Organochlorine pesticide	Positive samples for pesticides		Average concentration, ng/g wet weight
	No.	Detection frequency	
α-HCH	58	38.9	1.48 ± 1.69
β-HCH	21	14.1	1.08 ± 0.98
Lindane	50	33.6	1.97 ± 1.05
δ-HCH	49	32.9	1.33 ± 0.79
Heptachlor	47	31.5	7.41 ± 5.05
Heptachlor epoxide	71	47.6	1.61 ± 1.11
Aldrin	3	2.0	1.09 ± 0.12
Dieldrin	19	12.8	5.75 ± 2.4
Endrin	7	4.7	0.5 ± 0.08
α-endosulfan	78	52.3	2.76 ± 2.27
β-endosulfan	0	0	NA[a]
p,p'-DDE	28	18.8	0.71 ± 0.21
p,p'-DDD	0	0	NA[a]
p,p'-DDT	12	8.1	0.82 ± 0.27

[a] NA, not available.

GC Determination

The concentrations of organochlorine pesticides in the extracts were monitored by GC–ECD using a PTE-5 fused-silica capillary column. One microliter of each sample was injected into the GC system for separation and determination of organochlorine pesticides. GC peaks were identified through accurate assignment of retention times of standards (± 1%) and internal standards (pentachloronitrobenzene and decachlorobiphenyl). Identified peaks were checked by GC–ECD using an SPB-608 fused-silica capillary column. The SP-608 column was kept at 100°C, heated to 200°C at 6°C/min and then programmed to 250°C at 8°C /min, and held for 15 min. Residues of organochlorine pesticides were determined by comparing the peak area of the samples and the calibration curves of the standards. Correlation coefficients (R) of calibration curves of organochlorine pesticides were all > 0.998.

Quality Assurance

Recoveries of organochlorine pesticides by this method were determined by analysis of crab, shark, and pork samples ($n = 3$). Samples were spiked with 25 ng organochlorine pesticides (final concentration was 25 ng/mL). For every set of 10 samples, a procedural blank and spike sample consisting of all reagents were run to check for interference and cross-contamination. The quality of analytical data was assured by using the SRM 1945 whale blubber standard. Recoveries of α-HCH, lindane, *p,p'*-DDE, *p,p'*-DDD, and *p,p'*-DDT were 103, 103, 105, 77, and 88%, respectively.

Results and Discussion

Table 1 shows recoveries and detection limits of organochlorine pesticides from shrimp, shark, and pork. Recoveries ranged from 80 to 110%, in agreement with FDA recommendations. Detection limits ranged from 0.09 ng/g wet weight for α-endosulfan to 0.47 ng/g wet weight for aldrin.

Table 2 shows the residues of organochlorine pesticides in 14 kinds of foods. No organochlorine pesticide was detected in fruit. This may be due to the low fat content and high water content of fruits. Also, removals of peels during pretreatment may also eliminate any pesticide residue.

Residues of heptachlor, α-endosulfan, and heptachlor epoxide were found in cereals. Twelve of the 18 pesticides studied were found in seafood and meats at levels greater than the method detection limit: α-HCH, β-HCH, lindane, δ-HCH, heptachlor, heptachlor epoxide, aldrin, dieldrin, endrin, α-endosulfan, *p,p'*-DDE, and *p,p'*-DDT. Endosulfan sulfate, endosulfan aldehyde, endrin aldehyde, and methoxychlor were not detected. The concentrations of detected pesticides ranged from 0.26 to 10.22 ng/g wet weight. The order and pattern of contaminant concentrations varied with food type. In general, average concentrations followed the order heptachlor > dieldrin > α-endosulfan > HCH isomers > heptachlor epoxide > DDT.

Frequencies of detection of organochlorine pesticide residues (Table 3) ranged from 2.0 to 52.3%. α-Endosulfan was the most frequently detected, followed by heptachlor epoxide (47.6%) and α-HCH (38.9%). Although α-endosulfan was the most frequently detected pesticide, no β-endosulfan was found in any sample, perhaps because β-endosulfan is degraded under aerobic conditions to α-endosulfan. Lee (12) reported that β-endosulfan could be transformed into α-endosulfan by *Pseudomonas* species in soil. The half-lives of α-endosulfan and β-endosulfan are 800 days and 60 days, respectively.

The high detection frequencies of endosulfan, heptachlor, and HCH may result because they are still used in some developing countries and may still be contaminating foods via atmospheric or seawater deposition. Tanabe et al. (13, 14) speculated that the increasing use and disposal of organochlorine

Table 4. Estimated daily intakes (EDIs) of organochlorine pesticides from foods by an average person

Analytes	EDI, μg, by an average person from the indicated amount of food							
	Pork (128.63 g)	Beef (7.45 g)	Chicken (74.19 g)	Shellfish (16.91 g)	Fish (133.47 g)	Cereal (421.56 g)	Fruit (136.5 g)	Total, μg
α-HCH	0.041	0.011	0.020	0.021	NA[a]	NA	NA	0.093
β-HCH	0.286	NA	0.033	NA	0.025	NA	NA	0.344
Lindane	0.349	0.009	0.000	NA	NA	NA	NA	0.358
δ-HCH	0.174	NA	0.118	0.013	0.037	NA	NA	0.342
Heptachlor	1.315	NA	0.027	0.069	0.618	0.118	NA	2.147
Aldrin	NA	NA	NA	0.002	NA	NA	NA	0.002
Heptachlor epoxide	0.268	0.012	0.060	NA	0.288	0.074	NA	0.702
α-endosulfan	0.171	0.015	0.098	0.039	0.251	0.050	NA	0.624
p,p′-DDE	0.067	NA	0.050	0.002	0.041	NA	NA	0.160
Dieldrin	NA	NA	NA	0.059	NA	NA	NA	0.059
β-endosulfan	NA	NA	NA	NA	NA	NA	NA	NA
Endrin	NA	NA	0.037	NA	NA	NA	NA	0.037
p,p′-DDE	NA	NA	NA	NA	NA	NA	NA	NA
p,p′-DDT	0.105	NA	NA	NA	NA	NA	NA	0.105

[a] NA, not available.

pesticides in some developing countries would accelerate the pollution of seawater. Oehme et al. (15) measured the seasonal concentration changes of organochlorines in the European Arctic and found that long-range air transport from more polluted source areas might lead to a significant concentration change in Arctic air. As expected from the liposolubility of these residues, the detection frequencies and mean levels of organochlorine pesticides in high-fat foods such as pork, beef, and chicken were higher that those in low-fat foods. Therefore, more attention should be paid to the intake of the studied pollutants from high-fat foods.

Residues of total HCH were detected in about 75% of samples. The detected levels in meats ranged from 2.31 ng/g wet weight in chicken to 6.6 ng/g wet weight in pork. These levels were slightly higher than those in seafood, which ranged from 0.38 to 6.36 ng/g wet weight. The concentrations of total HCH in shrimp, clam, and roach were 0.38, 3.9, and 1.43 ng/g wet weight, respectively. No total HCH was detected in oyster, salmon, shark, and cereal. α-HCH was the most frequently found pesticide in total HCH, but lindane gave the highest mean level of residue.

Seafood and meats were highly contaminated with heptachlor and heptachlor epoxide. Average concentrations of heptachlor and heptachlor epoxide were 3.53 and 1.51 ng/g wet weight for meats and 8.82 and 1.69 ng/g wet weight for seafood, respectively. Also, heptachlor and its metabolite were detected in the cereal group at mean concentrations of 0.285 and 0.15 ng/g wet weight, respectively. Heptachlor is still used in some Asian countries. Although the average concentration of heptachlor epoxide, a metabolite of heptachlor, was lower than that of heptachlor, the detection frequency of heptachlor epoxide was higher than that of heptachlor.

Contamination of foods with cyclodienes (aldrin, dieldrin, endrin) was very low. Detection frequencies of aldrin and dieldrin and endrin were 2.0 and 12.8%, respectively. Aldrin was detected only in oysters, and dieldrin was detected only in clams. The average concentrations of aldrin and dieldrin were 1.68 and 5.75 ng/g wet weight, respectively. The detection frequency of endrin was 4.7% and the detected concentration in chicken was 0.5 ng/g wet weight, which was lower than that of dieldrin.

DDT contamination of meat and seafood was low. Total DDTs were detected at mean levels ranging from 0.67 ng/g wet weight in chicken to 1.34 ng/g wet weight in pork. Among the DDT metabolites, *p,p′*-DDE was most frequently detected (19%), at a mean level of 0.71 ng/g wet weight. *p,p′*-DDT was detected in only 8.1% of samples but at an average concentration of 1.23 ng/g, wet weight higher than that for *p,p′*-DDE.

The ratio of DDE to DDT was used to determine whether recent exposure to DDT had occurred. A ratio of 2.5 suggests an earlier use of DDT rather than a recent exposure. The concentrations of DDT still observed in the samples might be due to long-range atmospheric transport from regions where DDT is still used.

Table 4 gives EDIs of organochlorine pesticides. These values were calculated by multiplying the concentration of or-

Table 5. Comparison of estimated daily intake (EDI) and acceptable daily intake (ADI) for some organochlorine pesticides

Organochlorine pesticide	EDI, μg	ADI[a], μg	% ADI reached
Lindane	0.358	480	0.075
Heptachlor + heptachlor epoxide	2.849	6	47.48
Aldrin + dieldrin	0.061	6	1.01
Total DDT	0.541	1200	0.045

[a] Recommended by FAO/WHO. The body weight for an average person is 60 kg.

ganochlorine pesticide in each food by the weight of an average person. The total daily intakes from foods were 1.137 μg for total HCH, 2.147 μg for heptachlor, 0.702 μg for heptachlor epoxide, 0.624 μg for endosulfan, 0.541 μg for total DDT, and 0.098 μg for cyclodienes. Among the samples analyzed, pork and fish contributed the most to the EDIs of organochlorine pesticides, especially for total HCH, heptachlor, heptachlor epoxide, and α-endosulfan. The contributions of pork were 74.8% for total HCH, 61.2% for heptachlor, and 38.2% for heptachlor epoxide. The contributions of fish were 46.2% for heptachlor epoxide and 40.2% for α-endosulfan. The distribution pattern of daily intake determined in this study is different from that determined by other researchers (16, 17). The intake of total HCH in this study was much higher than that in earlier reports, whereas the intake of total DDT was lower than that in the same reports. Nakagawa et al. (10) showed that daily intakes of total HCH per person in Japan, Canada, and the United States were 0.56, 0.39, and 0.16 μg, respectively, whereas the intakes of total DDT were 1.42, 6.38, and 1.30 μg, respectively. The intakes of total HCH and total DDT determined in this study were 1.137 and 0.265 μg, respectively. Lindane was banned in Taiwan in 1985, about 10 years after DDT was banned there.

We assessed the hazardous effects of residues of organochlorine pesticides on human health by using the acceptable daily intakes (ADIs) proposed by the Joint Food and Agriculture Organization/World Health Organization Meeting on Pesticide Residues and by assuming that the body weight of an average adult is 60 kg. As shown in Table 5, EDIs were only 0.075% of ADI for lindane, 47.5% of ADI for heptachlor and heptachlor epoxide, 0.045% of ADI for total DDT, and 1.01% of ADI for aldrin and dieldrin. These results show that consumption of the foods analyzed does not pose a health risk on the basis of organochlorine pesticide content.

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