J. Agric. Food Chem. 1994, 42, 369–373

Effects of the Processing Steps in Tofu Production on Pesticide Residues

Makoto Miyahara* and Yukio Saito

Division of Foods, National Institute of Hygienic Sciences, 1-18-1 Kamiyoga Setagaya-ku, Tokyo 158, Japan

Tofu is a traditional Japanese food that has become an international food. Soybeans are sprayed with pesticides to prevent damage by pests during storage and shipment. To test the effect of each stage of soybean processing on these pesticides, soybeans were spiked with four pesticides. The soybeans were subjected to tofu production conditions in the laboratory. The concentrations of the pesticides in the washed soybeans, soybean milk, okara, and tofu were determined immediately after each process. Conclusions from these experiments are as follows: (1) The persistence of each of the tested pesticides after tofu production is negligible (<0.5%). (2) With the exception of captan, the concentrations of pesticides in the soybean milk were essentially the same as in the raw soybeans and were not decreased with further cooking. Captan levels dropped to less than 10% of the original values at 2.5 min. (3) The washing of soybeans caused the concentrations of dichlorvos, malathion, chlorpyrifos, and captan in or on the soybeans to decrease markedly. The residues in the soybeans decrease with the first two washings; however, there is no further reduction with subsequent washings.

INTRODUCTION

We have been studying pesticide residues in processed food. Soybeans are one of the major imported crops and are processed into many foods in Japan. During storage and transportation, soybeans are treated with pesticides to prevent losses due to pest damage. The applied amounts and the residual levels are very high compared with preharvest application (Elms et al., 1972; Bates and Rowlands, 1964). Also, pesticides applied after harvest tend to persist because losses due to weathering or translocation are negligible. Interest has focused on removal of the pesticides by food production processes. We studied the removal of pesticides in or on soybeans by the oil refining process and concluded that no pesticides remain in the soybean oil after appropriate refining (Miyahara and Saito, 1993). We also concluded that the deodorizing process is an important process in minimizing pesticide residues in the final products. The oil refining process involves drastic conditions such as steam distillation in vacuum (5 mmHg) and at high temperature (250 °C), which eliminate pesticides from the crude oil.

Tofu and soy sauce are made from soybeans and are important traditional foods in Japan. Tofu is manufactured by a few steps (National Food Research Institute, 1966; Watanabe, 1970, 1987; Sata, 1988). The process involves only cooking, washing, and coagulating stages. Therefore, persistence of pesticide residues is of concern.

Studies on the removal of pesticides from processed food have been limited to oil-refining processes (Addison et al., 1978; Chaudy et al., 1978; Hashemy-Tonkabony and Soleimani-Amiri, 1976; Smith et al., 1968; Kanematsu et al., 1974). No studies were found that dealt with tofu, which is widely consumed today in the world. Studies were needed to determine the effects of tofu production on soybean pesticide residue levels (Figure 1) in process intermediates. The pesticides we studied are representative of those that are used on soybeans.

For the purpose of this study, simulated commercial processing procedures were established. Since tofu is a traditional food, there are many variations of commercial production, and many kinds of tofu are produced in Japan (Watanabe, 1970). Standard tofu production procedures were selected, and simulated processing procedures were established for laboratory study.

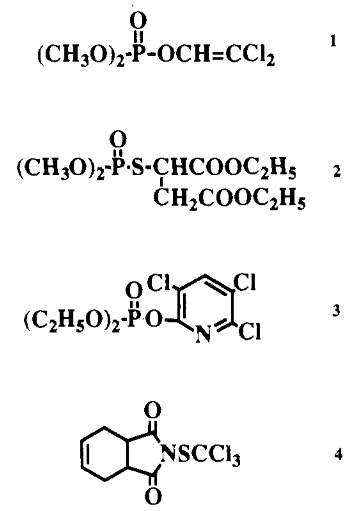

Figure 1. Structures of pesticides.

EXPERIMENTAL PROCEDURES

Apparatus. (*a*) *A Shimadzu Model SPL14 gas chromatograph with electron capture detector* and splitter was used. A DB-1 capillary column (30 m × 0.32-mm i.d.) with a film thickness of 0.25 μm was used with helium carrier gas at 3 mL/min and a helium septum purge at 3 mL/min. A split ratio of 20 was used. Injector and detector temperatures were 250 and 280 °C, respectively. The column oven temperature was initially maintained 40 °C for 1 min and then increased to 150 °C at 20 °C/min, held 150 °C for 1 min, and then increased at 4 °C/min to 210 °C, and finally maintained 210 °C for 30 min. A 1-μL injection in the splitless mode was utilized followed by a waiting time of 1 min. Makeup gas consisted of nitrogen at 50 mL/min.

(*b*) *A Shimadzu Model GC9A gas chromatograph with flame photometric detector* and splitter was used. A DB-5 capillary column (30 m × 0.32-mm i.d.) with a film thickness of 0.25 μm was used with helium carrier gas at 3 mL/min and a helium septum purge at 3 mL/min. A split ratio of 20 was used. Injector and detector temperatures were 250 °C. The column oven temperature was initially maintained 120 °C for 1 min and then increased to 150 °C at 20 °C/min, held 150 °C for 1 min, and then increased at 4 °C/min to 210 °C, and finally maintained 210 °C for 10 min.

0021-8561/94/1442-0369$04.50/0

A 1-μL injection in the splitless mode was utilized and followed by waiting time of 1 min. Makeup gas consisted of nitrogen at 50 mL/min.

(*c*) *A Horiba B201 pH meter* was used.

(*d*) *A corrosion-resistant freeze-dryer* Model FD8-84, FTS Systems Co., New York, was used.

Reagents. (*a*) *Pesticide standards* included dichlorvos (1), *O*,*O*-dimethyl *O*-(2,2-dichlorovinyl) phosphate >98%; malathion (2), *S*-(1,2-dicarboxyethyl) *O*,*O*-dimethyl dithiophophate >98%; chlorpyrifos (3) *O*,*O*-diethyl *O*-(3,5,6-trichloro-2-pyridyl) phosphorothioate >99%; captan (4), *N*-[(trichloromethyl)thio]-4-cyclohexene-1,2-dicarboximide >98%. These standards were purchased from Wako Pure Chemicals Co.

(*b*) *All organic solvents* for analysis were of pesticide residue grade.

(*c*) *Caution:* All the organic solvents and the pesticide standards used in this study could be harmful to health. They have to be handled carefully.

Samples. A sample of soybeans was obtained from a retail store in Tokyo. Soybeans were hand sorted to eliminate contaminants.

Preparation of Soybeans with Pesticides. A mixture of pesticide standards in acetone was sprayed onto the surface of the sample in a rotating flask. After 15 min, the sample was stored in the refrigerator until processed.

Preparation of Tofu for Specifications. A sample (50 g) was soaked in 100 mL of water at 20 °C for 12 h. After soaking, the soybeans in 400 mL of water were thoroughly ground with a mixer for 5 min. The mixture was heated at 100 °C for 5 min. The hot mixture or go (above 75 °C) was filtered through two layers of gauze to separate okara. The filtrate (soybean milk) was heated and maintained at 75 °C for 1 min. The soybean milk was coagulated with 1 g of suspended calcium sulfate in 10 mL of water. After coagulation was complete, coagulated soybean (crude soybean curd) was collected in a small box with holes and pressed with 500 g of weight for 10 min. The crude soybean curd was rinsed with water for 30 min. The tofu was subjected to analysis immediately after preparation.

Soybean Washing Procedure. The sample (100 g) was placed in a 500-mL beaker, 200 mL of water was added, and the mixture was left for 12 h. It was stirred slowly for 10 min. The water was decanted, and 10 g of sample was collected for analysis. The remaining 90 g was washed with 160 mL of water by stirring for 10 min. The procedure was repeated two times. Each sample was ground in a Waring blender and then subjected to analysis.

Cooking Procedure. The spiked and ground sample (200 g) was heated to make go at 100 °C with 2000 mL of water. The temperature was maintained at 100 °C with careful stirring to prevent sticking. Samples (10 g) were taken from this mixture periodically (0, 2.5, 5, and 10 min) and cooled in an ice bath immediately. The cooled sample was analyzed.

Preparation of Okara and Soybean Milk with Pesticides. A spiked sample (20 g) was soaked in 40 mL of water at 20 °C for 12 h. After soaking, the soybeans and 160 mL of water were thoroughly ground in a Waring blender for 5 min. The mixture was heated at 100 °C for 5 min. The hot mixture, go, (above 75 °C), was filtered through two layers of gauze to separate okara. This wet okara was subjected to analysis after cool-down. The filtrate (soybean milk) was heated and maintained at 75 °C for 1 min. The soybean milk was subjected to analysis after cooling.

Preparation of Tofu with Pesticides. The pesticide-treated soybeans were used without washing. The sample (20 g) was soaked in 40 mL of water at 20 °C for 12 h. After soaking, the soybeans were thoroughly ground with 150 mL of water for 5 min. The mixture was heated at 100 °C for 5 min. The hot mixture (above 75 °C) was filtered through two layers of gauze to separate okara. The soybean milk was heated and maintained at 75 °C for 1 min. The soybean milk was coagulated by addition of 0.4 g of suspended calcium sulfate in 4 mL of water. After coagulation was complete, crude soybean curd was collected in small box with holes, lapped with gauze, and pressed with a weight (500 g) for 10 min. The crude soybean curd was rinsed with water for 30 min. The tofu was subjected to analysis immediately after preparation.

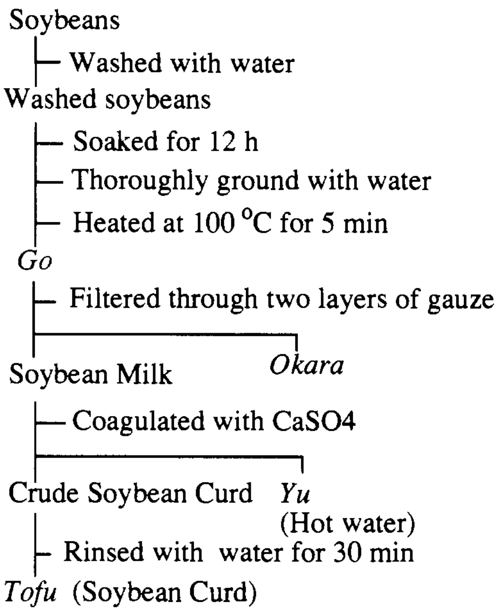

Figure 2. Tofu production steps.

Measurement of Water Content in Soybeans. The ground sample (10 g) was stored at 130 °C for 3 h. The weight loss was measured after the sample was cooled in a desiccator.

Measurement of Dry Content in Okara and Tofu. The sample was dried by freeze-drying at room temperature for 48 h.

Analytical Procedure for Soybeans and Washed Soybeans. Pesticides were extracted from the ground sample (10 g) with two 50-mL portions of acetonitrile. After filtration, the solvent was evaporated under reduced pressure. The residue was dissolved in 15 mL of hexane, and pesticides were removed by extraction with three 30-mL portions of acetonitrile. The combined acetonitrile extracts were evaporated under reduced pressure, and the residue was dissolved in 5 mL of hexane. For the determination of the organophosphorus pesticides, the solution was analyzed by gas chromatography without further purification. For the determination of captan, the solution was subjected to Florisil column chromatography.

Florisil PR (20 g) was packed in a chromatographic column (22-mm i.d. × 300 mm) as a slurry in hexane. The column was washed with 200 mL of hexane. The pesticides were eluted with 100 mL of a mobile phase consisting of ethyl acetate and hexane (1:4). The eluate was collected, and the solvent was evaporated under reduced pressure. The residue was dissolved in 5 mL of hexane. The recoveries for dichlorvos, malathion, chlorpyrifos, and captan in soybeans were 91%, 97%, 96%, and 83%, respectively.

Analytical Procedure for Okara and Soybean Milk. Each sample was extracted twice with 100 mL of ethyl acetate. The solvent was evaporated under reduced pressure. The residue was dissolved in 15 mL of hexane. This was extracted with three 30-mL portions of acetonitrile. The combined acetonitrile extracts were evaporated and the residue was dissolved in hexane prior to Florisil chromatography as described in the previous section.

Hardness Test. Disk (10-mm diameter) was set on sample and weight was loaded on it. Hardness was determined by total weight on it required to break the sample surface.

RESULTS AND DISCUSSION

Simulation of Commercial Tofu Production Process. The procedures in this study are comparable with commercial tofu production processes (Figure 2). To confirm this, yields and several other parameters were monitored. These parameters were measured by standard analytical methods for tofu which are authorized by the

Table 1. Yields for Each Stage of Processing

	weight				ref values[a]			
	wet		dry[b]		wet		dry	
material	g	%	g	%	g	%	g	%
soybeans	50	100	43	100	1000	100	880	100
washed soybeans	115	230			2200	220		
soybean milk[c]	436.5	873	30.3	70	8700	870	643	72
okara	63.5	127	12.7	30	1300	130	246	28
tofu	172	346	19.0	44	4000	400	480	54

[a] Reference (National Food Research Institute, 1966). [b] Dry weight after freeze–drying for 48 h. [c] Calculated from weights of soybeans and okara.

Table 2. Specifications for Tofu

parameter	tofu	ref values[a]
extracting rate[b] (%)	70	66–68
yield[c] (%)	44	50–55
coagulating rate[d] (%)	63	70–80
pH	6.2	6.0–6.5
hardness (g/cm^2)	40	30–50
solid[e] (%)	11	9.8

[a] Reference (National Food Research Institute, 1966). [b] DWsm/DWsb × 100.[f] [c] DWtf/DWsb × 100.[f] [d] DWtf/DWsm × 100.[f] [e] DWtf/WWtf × 100.[f] [f] Key: DWsm, dry weight of soybean milk; DWsb, dry weight of soybeans; DWtf, dry weight of tofu; WWtf, wet weight of tofu.

Table 3. Concentration (ppm) of Pesticides at Each Stage of Processing

	dichlorvos		malathion		chlorpyrifos		captan	
sample	ppm[a]	μg[b]	ppm	μg	ppm	μg	ppm	μg
soybeans	5.01	250.5	7.9	395	11.2	560	2.87	143
soybean milk	0.20	87.3	0.89	388	1.2	541	0.053	23
okara	0.13	8.3	0.43	27	0.59	37	0.017	1.1
tofu	0.06	10.3	0.72	123	0.82	141	0.014	2.4

[a] Based on the wet weight. [b] concentration × weight.

National Food Research Institute of Japan (National Food Research Institute, 1966). As shown in Tables 1 and 2, the products prepared by the laboratory do meet the specifications for commercially produced tofu. The dry weight of the sample soybeans (50 g) was 43 g, which corresponds to a water content of 14%. The sample was harvested in 1990 and washed, packed, and sold in the spring of 1991. The sample was fresh and maintained with water in the package. Under such storage conditions, the water content was greater than that of imported soybeans (<12%) (Miyagawa, 1988). The weight for washed soybeans increased because soybeans absorb water. The increase of 230% is quite normal and consistent with the reference value (220%). This implies the soybeans used were suitable for tofu. The weight of the cooking mixture (go) was adjusted by water addition. The total weight of go was 10 times the initial soybean weight. In this case, the total weight was adjusted to 500 g. To elucidate the suitability of the filtration, yields based on soybeans (50 g) were examined. Wet and dry weights of soybean milk were estimated from the weights of soybeans and okara and were 436 and 31 g, respectively. The yield of okara (63.5 g, 127%) was comparable to the commercial specification. Dry weight and yield, based on wet soybeans (50 g), of okara were 12.5 g and 30%, respectively. The values were larger because the soybeans were ground too fine to filter off. However, the deviation from the reference value was negligible. These results illustrate that the filtration process adequately simulates the commercial process. To examine the coagulating process, the yields of tofu (based on soybeans) were also calculated. The weight of tofu and the yield based on wet soybeans were 172 g and 346%, respectively. The dry weight of the tofu was 19 g, and the yield based on dry soybeans was 44%. Those values were rather less than reference values (440 and 54%). This means that water-soluble components in soybean milk were in excess for the tofu process. Total weight loss of our experiments was 26%, and it ranged from 24 to 28%. These values are greater than the reference value of 19%. On the basis of these results, the coagulating components of the soybeans we used may be reduced, and as a result, we obtained a poor yield. However, the coagulating process was judged to be adequate.

This conclusion is supported by the parameters shown in Table 2. These data are comparable to the reference values (Watanabe, 1970, 1987) in Table 2. The results indicate that the extracting and coagulating processes were quite similar to the commercial processes. The extracting process involves homogenizing, cooking, and filtration. The extracting rate was 70% and is consistent with the reference value. The extraction rate was defined as ratio of soybean milk (dry weight) and soybeans (dry weight). This parameter indicates that the extraction process was properly carried out.

The isolation of the tofu requires coagulating, separating of water from soybean curd, pressing, and rinsing. The yield of tofu was 44% (defined as ratio of tofu (dry weight) and soybeans (dry weight), and the coagulating ratio was 63%. These values are less than the reference values. These results suggest that the coagulating components in the soybeans we used were reduced relative to those in commercially processed soybeans. They may have contained more water-soluble components such as sugars, which are not coagulated. However, the production process proceeded normally and gave normal tofu.

The hardness and pH of tofu were measured to confirm the results on the laboratory production. The pH was 6.2 and shows that the quantity of coagulating agent ($CaSO_4$) was adequate. Hardness was 30 g, which confirms that the coagulating temperature was appropriate. The wet and dry weights of tofu were 172 and 19 g, respectively. The solid content of tofu was calculated from those two parameters and was 11%. These results were consistent with the reference value for tofu of 9.8%. As discussed above, deviations from specifications were observed, but in general, each process successfully simulated the commercial process. Therefore, the pesticide removing effects of the laboratory processes and the commercial processes are probably quite similar.

Residual Levels of Pesticides at Various Stages of the Process. Pesticides were added to the soybeans and the concentrations of the pesticides are shown in Table 3. The analytical procedures were slight modifications of reported procedures (Miyahara et al, 1992a; Miyahara and Saito, 1993). To obtain homogeneous samples special techniques were necessary. Without adequate rotation, variations in pesticide levels were excessive.

The process was stopped at the filtration stage to get samples for measurement of residual levels in soybean milk and okara. The washing process was omitted to ensure that enough pesticide remained to permit computation of the pesticide-eliminating effect of the particular step under study. The other runs for the tofu production process were completed for the same reason. The residual pesticides were measured as described in the Experimental Procedures immediately after each process was finished. The concentrations are based upon the wet sample weights. The concentrations of pesticides in soybean milk, okara, and tofu are also shown in Table 3. The residual levels are very low. The actual quantities are difficult to compute from the concentrations because the sample weights varied enormously from sample to sample. To estimate the amounts of pesticides in the samples, their quantities were calculated from the concentrations and weight data in Table 1.

Dichlorvos (1) was easily removed in every stage. Since dichlorvos is an unstable pesticide and very volatile (7 × 10^{-2} mmHg at 40 °C), it may have been vaporized by heat in the homogenizing and cooking stages (Saito et al., 1981). The levels in samples of the following stages ranged from 0.2 to 0.06 ppm. Thus, all stages of the process play a significant role in removing the pesticide.

Malathion and chlorpyrifos were extracted with hot water and retained in the hot extract (soybean milk); however, they were effectively removed by the coagulating steps. They do not decompose during coagulation and rinsing steps and are not coagulated in these conditions because both pesticides are stable in water (Norris et al., 1954; Miyahara et al., 1991). They remain in the water (yu) which separated from the soybean milk. This conclusion is supported by a previous study (Miyahara et al., 1991) that demonstrated that pesticides are not removed or decomposed by the coagulating agent which is an aqueous solution of 0.47 M phosphoric acid and 0.23 M ammonium chloride. The coagulating effectiveness for the reagent is 96%. The coagulation ability is much more efficient than that (63%) of calcium sulfate in the tofu production process. The pesticides are not incorporated in the coagulated matrix under these conditions. From these results and others, it is concluded that most pesticides remain in the water that is separated from the soybean milk. The warm water separated from soybean milk is called yu. In yu, many water-soluble components of soybeans are removed from the soybean curd. Residual pesticides will be removed from the curd matrix by the rinsing process. Crude soybean curd was washed with large volumes of water for 30 min to remove excess calcium sulfate. The solubility of calcium sulfate in water is 2000 ppm at room temperature. Solubilities of pesticides are 145 and 2 ppm for malathion and chlorpyrifos, respectively. Some of the pesticides in the matrix are removed from soybean curd by the rinsing process.

Captan was removed at an initial stage of the process because it is unstable in the homogenate. It decomposes in fresh vegetable homogenates (Wolfe et al., 1976) and is removed (Miyahara et al., 1992b; Cairns, 1983).

Removing Pesticides in the Washing Process. The washing process is quite effective in removing pesticide contaminants from the soybeans. To study this process, spiked soybeans were washed several times at room temperature. The concentrations of pesticides were monitored by gas chromatography (Miyahara et al., 1992a). After the soybeans were washed with water twice, the pesticides levels were reduced to about 10 and 20% of the initial levels (Figure 3). The solubilities of those pesticides range from 1000 to 2 ppm (dichlorvos, 1000 ppm; malathion, 145 ppm; chlorpyrifos, 2 ppm; captan, 3.3 ppm at room temperature); however, the residual levels were independent of those solubilities. The results suggest that sprayed pesticides remain as microparticles on the surface of the soybeans. These are easily removed by mechanical stirring in water. Thus, 80% of pesticides on the surface are washed away. The remaining pesticides may exist in the soybeans and are not removed by washing. The samples were stored for 2 weeks after pesticide application. If the washing process was carried out immediately after pesticide application, the residual levels were lower than those observed with aged samples.

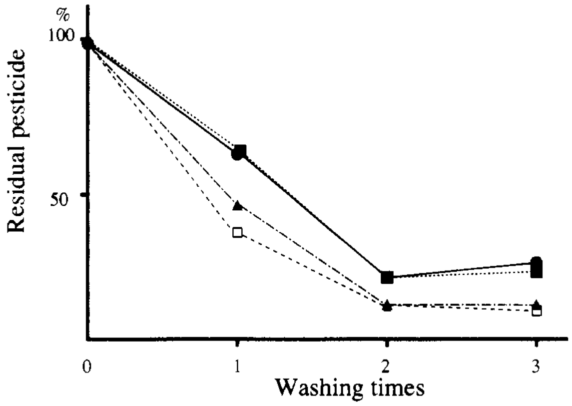

Figure 3. Disappearance of pesticides during washing: (▲) malathion; (■) chlorpyrifos; (●) dichlorvos; (□) captan.

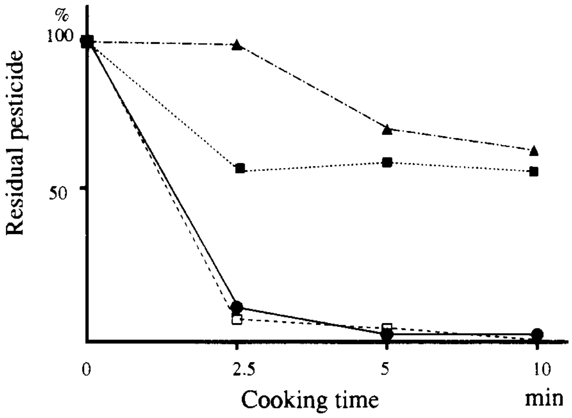

Figure 4. Disappearance of pesticides during cooking: (▲) malathion; (■) chlorpyrifos; (●) dichlorvos; (□) captan.

Disappearance of Pesticides in the Cooking Process. Dichlorvos and captan are efficiently removed by cooking (Figure 4). Dichlorvos was vaporized by heat. The residual level was very low after 2.5 min. Captan is unstable in the homogenate, perhaps due to decomposition by the components of soybeans (Miyahara et al., 1992b; Wolfe et al., 1976). The decomposition rate was accelerated by heat. The residual level after 2.5 min was also very low. Malathion and chlorpyrifos were not removed efficiently by this process and remained (50–60% of initial levels) in the go. Those pesticides are stable under the conditions we employed in this experiment. However, the levels were reduced gradually, and to some extent, this process affects the residual levels of both pesticides. However, the conditions for the cooking process are too mild to remove the contaminated pesticides from the mixture. Thus, this process is effective in reducing pesticide levels only when the pesticides are volatile and/or unstable in homogenate.

Retention of Pesticides. Using the results in Table 3 and Figure 3, the pesticide residues in each step of tofu production are calculated and the results are shown in Table 4. The effect of washing was considered in these calculations. The final product is essentially free of pesticides. The process should be carried out carefully to reduce residual pesticides in tofu. Washing, coagulation, and rinsing processes are important stages for reducing pesticides levels in tofu.

In conclusion, significant pesticide reduction is achieved in processing soybeans into tofu even in the absence of drastic conditions such as deodorizing, which is used in oil refinement.

Table 4. Theoretical Retention (%) of Pesticides at Each Stage of Processing

sample	dichlorvos	malathion	chlorpyrifos	captan
soybeans	100	100	100	100
washed soybeans	5	11	19	11
soybean milk	4	7	11	1.1
okara	0.069	0.83	0.13	0.09
tofu	0.085	0.37	0.49	0.20

ACKNOWLEDGMENT

We are thankful for Mr. F. Inaba, Turumaki Junior High School of Tokyo, for information on tofu production and kind instruction for it. We are grateful to Dr. C. R. Warner, U.S. Food and Drug Administration, who reviewed the manuscript.

LITERATURE CITED

Addison, R. F.; Zinck, M. E.; Ackman, R. G.; Sipos, J. C. Behavior of DDT, Polychlorinated Biphenyl's (PCBs), and Dieldrin at Various Stages of Refining of Marine Oils for Edible Use. *J. Am. Oil Chem.* **1978**, *55*, 391–394.

Bates, A. N.; Rowlands, D. G. Interference by Extractives in the Determination of Malathion Residues in Rice Bran. *Analyst* **1964**, *89*, 286–287.

Cairns, T.; Siegmund, E. G.; Doose, G. M. Incidence of cis-4-Cyclohexene-1,2-dicarboximide in Fruit Crops Treated with Captan. *Bull. Environ. Contam. Toxicol.* **1983**, *30*, 117–121.

Chaudry, M. M.; Nelson, A. I.; Perkings, E. G. Distribution of Aldrin and Dieldrin in Soybeans, Oil, and By-Products during Processing. *J. Am. Oil Chem.* **1978**, *53*, 851–853.

Elms, K. D.; Kerr, J. D.; Champ, B. R. Breakdown of malathion and dichlorvos mixtures applied to wheat. *J. Stored Prod. Res.* **1972**, *8*, 55–63.

Hashemy-Tonkabony, S. E.; Soleimani-Amiri, M. J. Detection and Determination of Chlorinated Pesticide Residues in Raw and Various Stages of Processed Vegetable Oil. *J. Am. Oil Chem.* **1976**, *53*, 752–753.

Kanematu, H.; Chimi, K.; Maruyama, T.; Niiya, I.; Imamura, M.; Oshiba, K.; Kawakita, H.; Matumoto, T. Removal of Chlorinated Pesticide Residues from Crude Oils and Fats by Processing Procedures. *Yukagaku* **1974**, *23*, 49–52.

Miyagawa, T. *Syokuyou Abura Seizo no Jissai*; Saiwai Syobo: Tokyo, 1988; pp 25–29.

Miyahara, M.; Saito, Y. Pesticides removal efficiencies of soybean oil refining process. *J. Agric. Food Chem.* **1993**, *41* , 731–734.

Miyahara, M.; Sasaki, K.; Suzuki, T.; Saito, Y. Expanded coagulating cleanup procedures for simultaneous gas chromatographic determination of organophosphorus pesticides in crops and fruits. *Chem. Pharm. Bull.* **1991**, *39*, 1055–1058.

Miyahara, M.; Suzuki, T.; Saito, Y. Multi-residue Methods for Some Pesticides in Lanolin by Capillary Gas Chromatography with Detection by Electron Capture, Flame Photometric, Mass Spectrometric, and Atomic Emission Techniques. *J. Agric. Food Chem.* **1992a**, *40*, 64–69.

Miyahara, M.; Suzuki, T.; Saito, Y. Capillary Gas Chromatographic Determination of Captafol in Vegetables, Fruits and Grains. *J. Agric. Food Chem.* **1992b**, *40*, 1150–1153.

National Food Research Instates: Process technology for soybean food; Food process technology series; Ministry of Agriculture, Forestry and Fisheries: Tokyo, 1966; Vol. 4, Chapter 10, pp 22–37.

Norris, M. V.; Vail, W. A.; Averall, P. R. Colorimetric estimation of malathion residues. *J. Agric. Food Chem.* **1954**, *2*, 570.

Saito, K.; Saito, N.; Nagayoshi, H.; Suzuki, K. Studies on Thermal Decomposition of Pesticides II. Thermal Decomposition of Various Pesticides in Closed Tube. *Bull. Agric. Chem. Inspect. Stn.* **1981**, *21*, 25–31.

Sata, M. *Process and storage of Crops and Meats*; No-san-gyo son Bunnka Sya: Tokyo, 1988; Chapter 4, pp 100–103.

Smith, K. J.; Polen, P. B.; De Vries, D. M.; Coon, F. B. Removal of Chlorinated Pesticides from Crude Vegetable Oils by Simulated Commercial Processing Procedures. *J. Am. Oil Chem.* **1968**, *45*, 866–869.

Watanabe, A. *Tofu and related food, Sogo Syokuryo Kougyou*; Koseisya-Kouseikaku Co.: Tokyo 1970; Chapter 16, pp 306–321.

Watanabe, A. *Tofu and related food. Soybeans and Process I*; Kenpakusya Co.: Tokyo, 1987; Chapter 2, pp 32–62.

Wolfe, N. L.; Zepp, R. G.; Doster, J. C.; Hollis, R. C. Captan Hydrosis. *J. Agric. Food Chem.* **1976**, *24*, 1041–1045.

Received for review July 26, 1993. Accepted November 10, 1993.⊗

⊗ Abstract published in *Advance ACS Abstracts*, January 1, 1994.