

# PESTICIDE RESIDUE PROBLEMS IN JAPAN

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## Abstract

After the Second World War, pesticide residue problems have arisen in Japan as a result of the extensive use of organosynthetic pesticides, especially persistent pesticides such as organomercury fungicides and organochlorine insecticides. The successive problems encountered can be summarized according to the following chronological order.

1) Mercury contamination of rice grains, 2) Contamination of cow milk with BHC isomers, 3) Contamination of vegetables with dieldrin residues in soil, 4) Contamination of waterways with organochlorine insecticides, and 5) Pesticide effects on fish and shellfish.

## Introduction

After the Second World War, several organosynthetic pesticides which had been developed in America and in the European countries were successfully introduced to Japan for the control of pests, diseases and weeds of crops. It had been considered at that time that the efficacy of pesticides for the control of the pests should have the priority, while safety for the health of users was thought to be secondary. As a result little attention was paid to the pesticide residue problems in crops.

But in 1956, mercury contamination of rice grains resulting from the spraying of rice plants with organomercury fungicides was revealed first by Tomizawa and BHC and DDT residues were continuously detected in rice grains. In the 1960s, the contamination of many kinds of foods and of the environment by organochlorine insecticides was detected. Especially, severe contamination of cow milk with BHC isomers was first reported by Ueda in 1966.

As a result, there was an increasing awareness of the problem of pesticide residues not only on the part of government officials but also throughout the nation. Consequently, since 1963 the establishment of tolerance levels for pesticide residues in agricultural commodities and instructions for safe use of the pesticides have been implemented through the joint collaboration of the Ministry of Health and Welfare and the Ministry of Agriculture and Forestry. Moreover, the effect of pesticides on wildlife, especially fishes and shrimps became increasingly troublesome with the increasing amounts of pesticides used.

This review deals primarily with the pesticide residue problems in agricultural crops, and secondarily with the environmental problems of pesticides and their effects on wildlife.

## Mercury contamination of rice grains

Since the 1910s organomercury compounds have been extensively used as seed disinfectants. After 1945, it was found that these compounds were useful for the control of rice blast which is the most destructive disease of the rice plants in Japan, and dusts which contained phenylmercury acetate (PMA) were mainly sprayed on the leaves and stems of rice plants. These organomercury fungicides were so effective that the control of the incidence of blast under the conditions of bad weather and heavy fertilization of nitrogen was established, and this contributed to the yearly increase of rice yield. Also, these fungicides were effective not only for rice plant diseases but also for various diseases of vegetables and fruit trees, and they were successfully sprayed on the field

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crops. In foreign countries, organomercury fungicides have been mainly used as seed disinfectants, and only sprayed experimentally for the control of apple scab and tomato leaf mold disease in glasshouses. The spraying of organomercury fungicides directly on leaf and sheath of many kinds of crops was performed only in Japan.

In 1956, Tomizawa first reported that rice plants absorbed  $^{203}\text{Hg}$ -labelled mercury compound through not only leaf and sheath but also root, and that radioactive mercury was translocated into the rice grains when  $^{203}\text{Hg}$ -labelled PMA was sprayed on rice plants. This was the first warning against the indiscriminate use of pesticides in Japan. Based on these results, an Investigation Committee for the mercury residue problem was organized by the concerned scientists of the Ministry of Agriculture and Forestry, and a report was published in 1965.

Among the analytical results on a total of 80 samples of rice grains harvested in various districts in Japan in 1957, a part of the data is shown in Table 1. As seen in this Table, mercury

**Table 1 Relationship between the content of mercury residues in raw rice grains and treatment with phenylmercury acetate-dusts<sup>a</sup>.**

Location (variety)		Mercury contents of dust (%)	Hg, g/ha	Application		Hg ppm
				Date and growth stage <sup>a</sup>	No.	
Tohoku (Norin 41)	A	0.25	50	7-20(T), 8-8(J)	2	0.16
	B	0.25	100	7-20(T), 8-8(J)	2	0.20
	C	0.25	50	8-16(H)	1	0.17
	D	0.25	100	8-16(H)	1	0.23
	E	0.25	50	8-23(F)	1	0.19
	F	0.25	100	8-23(F)	1	0.40
	G	0.25	50	7-20(H), 8-23(F)	2	0.16
	H	0.25	100	7-20(H), 8-23(F)	2	0.40
	I	Untreated				
Kyushu (Jukkoku)	1	0.16	48	8-3(T), 8-11(T), 8-30(J)	3	0.09
	2	0.16	48	8-11(T), 8-30(J), 9-17(H)	3	0.16
	3	0.16	48	8-11(T), 8-30(J), 9-17(H), 9-26(F)	4	0.38
	4	Untreated				
Kantosan (Norin 25)	1	0.16	48	7-19(T), 7-27(T), 8-28(J)	3	0.24
	2	0.16	48	7-27(T), 8-27(J), 9-9(J)	3	0.56
	3	0.16	48	7-27(T), 8-27(J), 9-9(H), 9-26(F)	4	0.60
	4	Untreated				

<sup>a</sup> Abbreviations used: T, Tillering stage; J, Jointing stage; H, Heading stage; F, Full heading stage; 7, July; 8, August; 9, September.

contents were higher in rice grains from plants treated at later stages. Raw rice grains from treated plants contained 0.05 to 0.60 ppm mercury, with one exception of 1.31 ppm, and the average content was 0.20 ppm. Samples containing 0.1 to 0.2 ppm mercury accounted for 40% of the total samples. The average mercury contents in untreated samples was 0.04 ppm. The content of

mercury in rice grains was highest in bran, followed by husk and albumen (polished rice). About 40% of the mercury in raw rice grains was removed by standard polishing. Although the chemical form of residual mercury was not well known, it was considered that 11 to 17% of total mercury in raw rice grains was in the form of organomercury compounds. Organomercury compounds are generally absorbed by the roots of rice plants which are likely to absorb the water soluble and undissociated compounds.

Presently, the effect of mercury residues in rice grains on human health is not well known. However, an incident which was directly related to poisoning by organomercury compounds occurred in Japan at the same time. It became apparent that the outbreak of Minamata disease in the Prefectures of Kumamoto and Niigata was caused by the accumulation of methylmercury compounds in fish and shellfish from the drainage water of a factory. The mercury compounds used for rice blast were phenylmercury compounds, which differ from methylmercury compounds, and are less likely to cause Minamata disease. However, mercury as an element always remains in the natural environment. Moreover, it was found that microorganisms were able to synthesize methylmercury compounds from elemental mercury. Therefore, although mercury residues in rice grains do not affect immediately human health, prolonged use of organomercury fungicides leads to an increase in mercury residues in human body and environment, and its effects on human health may become quite unexpected. As this problem was related to the safety of the staple food of the nation, it had to be tackled. On the other hand, with the development of substitutes for organomercury fungicides for rice blast control, sprays of organomercury fungicides on rice plants were eventually prohibited in 1968 by the authorities concerned of the Ministry of Agriculture and Forestry. Registration and production of organomercury fungicides for the control of horticultural crops and soil diseases were banned in 1970. At present, the use of organomercury compounds for seed disinfection has also been prohibited.

According to the statistics, total consumption of organomercury fungicides during the period 1953–1972 in Japan amounted to 2,325 tons as metallic mercury. The acreage of rice fields in Japan at that time was approximately 3,200,000 ha, and therefore, the amount of mercury applied per ha was 0.726 kg on the average. If mercury at this dosage is assumed to remain chiefly in the soil layers extending from the surface to a depth of 10 cm, and if one takes into account the specific gravity of soil, mercury residues can theoretically be estimated to amount to 0.726 ppm. However, in 1973 four years after the use of mercury compounds had been banned, the amount of mercury residues in the surface of 13 paddy soils in the Prefectures of Niigata and Kagawa ranged from 0.13 to 0.44 ppm. If the amount of mercury present in the soil is assumed to be 0.1 ppm, it can be considered that about 70% of mercury applied in the past has been lost through volatilization and/or leaching. On the other hand, PMA has been applied to soils at the dosage of 3 kg and 15 kg (as metallic mercury) per ha, which corresponds to a 20 and 100 year's continuous application, respectively. Though an amount of 4.2 and 5.3 ppm of mercury residues in the paddy soils remained in 1973 after 8 years of application, grains of raw rice harvested from these paddy fields in 1973 contained only 0.05 and 0.07 ppm mercury, respectively. This result suggests that organomercury compounds in soil are rapidly converted to a form which is not readily taken up by rice plants. Moreover, there was no evidence of the existence of phenylmercury compounds in the soil after 8 years. This problem of contamination with mercury compounds is a typical instance of direct contamination of agricultural commodities with pesticides.

### **Contamination of cow milk with BHC isomers**

The promotion of countermeasures for the control of pesticide residues thus became a pressing need. Monitoring surveys on pesticides residues in market foods started from 1964 and were organized by the Ministry of Health and Welfare for the establishment of the tolerance levels of pesticide residues in foods. Although the research aimed first at selecting agricultural commodities related to practical use and diet habits, a survey on dairy products was started in 1969. As a

result, severe contamination of market cow milk by BHC isomers was discovered in 1969, and became an important social problem.

BHC had been extensively used for rice plant cultivation and forestry since 1949. Technical grade BHC consisted of the following ratios of isomers on the average: 68% of  $\alpha$ -BHC, 8.5% of  $\beta$ -BHC, 16% of  $\gamma$ -BHC, 5.5% of  $\delta$ -BHC and 2% of other impurities. Only  $\gamma$ -BHC is endowed with insecticidal activity and this product is called lindane. Lindane formulation had been mainly used for the control of insects in house, orchards and vegetable gardens in Japan. But the formulation of technical grade BHC had been used for the control of rice stem borers and leafhoppers for economical reasons. The amount used totalled 33,000 to 40,000 tons each year.

Though each isomer of BHC had been detected in cow milk, in general, the amount of  $\beta$ -BHC was particularly high. Based on the analytical results of cow milk from January to February 1970, the content of  $\gamma$ -BHC ranged from 0 to 0.05 ppm, while that of  $\beta$ -BHC ranged from 0.001 to 2.68 ppm. The amount of  $\alpha$ -BHC which accounted for the highest ratio was only less than half that of  $\beta$ -BHC. The amount of  $\delta$ -BHC was similar to that of  $\gamma$ -BHC. The analytical results of raw cow milk in 1970 are shown in Fig. 1. Generally, the contamination of cow milk with BHC

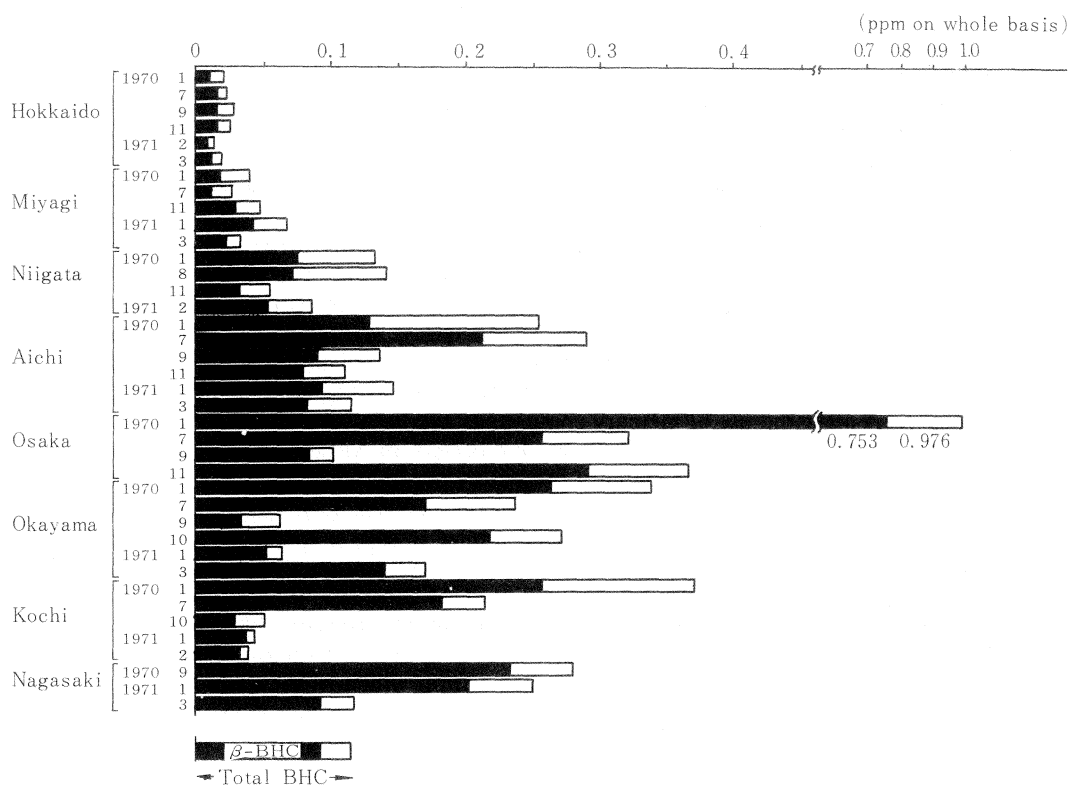


Fig. 1 Variation in the amount of BHC residues in raw milk with the location and season.

from different areas almost paralleled pesticide use. The contamination levels of BHC in the south-western part of Japan were generally higher than those of the north-eastern part of Japan including Hokkaido, Tohoku and Kanto, in spite of the fact that these areas produced a large

amount of cow milk. Moreover, DDT and dieldrin were also detected in cow milk, at the lowest amounts.

Research on the feeding methods of milking cows and analysis of BHC in feed were carried out by the Ministry of Agriculture and Forestry. On the basis of the results obtained, the following facts became apparent. There was a significant correlation between the degree of BHC contamination of cow milk and that of feed. The contamination levels of cow milk from cows fed rice straw as roughage were high. On the other hand, there was a high level of contamination with BHC in rice straw, while the content of BHC in the other feeds was very low. The amounts of BHC residues in rice straw were correlated with the application time of the pesticide, and when the application took place at the later stages of growth to control rice leafhoppers, the amount of residues was about ten times higher than when the pesticide was applied in the early stages of growth to control rice stem borers. It is interesting to note that the ratio of  $\gamma$ -BHC to  $\beta$ -BHC in rice straw was 1:4 in spite of the fact that the ratio of these compounds in technical grade BHC was 1:0.7. Moreover, the ratio in cow milk was 1:50 to 100. It appears that  $\beta$ -BHC is less volatile than  $\gamma$ -BHC, and is liable to remain in agricultural crops. It is considered that the speed of metabolism of  $\beta$ -BHC in animal body is lower, and that the compound tends to accumulate in animal adipose tissue.

Also the BHC residues in rice grains amounted to about 0.15 ppm as total BHC in the case of standard use, and the ratio of  $\beta$ -BHC was only a little higher than that in the formulation. From the above results, it was considered that the main route of BHC contamination of cow milk was represented by the use of rice straw as feed, to which BHC had been applied.

During the survey on cow milk, data on the contamination of human milk with  $\beta$ -BHC were reported, and the urgent need for a broad survey discussed. According to the analytical results of samples which were collected from hospitals within Tokyo City in 1971 (Iizuka *et al.*, 1971), the total amount of BHC residues in human milk ranged from 0.01 to 0.117 ppm, with that of the  $\beta$ -BHC residues ranging from 0.007 to 0.081 ppm, and the total amount of DDT ranged from 0.01 to 0.103 ppm, with that of dieldrin ranging from 0.0012 to 0.0087 ppm. The ratio of  $\gamma$ -BHC to  $\beta$ -BHC in human milk which was 1:7, was not particularly higher than that in cow milk.

From these results and others on the extensive contamination of wildlife such as fishes, birds and environment with organochlorine insecticides, the use of BHC in agricultural crops was prohibited by the Ministry of Agriculture and Forestry in 1971. Since BHC had been mainly used in paddy fields, and underwent rapid degradation under irrigation conditions, it was considered that it might be possible to disregard the effect of soil contamination with BHC applied in the past.

### Contamination of vegetables with dieldrin residues in soil

The quantity of aldrin used for the control of soil-borne insects has amounted to about 240 tons annually as technical grade product since 1954, mainly in areas where vegetables and tobacco are being cultivated. In December of 1969, tolerance levels of the residues of this insecticide were defined in 12 crops such as cucumber, potato, etc. The value expressed as the sum of aldrin and dieldrin level was very low, that is 0.02 ppm in cucumber, cabbage, radish, eggplant, etc, but the tolerance level could not be defined in the other crops. This residue tolerance level was established based on the acceptable daily intake for humans of the sum of aldrin and dieldrin level, that is 0.0001 mg/kg/day. It was already well known that aldrin was oxidized to dieldrin in the soil and living organisms. The Ministry of Agriculture and Forestry had been well inspired to prohibit the use of aldrin and dieldrin for the control of cucumber and potato. However, in the autumn of 1975, the Hygiene Institute of some Prefectures located in the production districts found that several market samples of cucumber and potato contained a quantity exceeding the tolerance level, so that shipment and discharge of these agricultural products were prohibited. Investigations showed that in one instance aldrin had been intentionally used by the farmer, but there were also

cases in which dieldrin was detected although aldrin had not been used in that particular year. Thereafter, extensive studies on residues of cyclodienes such as aldrin, dieldrin and endrin in soil and the uptake of these compounds by crops were carried out. On the basis of the results, the following facts have become apparent. Aldrin is converted rapidly to dieldrin in soil, and both compounds persist in soil for a long period of time. Therefore, the soil of fields where a large amount of aldrin had been used in the past became contaminated with dieldrin. Aldrin and dieldrin in soil were gradually lost by evaporation and degraded by microorganisms. As the speed of disappearance differs markedly depending on soil texture, temperature and cultivation conditions, etc., the amount of residues is reduced by one half within one year. As the level of uptake of aldrin and dieldrin in soil by crops varied with the kinds of crops and the soil texture, crops which absorbed the largest quantities were carrot, cucumber and peanut, followed by potato, radish, soybean and strawberry. Crops which did not absorb readily the compounds were tomato, eggplant, green pepper, cabbage, Chinese cabbage, lettuce, wheat, etc.

From the above results, aldrin and dieldrin were designated as soil residual pesticides with the revision of the Agricultural Chemicals Regulation Law in 1971, and the use of these insecticides was strictly regulated. Thereafter, registration of these insecticides was discontinued, and they are not any longer being marketed presently. Moreover, it was suggested that crops with a low capacity of absorption of these pesticides should be cultivated in soil with a high degree of contamination.

At present, levels of dieldrin residues amounting to 0.001 to 0.01 ppm can still be detected in some agricultural fields. As for the cause of the residues, it has been proposed that the degradation speed of dieldrin in soil can be retarded remarkably depending on the soil texture and weather conditions, and that the degradation of absorbed compounds may become difficult due to the activity of microorganisms.

### Contamination of water systems with organochlorine insecticides

Organochlorine insecticides such as DDT, BHC, dieldrin generally tend to persist in natural environments. Moreover, these insecticides are soluble in fat, so that they are likely to be accumulated into animal adipose tissue through food chain. Therefore, the contamination of the environment and wildlife with these insecticides has been observed worldwide.

The use pattern of organochlorine insecticides in Japan is characterized by larger amounts of formulations of technical grade BHC for agricultural production than in foreign countries. Based on the sales statistics, the use of BHC has increased annually to reach a peak in 1968, when 890 grams of  $\gamma$ -BHC were applied per hectare of arable land.

Monitoring surveys on the situation of organochlorine insecticides in the environment started from 1968 in Japan. The analytical results of BHC residues obtained by Tatsukawa *et al.* (1972) are summarized as follows. Since the residue levels of BHC in soil, water, air and various biota are roughly proportional to the amounts used, the western part of Japan would be expected to be more contaminated with BHC than the eastern part of the country. BHC residue levels in paddy soils are also higher in the western part of Japan than in the eastern part, but the overall concentrations of BHC in paddy field soils are not as high in spite of intensive spraying. BHC is degraded faster under water-logged conditions than under upland conditions. Among the BHC isomers,  $\gamma$ -BHC is the most rapidly degraded compound, followed by  $\alpha$ -BHC,  $\beta$ -BHC and  $\delta$ -BHC. Consequently, soils following rice harvesting contain high levels of  $\beta$ -BHC. The total load of BHC in the environment in Japan is estimated to be 2,000 to 7,000 tons, of which more than 90% is in soils, especially in arable soils. Approximately half of the total consists of  $\beta$ -BHC, and  $\gamma$ -BHC accounts only for a small part, probably less than 10% of the total. Since the persistent pesticides eventually reach the waterways, water samples from rivers, lakes, sea and rain are a good index to evaluate the environmental contamination with pesticides. Analysis of rain water in 1968 in the Tokyo area showed that it contained 0.195 ppb of DDT and its derivatives, 0.454 ppb of

$\alpha$ -BHC, 0.220 ppb of  $\beta$ -BHC, 0.388 ppb of  $\gamma$ -BHC and 0.171 ppb of  $\delta$ -BHC, and seasonal fluctuations in the BHC concentrations were also observed. Based on these results, it appears that at that time, the total BHC residue concentration was the highest in rain water, followed by water from river, sea, tap water and underground water in this order. In contrast, the total amounts of DDT residues were the highest in river water, followed by rain water, suggesting that BHC is more vaporized to the atmosphere than DDT.

Yamato *et al.* (1975) have studied the changes of BHC residue levels in the Onga River and Murasaki River of the Kitakyushu district of Japan from 1970 to 1974. The results for the Onga River are shown in Fig. 2. Furthermore in 1970 when the use of BHC was banned, thereafter

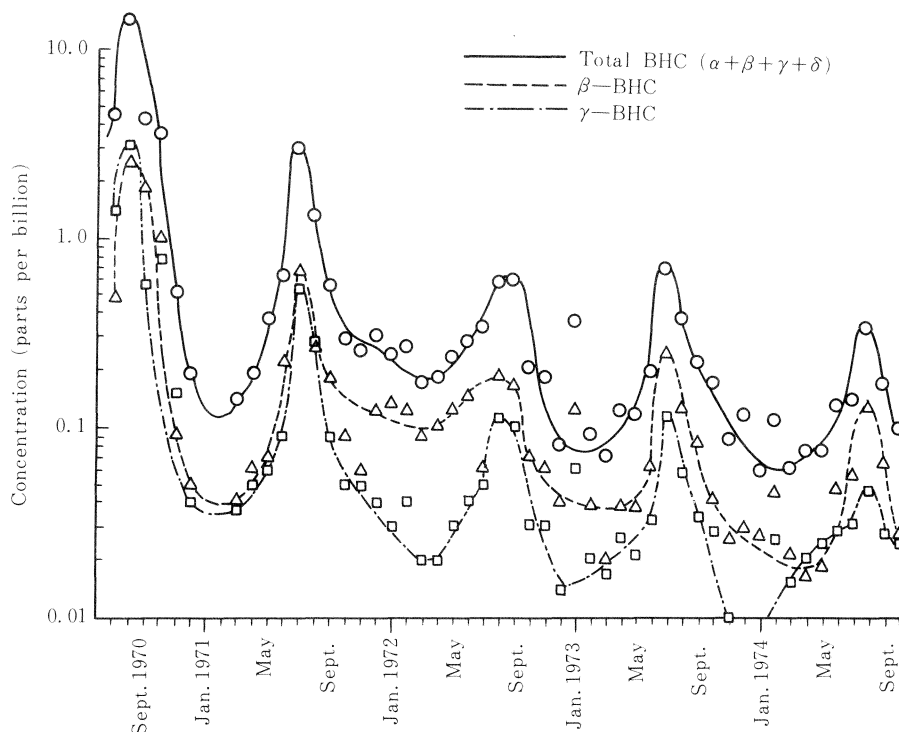


Fig. 2 Persistence of BHC in surface water samples collected in the Onga River, Kitakyushu.

changes in the BHC residues in river water were investigated. In the autumn of 1970, the total levels of BHC residues in the water of both rivers exceeded 10 ppb, but afterwards, the residue levels decreased exponentially, that is about 3 ppb in the summer of 1971, and about 0.7 ppb in the summer of 1972. Afterwards, from 1972 to 1974, the reduction speed was lower. The total levels of BHC residues in river water in the summer of 1974, 4 years after the use of the pesticide was banned, were 0.207 ppb in the Onga River and 0.252 ppb in the Murasaki River, respectively. Similarly high values recorded from 1972 to 1974 were considered to be due to the runoff of BHC residues from water of paddy fields. The maximum value in the summer season was 7 to 8 times higher than the minimum value of the winter season, presumably because the BHC residues in soil and water ways flowed away to the river through rain due to the high temperature in summer.

The contamination of natural waterways with the other pesticides except organochlorine

insecticides was almost limited to the period when the pesticides were used. Analysis of the water of the main irrigation canals in Tochigi Prefecture from June to August of 1974 showed a content ranging from 0.12 to 3.13 ppb for diazinon, 0.01 to 0.98 ppb for fenitrothion and 0.06 to 0.85 ppb for IBP (Hanu *et al.*, 1975). Analysis of sea water in Kagoshima bay from May to October of 1977 showed values ranging from 0.02 to 2.4 ppb for diazinon and 0.03 to 0.48 ppb for fenitrothion (Deshimaru, 1977).

Suzuki *et al.* (1977) detected 0.11 to 10 ppb of the herbicide thiobencarb in rivers and agricultural drainage water only immediately after application of the pesticide to the paddy fields in the northern district of Kyushu. However, thiobencarb residues could not be detected in water samples after the termination of the application of the herbicide to rice paddies, suggesting that the applied thiobencarb might have been degraded by microbial and physico-chemical activities such as photochemical reactions occurring in the soil surface.

Subsequently, Yamato *et al.* (1980) first reported the occurrence of the herbicide oxadiazon in surface and tap water. The high concentration of oxadiazon residues in water was detected immediately after the beginning of rice seedling transplantation and rapidly decreased in the Higashitani and Murasaki Rivers. The residue levels ranged between 0.001 and 1.953 ppb. The concentration of oxadiazon residues in tap water was about 3 to 23% that in raw water.

### Pesticide effects on fish and shellfish

As parathion had been used extensively for the control of the rice stem borer since 1952 in Japan, in the summer of the next year, the production of the krill, *Acetes japonicus* in Ariake bay was rapidly reduced. In 1954, the research staff of the University of Kyushu and Fisheries Experimental Station of Saga demonstrated that the cause of the decrease could be ascribed to the runoff of parathion from the water of paddy fields. This was the first report of damage to fisheries caused by pesticides in Japan. It was found that parathion was highly toxic to crustaceans. According to the studies conducted by Tomiyama *et al.* (1962), the  $LC_{50}$  (24 hrs) of parathion ranged from 0.01 to 0.033 ppb for *Acetes japonicus*, 0.11 ppb for *Metapenaeus joyneri* and 6.3 ppb for *Penaeus setiferus*.

Pentachlorophenol (PCP) had been used extensively for the control of weeds in paddy fields since 1956 in Japan. However, PCP is highly toxic to fishes. According to the survey conducted by the University of Nagoya and the Fisheries Experimental Station of Shiga, the  $LC_{50}$  (48 hrs) of PCP was 0.35 ppm for the carp, 0.28 ppm for the Japanese catfish, 0.056 ppm for the rainbow trout, 0.227 ppm for *Corbicula sandai*, and 0.38 ppm for *Charybdis japonica*. Also, massive death of the population of the bivalve, *Corbicula sandai* occurred on the shores of Lake Biwa in the City of Hikone, in July of 1960. Subsequently damage to the crucian carp, folktailed bullhead and lakeweed club was observed in the rivers and ponds of the City of Kusatsu. In 1961, extensive damage to fish and shellfish presumably due to PCP occurred in the Prefectures of Fukuoka, Kumamoto and Nagasaki. In 1963, in order to prevent the damage to the fishes, regulations for the use of the pesticides were enacted. Moreover, in 1971, PCP was incorporated in the list of pesticides responsible for water pollution following the revision of the Agricultural Chemicals Regulation Law. Afterwards, the amounts of PCP used decreased rapidly with the development of the other herbicides with low toxicity to fishes.

The herbicide molinate introduced from the U.S.A., has been used for the control of weeds after rice seedling transplantation since 1971. The acute toxicity of this herbicide to Japanese carp was relatively low, that is the  $LC_{50}$  for 48 hrs was 34 ppm. But the  $LC_{50}$  (48 hrs) of molinate to the rainbow trout was 0.29 ppm. Since 1974, massive death of breeding carps has been frequently observed in ponds near paddy fields in the Prefectures of Fukushima and Nagano. In these cases, the symptoms consisted of hemorrhage affecting mainly the gills, and it was soon revealed that the resultant anemia was induced by molinate after a few days of exposure to low concentrations of the herbicide. The pond water which caused the death of the carps contained



more than 10 ppb of molinate, with a maximum concentration of 50 ppb. On the basis of the studies conducted by Kawatsu (1977), subacute toxicity of this herbicide to carp corresponded to a  $LC_{50}$  (21 days) of 0.18 ppm. Due to this accident, the use of molinate was prohibited in the breeding districts of carp. The need to establish tests of subacute or chronic toxicity of pesticides to aquatic organisms was emphasized by this accident.

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### Discussion

**Ishikura, H.** (Japan): Could you explain the reason for the existence of seasonal fluctuations in the level of BHC residues in the Onga River water with high peaks in the summer after 1972 (the pesticide was banned in 1971). Could the high level of residues be caused by flooding of water in rice fields in the summer season?

**Answer:** I assume that the high levels observed in the summer can be ascribed to the fact that in this season the temperature is higher than in winter and precipitation is more abundant. As a result, runoff of BHC residues from paddy fields to the river is more likely to occur in the summer than in the winter.