Japanese Journal of Plant Science ©2007 Global Science Books

Drins Residue in Tokyo Soils and Regulation against Soil Pollution in Japan

Yoshiko Hashimoto

Tokyo Metropolitan Agriculture and Forestry Research Center, 3-8-1 Fujimicho, Tachikawa, Tokyo 190-0013, Japan

Correspondence: **y-hashimoto@tdfaff.com**

ABSTRACT

Dieldrin is an organochlorine insecticide and one of 12 chemical substances known as persistent organic pollutants (POPs). In Japan, aldrin and dieldrin were first registered in 1954, and used on farmland for 22 years until 1975 when the registration lapsed. Although dieldrin has not been used for 30 years, it remains in cucumbers at levels exceeding the tolerable limit for pesticides even today. Soil samples were obtained from 814 farms in Tokyo in 2002. Dieldrin residue was detected in 85 soil samples at concentrations ranging from 10 ng/g dw (dry weight) to 2600 ng/gdw. In this review, the actual residual level of dieldrin in Japan is compared with that in some other countries. In addition, although there is no tolerance for pesticide residue in soil, Japanese regulation against soil pollution is summarized.

Keywords: aldrin, dieldrin, endrin, organochlorine pesticide, persistent organic pollutants (POPs), persistent soil contamination

CONTENTS

INTRODUCTION

The pollution by persistent organic pollutants (POPs) is a global issue that needs international consideration and attention (Barra *et al*. 2006). Almost all POPs are organochlorine pesticides, and their use is now prohibited. However, they are very persistent, and are still present in the environment. Dieldrin over the tolerance level (0.02 ppm) for pesticide residue was found in cucumbers produced in Tokyo in 2002 (Kondo *et al*. 2003). Dieldrin is one of 12 chemical substances known as POPs (Jorgenson 2001). Although dieldrin has not been used for 30 years in Japan, it remains in cucumbers at levels exceeding the tolerable limit for pesticides. This is a very serious problem in Tokyo agriculture. Therefore, we examined residual concentration of dieldrin in soils in Tokyo in 2002. The horizontal distribution and the vertical distribution of dieldrin on some farms were also examined (Hashimoto 2005).

In this review, the data we obtained is presented and compared with dieldrin residual levels in some foreign countries at present. In addition, a system for surveillance of pesticide residue in soil in Japan is outlined.

DRINS USED IN THE PAST

Development and property

Dieldrin, aldrin and endrin are collectively called drins or drin pesticides (**Table 1**). These chemicals were synthesized from pentadiens which were obtained as secondary products of petrochemistry through the Diels-Alder reaction in 1948-1951 in the USA. Dieldrin is an oxidized form of aldrin, and is a stereoisomer of endrin. Dieldrin and endrin were effectively used as a foliar spray because their vapor pressure are low, while aldrin was used as a soil treatment because its vapor pressure is ten times higher than dieldrin and endrin and it also has a fumigative effect. Drins have lower mammalian toxicity and water pollution and higher insecticidal activity than DDT (dichloro diphenyl trichloroethane) or BHC (benzene hexachloride). Both DDT and BHC are organochlorine insecticides and POPs, the same as drins. They were developed in 1938-1942, a little before the synthesis of drins. Although they were widely used for upland and paddy pest insects, their use was replaced by drin use following their appearance, and lapsed in 1971 before the lapse of

Received: 3 March, 2007. Accepted: 9 April, 2007.

Invited Mini-Review

Table 1 Information for drins.

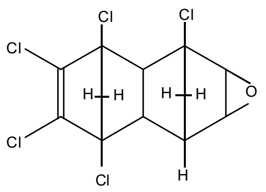
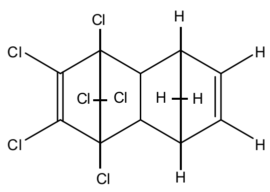
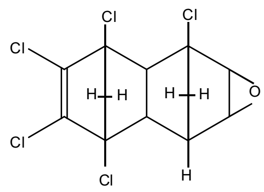
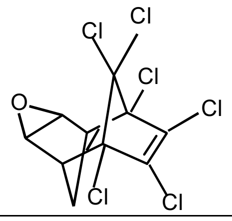
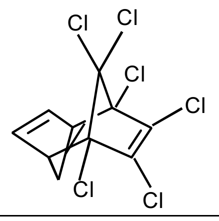
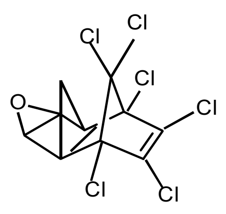
Name	**Dieldrin**	**Aldrin**	**Endrin**
Chemical structural formula			
Steric formula			
Molecular formula	$C_{12}H_8Cl_6O$	$C_{12}H_8Cl_6$	$C_{12}H_8Cl_6O$
Molecular weight	380.91	364.91	380.91
IUPAC	1,2,3,4,10,10-hexachloro -6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-exo-5,8-dimethanonaphthalene	1,2,3,4,10,10-hexachloro -1,4,4a,5,8,8a-haxahydro -1,4-endoexo-5,8-dimethanonaphthalene	1,2,3,4,10,10-hexachloro -6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-endo-1,4-exo-5,8-dimethanonaphthalene

Table 2 Use of drins in several countries around the globe in 1967.

	Japan	**USA**	**Canada**	**Italy**	**Greece**	**Poland**	**Spain**	**India**	**Israel**	**UAE**
Drins[a]	363	39,140	309	4,449	132	180	132	1,365	123	520
Cultivated area[b]	5,753	176,440	43,404	15,213	3,851	15,518	20,482	162,434	411	2,801
a / b[c]	63.1	221.8	7.1	292.4	34.3	11.6	6.4	10.3	299.3	185.6

[a] Effective ingredient conversion, t
[b] 1000 ha
[c] kg / ha
Source: Goto 1970

drins. Since aldrin particularly indicated very excellent activity on soil pest insects, namely *Gryllotalpa orientalis*, *Collembola*, *Phylloxeridae*, *Paragordius tricuspidatus*, *Tipula aino*, and others, it was applied to soil as a dust formulation or pesticide-fertilizer mixture. Aldrin is readily converted to dieldrin in soil, because much monitoring data in the 1970s detected dieldrin after the application of aldrin only in soil (Machimura and Nasuda 1972; Ishimoto *et al.* 1973; Suenaga 1973; Beyer and Gish 1980; Nagami 1997). Aldrin changes into dieldrin and its half-life is about 1.5-5.2 years in soil. The half-life of dieldrin is longer, 2-15 years in soil. In addition, it takes 25 years for 90% of dieldrin to disappear (Kiritani 1971; Jorgenson 2001).

Use of drins in Japan

In Japan, aldrin, dieldrin and endrin were first registered in 1954, and used on farmland for 22 years until 1975 when the registration lapsed (Ministry of Agriculture, Forestry and Fisheries). In 1967, for example, the quantity of drins used in Japan was 360 t as the amount of active ingredient, and they were mainly used for cultivating vegetables. The recommended time of application for drins detailed in the Direction for safe use of agricultural chemicals was as follows: (1) aldrin dust containing 4% of ingredient incurporated 20-30 kg/ha for control of soil pest insects, its suspension containing 40% of ingredient drenched after 1000-5000 times dilution for control of soil pest insects, and its emulsion containing 24% of ingredient sprayed after 300-500 times dilution for control of soil and aerial part pest insects; (2) dieldrin dust containing 4% of ingredient incorporated 30 kg/ha for control of soil pest insects, its suspension containing 50% of ingredient sprayed after 500-1500 times dilution for control of aerial part pest insects, and its emulsion containing 18.5% of ingredient sprayed after 200-500 times dilution for control of aerial part pest insects or drenched for control of soil pest insects; (3) endrin dust containing 1.5-2% of ingredient incorporated 30-60 kg/ha for control of soil pest insects but with a lower effect than aldrin and dieldrin, its emulsion containing 19.5% of ingredient sprayed after 200-1000 times dilution for control of aerial part pest insects, and its granules containing 2-5% of ingredient could be used similarly to its dust (Fukunaga 1967).

The tendency of past drins use was not exactly clear, however it can be guessed as follows. All prefectural governments in Japan made independent annual manuals for pest control which reflected local necessity for pest control from the 1950s. It is likely that farmers used pesticides according to this manual in the 1950s-1960s, because they could not get any kind of information on pesticide use that is available nowadays. Therefore, it is reasonably that the mentions in the manual suggest drins use in the past almost exactly. For example, the manual of Tokyo in 1969 noted the use of aldrin dust for rice, wheat, tomato, eggplant, green pepper, burdock, potato, cucumber, pumpkin, sweet potato, lettuce, green soybeans, rape vegetables and watermelon, the use of endrin emulsion for cucumber, pumpkin, sweet potato, lettuce, green soybeans, rape vegetables, watermelon, broad bean and celery while mention of dieldrin was only as an emulsion for watermelon. Therefore, in Tokyo, it is possible that almost all the residual dieldrin at present originates from soil-applied aldrin. On the other hand, the use of endrin was mentioned as an emulsion in the manual. In other prefectures, also, aldrin was treated on soil and dieldrin and endrin were applied on foliage (Hankawa 1993).

Use of drins outside Japan

In 1967, drins were used in Canada as they were in Japan, and more than four times the amount of drins were used in India compared to Japan, more than ten times the amount were used in Italy, and a hundred times more drins were used in the USA (Goto 1970). They seemed to be used for corn cultivation in the USA, although no information could be obtained on any of the other countries (Jorgenson 2001). Considerable quantities of drins were used in other countries, too. The amount of drins used per unit area in Japan was lower than that in the USA, Italy, Israel and UAE (**Table 2**).

PRESENT DIELDRIN CONCENTRATION IN SOIL

Drins concentration in Tokyo soils

The sampling test for pesticide residue in food-containing agricultural crops and processed food goods was done based on the Food Sanitation Law by the institutes of health sciences in every prefecture in Japan. Samples are generally collected at sales points but not at production locations, therefore, who produced the sample is unknown. However, in recent years, in Tokyo, direct sales style has increased by producers in direct sales shops and in front of their fields, and the number of samples that could be traced back to the producer increased. In 2002, cucumbers which contained dieldrin exceed the tolerance level specified by their producer and the production location. This resulted in the clearing of fields containing dieldrin in soil. Such a case has seldom happened, but might be a reality in Tokyo from now on. Tokyo, being the biggest consumption area, attracts the attention of food safety, which is ignored in agriculture areas. Therefore, it was necessary that we pay attention to field soil in Tokyo, and study drins residue in soil in Tokyo for the establishment of safe agriculture.

Soil samples were obtained from 814 places in Tokyo in 2002. All sampling points were used as a field. Almost all samples were Andosol. The number of fields sampled was decided based on the area ratio of farmland in Tokyo. There is little farmland within a 15-km radius and beyond a 45-km radius of Tokyo Station. Most agricultural land in Tokyo lies between 15 km, which is residential, to 45 km from Tokyo Station. The soil was collected from five sites chosen at random and from a depth of 0-15 cm using a core sampler 10 cm in diameter.

Aldrin was not detected in any soil samples, and endrin was detected in only three. On the other hand, dieldrin was detected in 85 of the 814 samples, i.e., 10.4%. In 70% of the samples in which dieldrin was detected, the concentration of residue was 100 ng/gdw or less. The concentration was 500 ng/gdw or less for 95% of these samples (Hashimoto 2005).

The horizontal distribution of dieldrin was examined in three places where dieldrin remains on farms in Tokyo. The area of the three sampling plots were 10 m × 30 m, 10 m × 36 m and 20 m × 20 m, respectively, and the number of soil samples was 15, 18, and 25, respectively. Dieldrin was not uniformly distributed, with the concentration varying from less than the limit of detection (10 ng/gdw) to 730 ng/gdw, the limit of detection to 160 ng/gdw and from 60 ng/gdw to 260 ng/gdw, respectively. The area of each field was between 300 m^2 and 400 m^2, which is normal for the cultivation of a single crop in Tokyo. Even over a distance of only two or three meters within one small farm, dieldrin residue levels are 4-70 times different (Hashimoto 2005). Residual dieldrin in Tokyo soil diffused during long term after use, although soil-applied pesticide was generally detected on every site on the field just after application.

The vertical distribution of dieldrin was examined in three places where there were differences in the horizontal distribution of dieldrin remains on farms in Tokyo. One had not been plowed deeply over the past 30 years, while the others were regularly plowed deeply. The soil was gathered every 10 cm from the surface of the earth after the soil was dug down to a depth of 1 m. Dieldrin was distributed to a depth of 30 cm from the soil surface on farms which had never been plowed deeply. On the other hand, dieldrin was scattered from the surface to a depth of 50-70 cm on farms which had been plowed deeply. In about 1970, it was suggested that deep cultivation had a restraining effect on drin residues in crops, and a diluting effect of the residual amount in soil (Yamamoto and Nutahara 1974). Therefore, many fields were cultivated deeply. Deep cultivation is a method to improve soil physical property by the addition of permeability to soil, and is usually conducted reasons other than residue evasion. of the decision to plow deeply is made by each farmer, and it is regularly done among farmers who plow deeply. Dieldrin seldom moves in soil because of its low water solubility, and it was localized only at 30 cm from the soil surface 1-10 years after application, however residual dieldrin in soil moves automatically by plowing deeply (Kawahara *et al*. 1971; Kiritani 1971; Kawahara 1973; Otani 1988). It is thought that the use of a farm affects dieldrin distribution in the soil.

In other prefectures, drin concentrations in soil had been detected in the 1970s (**Table 3**). Aldrin and dieldrin were seldom detected in soil of paddy rice field in Hokkado in 1973 (Saito and Kitayama 1974) and in Sendai in 1978 (Takahashi *et al*. 1978). On the other hand, in vegetable fields, total concentration of dieldrin and aldrin was detected in Saitama at 20-850 ng/gdw, 9-319 ng/gdw and 10-30 ng/gdw in 1971, 1972-1975, 1977, respectively (Nakamura 1993), and in Hyogo 30-140 ng/gdw in 1977-1979 (Otani 1988). Separate concentrations of dieldrin and aldrin were detected in Sendai (Miyagi prefecture), Fukuoka and Niigata. No aldrin was detected while dieldrin at 442 ng/gdw, ND-324 ng/gdw, and 120-680 ng/gdw in the soil of vegetable fields in 1975, 1976 and 1978, respectively were detected in Sendai (Osawa *et al*. 1975; Hiroshima *et al*. 1978); aldrin at 19 ng/gdw and dieldrin at 745 ng/gdw was detected in the soil of lettuce and spinach fields in Fukuoka in 1978 (Suzuki *et al*.1978), and aldrin at 3 ng/gdw and dieldrin at 200 ng/gdw in the soil in Niigata in 1980 (Homma *et al*. 1980). There are few reports on the concentration of drins in soil after the 1980s.

Table 3 Dieldrin and aldrin concentration in soil in Japan.

Year	Prefecture	Sample	Dieldrin (ng/gdw)	Aldrin (ng/gdw)	Reference
1973	Hokkaido	paddy field	0	0	Saito 1974
1971	Saitama	vegetable field	20-850		Nakamura 1993
1972-1975	Saitama	vegetable field	9-319		Nakamura 1993
1977	Saitama	vegetable field	10-30		Nakamura 1993
1975	Miyagi (Sendai)	vegetable field	442	0	Osawa 1975
1976	Miyagi (Sendai)	vegetable field	ND-324	0	Hiroshima 1978
1978	Miyagi (Sendai)	vegetable field	120-680	0	Hiroshima 1978
1977-1979	Hyogo	vegetable field	30-140		Otani 1988
1978	Fukuoka	vegetable field	745	19	Suzuki 1978
1980	Niigata	vegetable field	200	3	Homma 1980

Table 4 Dieldrin and aldrin concentration in soil in recent years in selected countries around the world.

Year	Country (City)	Sample	Dieldrin	Aldrin	Reference
			(ng/gdw)		
1992	Jamaica	sediment	1.88	0	Mansingh 1995
1995-1996	Egypt	soil	8-9.5		Mohamed 1998
1996-1997	Thailand	soil	0.009-249.3	0.017-1.041	Thapinta 2000
1999	Alabama	soil	0.53-23.8[a]		Harner 1999
2002	Brazil	soil	4-11,838	2.8-24,280	Barra 2006
2002	Japan (Tokyo)	soil	20-2600	0	Hashimoto 2005
2005	China (Beijing)	sediment	7.59-36.0[b]		Xue 2005
2005	China	soil	5.04-214.9[a]		Zhou 2006

[a]: Total OCPs contained aldrin, dieldrin, endrin, heptachlor and heptachlor epoxide.
[b]: Total OCPs (top soil to 50 cm depth).

Drin concentration in other countries

In many countries, research and monitoring of organochlorine pesticides containing drins efforts have continued on soil and river sediment (Wegman and Hofstee 1982), water in rivers and bays (Zimmerman *et al*. 2000; Xu *et al*. 2007), surface water (Nwankwoala and Osibanjo 1992; Hung and Thiemann 2002), earthworms and fishes (Nair *et al*. 1995; Shokrzadeh *et al*. 2005) and air (Manay *et al*. 2004), among other examples. Focusing on soil after the 1990s, dieldrin and aldrin concentrations in soil were 0.009-249, 300 ng/gdw and 0.017-1041 ng/gdw, respectively in 1996-1997, in Thailand (Thapinta and Hudak 2000; **Table 4**). In China, the concentrations of total organochlorine pesticides (aldrin, dieldrin, endrin, heptachlor, heptachlor epoxide) in soil was 5-215 ng/gdw in 2005 (Zhou *et al*. 2006), and the total concentration of 31 pesticides containing organochlorine pesticides in the Beijing river sediment was 7.6-36 ng/gdw (Xue and Xu 2005). On the African continent, the concentration of total organochlorine pesticides containing dieldrin from the top to 50 cm depth soil was 8-9.5 ng/gdw in Egypt in 1995-1996 (Ahmed *et al*. 1998). On the American continent, dieldrin concentrations was 0.53-23.8 ng/gdw in Alabama, USA in 1999, 1.88 ng/gdw in Jamaica in 1992 and 4-118,380 ng/gdw in Brazil in 2002 (Mamsingh and Wilson 1995; Harner *et al*. 1999; Barra *et al*. 2006). Although these data should not be directly compared because of incomplete information and historical use of soil, it is suggested that dieldrin widely persists in soil all over the world at present.

Half-life of drins in soil

In Nagano prefecture in 1971, the total average concentration of aldrin and dieldrin was reported to be 400 ng/g dw in fields for chinese cabbage and burdock after aldrin had been used for ten years until 1971 (Kawahara *et al*. 1971).

In Tokyo in 2002, the concentration of dieldrin residue was less than 100 ng/g dw in almost all soil samples, but the concentration of high residual soil was 2600 ng/g dw. When dieldrin is used in a field at a rate of 0.6 kg/ha, the half-life of the soil obtained from the field is an average of 2.6 years, at 2.2 kg/ha, it is 4.1 years, and at 9.6 kg/ha it is 12.5 years (Beyer and Gish 1980; Jorgenson 2001). In brief, if the application rate of drins to soil increases 15 times, the half-life of drins becomes 5 times longer (Nagai 1973; Jorgenson 2001). From these findings one can conclude that it is not an unusual phenomenon for current dieldrin residue levels in Tokyo to range widely. It is thought that the various levels of dieldrin residue have been caused by treatment of various quantities of drins in the 1960's on each farmland.

Dieldrin residue in soil caused by aldrin use in Tokyo

The residual dieldrin concentrations in soil are high, suggesting either that the rate of pesticide degradation in soil is low, or that a lot of pesticide was applied to the soil.

Information on the cultivated acreage of Ando soils, pesticide purchase amounts, and the main crops harvested were selected from agricultural censuses on all farmers in Tokyo in February, 1965. It could be seen that the amount of pesticide purchased per cultivated acreage in cities with high dieldrin residues was more than in cities with low dieldrin residues. Moreover, in 1965, the main crop in cities with high residual dieldrin levels was wheat, used as a food crop (Tokyo Metropolitan Government 1965). Wheat is prone to being damaged by soil insect pests very soon after sowing, and drins were the most effective in preventing this (Masaki 1958, 1959; Fujiyoshi 1984). Aldrin, in particular, could be used by mixing with fertilizer at dissemination, because of the stability of the active ingredient under both acidic and alkaline conditions (Fukunaga 1967).

On the other hand, the residual concentration of pesticides in soil is also effected by its reduction rate and some soil properties participate in its reduction rate. The factors affecting the reduction rate of residual pesticides in soil are soil temperature, soil moisture, humus content, pH, availability by microbial action in soil (Adams 1973; Hankawa 1980; Ibrahim *et al*. 1981). The half-life of DCPA (3',4'-dichloropropionanilide) ranged from 92 d at 10°C to 18 d at 30°C and 49, 33, and 31 d in 0.1, 0.2, and 0.4 kg H_2O kg^{-1} soil moisture levels, respectively (Choi *et al*. 1988). The persistence of dieldrin residues in soil was proportional to the organic content of the soil when soil types were used with organic contents ranging from 1 to 70% (Harris and Snas 1972). Toxic activity of dieldrin increased as pH increased since the pH of a soil is a reliable soil property in predicting pesticide adsorption (Hermason and Forbes 1966; Adams 1973). BHC is easily degraded when there is a low level of nitrogen sources, for example in sandy soil rather than clay soil (Singh *et al*. 1989). This suggests that BHC was used by soil microbes because no other substrates were available (Singh *et al*. 1989). Additionally, some organochlorine pesticides are likely to be used by microbes when their adsorption to soil is weak (Ghadiri *et al*. 1995). Bioavailability can also be estimated by the strength of pesticide adsorption to soil. This adsorption strength increases in soils with higher moisture contents and larger CEC (Choi *et al*. 1988; Kottler *et al*. 2001). However, the chemical properties of the soil in Tokyo such as humus content, pH, EC and base cations, cation exchange capacity and total carbon contents did not differ between high and low dieldrin residual soil (results in submission). Therefore these soil factors might not affect different levels of residual dieldrin in Tokyo.

LAW CONCERNING SOIL CONTAMINATION IN JAPAN

Agricultural Chemicals Regulation Law in Japan

Regulation concerned drins in soil contamination are found in the Agricultural Chemicals Regulation Law and the Agricultural Land Soil Pollution Prevention Law while the Soil Contamination Provision Law targets no agricultural chemicals. The Agricultural Chemicals Regulation Law was legislated in 1948. When it was revised in 1971, dieldrin and aldrin were specified as a soil persistent pesticide and their use was regulated (Goto 1971; Sasaki 1973; Nakamura 1973; Yoshioke 1974). Their registration lapsed in 1975 in Japan and their use was prohibited in the Stockholm Convention on Persistent Organic Pollutants adapted in 2001 (Environment Agency 1977). Therefore, all mentions of dieldrin and aldrin as a soil persistent pesticide were eliminated from the Agricultural Chemicals Regulation Law in 2002.

The Agricultural Land Soil Pollution Prevention Law regulates only farmland and it specifies Cd (cadmium), Cu (copper) and As (arsenic) as specific hazardous substances, but drins are not mentioned.

Standard value for pesticide residue in soil in Japan

Environmental Quality Standards for Soil Contamination is prescribed for five pesticides, namely organophosphorus pesticides, 1,3-dichloropropene, thiuram, simazine and thiobencarb based on the Basic Environment Law. However there is no tolerance for drins. It is difficult to set a uniform standard value for an aged drin in soil and it might be significantly scarce. It is more important to clarify the actual drin uptake to crops in various statuses of drin residue in soil of many prefectures (Hashimoto 2007).

CONCLUDING REMARKS

There is much data of dieldrin in the environment, which ranges widely. The meaning or impact of such concentrations is not fully known but the existence of dieldrin in the soil or environment is not good. Therefore, a tolerance for drin residue in soil has not yet been legislated. A more extensive understanding about residue status in soil of dieldrin, drins and POPs is necessary.

REFERENCES

Adams RSJ (1973) Factors influencing soil adsorption and bioactivity of pesticides *Residue Review* **47**, 1-54

Ahmed MT, Ismail SMM, Mabrouk SS (1998) Residues of some chlorinated hydrocarbon pesticides in rain water, soil and ground water, and their influence on some soil microorganisms *Environment International* **24**, 665-670

Barra R, Colombo JC, Eguren G, Gamboa N, Jardim WF, Mendoza G (2006) Persistent organic pollutants (POPs) in Eastern and Western South American countries. *Reviews of Environmental Contamination and Toxicology* **185**, 1-33

Beyer WN, Gish CD (1980) Persistence in earthworms and potential hazards to birds of soil applied DDT, dieldrin and heptachlor *Journal of Applied Ecology* **17**, 295-308

Choi JS, Fermanian TW, Wehner DJ, Spomer LA (1988) Effect of temperature, moisture, and soil texture on DCPA degradation *Agronomy Journal* **80**, 108-113

Environment Agency (1977) The *status quo* of environmental pollution due to agricultural chemicals and its countermeasures *Environment White Paper* **52**, 261-264

Fujiyoshi N, Yamanaka M, Takahashi T (1984) Emergence inhibition by snowfleas, *Onychurus* spp. on wheat and several methods of control. *Bulletin of the Fukuoka Agricultural Research Center* **A-4**, 43-46

Fukunaga K (1967) *Pesticide Handbook*, Japan Plant Protection Association, pp 18-22

Ghadiri H, Rose CW, Connell DW (1995) Degradation of organochlorine pesticides in soils under controlled environment and outdoor condition. *Journal of Environmental Management* **43**, 141-151

Goto S (1970) Status and problem on pesticide use. *Shokubutu Boueki* **24**, 501-506

Goto S (1971) Revision of the Agricultural Chemicals Regulation Law. *Shokubutu Boueki* **25**, 123-127

Hankawa Y (1980) Studies on organochlorine insecticides in crops and soil (3). Effects of the humus content of soil on the persistence of aldrin, dieldrin and endrin. *Report of the Hiroshima Prefecture Agricultural Experiment Station* **3**, 60

Hankawa Y (1993) Studies on safe utilization and rational application of pesticides in crop production. *Bulletin of the Hiroshima Prefectural Agriculture Research Center* **59**, 1-169

Harner T, Wideman JL, Jantunen LMM, Bidleman TF, Parkhurst WJ (1999) Residues of organochlorine pesticides on Alabama soils. *Environmental Pollution* **106**, 323-332

Harris CR, Snas WW (1972) Behavior of dieldrin in soil: microplot field studies in the influence of soil type on biological activity and absorption by carrots. *Journal of Economic Entomology* **65**, 333-335

Hashimoto Y (2005) Dieldrin residue in the soil of agricultural fields and in cucumbers in Tokyo. *Journal of Pesticide Science* **30**, 397-402

Hashimoto Y (2007) Reduced dieldrin concentration in cucumber fruits using *Cucurbita* rootstocks and activated carbon. *Journal of Pesticide Science* **32**, in press

Hermanson HP, Forbes C (1966) Soil properties affecting dieldrin toxicity to *Drosophilia melanogaster*. *Proceedings of the Soil Science Society of America* **30**, 748

Hiroshima K, Kikuchi M, Sugano T, Takano F, Mishima Y, Magariya N (1978) Survey on pesticide residues 6th report on organochlorine pesticide residue in vegetables and soils. *Report of the Sendai Municipal Institute of Public Health* **8**, 268-270

Homma I, Mukai T, Maho M, Masuyama K, Soma K, Arakawa H (1980) Investigation on the residue of chlorinated cyclodienes – correlation of the residue between crop and soil. *Japanese Journal of Public Health* **27**, 644

Hung DQ, Thiemann W (2002) Contamination by selected chlorinated pesticides in surface waters in Hanoi, Vietnam. *Chemosphere* **47**, 357-367

Ibrahim O, Wheeloccck DE, Cotton JV (1981) Factors affecting soil residues of dieldrin, endosulfan, γ-hch, dimethoate, and pyrolan. *Ecotoxicology and Environmental Safety* **5**, 135-160

Ishimoto S, Yamamoto M, Nutahara M (1973) Provision for pesticide residue in Kouchi prefecture. *Shokubutsu Boueki* **27**, 425-427

Jorgenson JL (2001) Aldrin and dieldrin: a review of research on their production, environmental deposition and fate, bioaccumulation, toxicology, and epidemiology in the United States. *Environmental Health Perspectives* **109**, 113-139

Kawahara T, Takanuma S, Wada T, Kureha Y, Nakamura H (1971) Absorption and translocation of chlorinated hydrocarbons into crops in the high-cooling region. *Bulletin of the Agricultural Chemical Inspection Station* **11**, 67-72

Kawahara T (1973) Residue and fate of organochlorine insecticides in soil. *Shokubutu Boueki* **27**, 402-406

Kiritani K (1971) A Review on environmental pollution by chlorinated hydrocarbon insecticides. *Proceedings of the Association of Plant Protection, Sikoku* **6**, 1-44

Kondo H, Amakawa E, Sato H, Yasuda K, Onuki K, Akiba M, Kanaya K (2003) Case study on detection of dieldrin in cucumbers cultivated in Tama region, Tokyo. *Annual Report of Tokyo Metropolitan Institute of Public Health* **54**, 132-135

Kottler BD, White JC, Kelsey JW (2001) Influence of soil moisture on the sequestration of organic compounds in soil. *Chemosphere* **42**, 893-898

Machimura N, Nasuda K (1972) Residue in soil and uptake into crops of organochlorine pesticides. *Annual Report of the Hokuriku Plant Protection Society* **20**, 71-75

Mamsingh A, Wilson A (1995) Insecticide contamination of Jamaican environment. Baseline studies on the status of insecticidal pollution of Kingston Harbour. *Marine Pollution Bulletin* **30**, 640-645

Manay N, Rampoldi O, Alvarez C, Piastra C, Heller T, Viapiana P, Korbut S (2004) Pesticides in Uruguay. *Reviews of Environmental Contamination and Toxicology* **181**, 111-138

Masaki J (1958) Insect pest control of wheat at sowing. *Shokubutu Boueki* **12**, 437-440

Masaki J (1959) Studies on rice crane fly (*Tipula* Aino Alexander, Tipulidae, Diptera) with special reference to the ecology and its protection. *Kantotozan Nogyo Shikenjo Kenkyu Hokoku* **13**, 1-202

Ministry of Agriculture, Forestry and Fisheries. Access online: http://www.acis.go.jp

Nagai Y (1973) Provision on pesticide residue in Tokushima prefecture. *Shokubutu Boueki* **27**, 423-424

Nagami H (1997) Dieldrin and chlordane residue in agriculture fields. *Bulletin of Environmental Contamination and Toxicology* **59**, 383-388

Nair A, Dureja P, Pillai MKK (1995) Levels of aldrin and dieldrin in environmental samples from Delhi, India. *Journal of Environmental Management* **43**, 141-151

Nakamura H (1973) Crop persistent and directionfor safe use of agricultural chemicals. *Shokubutu Boueki* **27**, 397-401

Nakamura K (1993) Studies on behavior and fate of pesticides in soil and aquatic environment of agricultural lands. *Bulletin of the Saitama Agriculture Experimental Station* **46**, 1-124

Nwankwoala AU, Osibanjo O (1992) Baseline levels of selected organochlorine pesticide residues in surface waters in Ibadan (Nigeria) by electron capture gas chromatography. *Science of the Total Environment* **119**, 179-190

Osawa T, Takahashi K, Inoue Y, Sugano T, Mishima Y, Kato T (1975) On the results of determination of pesticides for agricultural use in irrigation water and soil in paddy rice field. *Annual Report of Sendai Municipal Institute of Public Health* **8**, 232-234

Otani Y (1988) Behavior of soil-applied pesticides in soil. *Bulletin of the Hyogo Central Agriculture Technical Centre* **12**, 133-147

Saito M, Kitayama M (1974) Studies on the prevention of poisoning due to agricultural chemicals, part 19, organochlorine pesticide residues in water and soil. *Annual Report of the Hokkaido Institute of Public Health* **24**, 126-127

Sasaki T (1973) Status and provision on pesticide residue. *Shokubutu Boueki* **27**, 395-396

Shokrzadeh M, Ebadi AG, Heidari R, Zaree S, Pourhosseini M (2005) Survey and measurement of residues of dieldrin and endosulphan in four species of fishes in Casplan sea, Iran. *International Journal of Biology and Biotechnology* **2**, 47-51

Singh VN, Prasad CR, Singh AK, Sinha RP (1989) Residue of gamma BHC in calcareous soil under different levels of nitrogen as urea. *Indian Journal of Entomology* **51**, 294-299

Suenaga H (1973) Provision on pesticide residue in Fukushima prefecture. *Shokubutu Boueki* **27**, 418-422

Suzuki M, Yamato Y, Koga M (1978) Field desorption mass spectrometric analysis of organic compound residues in the environment – organochlorine insecticides. *Biomedical Mass Spectrometry* **5**, 518-523

Takahashi K, Suzuki N, Magariya N (1978) On the results of determination of pesticides for agricultural use in irrigation water and soil in paddy rice field. *Annual Report of the Sendai Municipal Institute of Public Health* **8**, 232-234

Thapinta A, Hudak PF (2000) Pesticide use and residual occurrence in Thailand. *Environmental Monitoring and Assessment* **60**, 103-114

Tokyo Metropolitan Government (1965) Report of middle agricultural census in 1965, pp 17-67

Wegman RCC, Hofstee AWM (1982) Determination of organochlorines in river sediment by capillary gas chromatography – Application to Dutch river sediment. *Water Research* **16**, 1265-1272

Xu X, Yang H, Li Q, Yang B, Wang X, Lee FSC (2007) Residues of organochlorine pesticides in near shore waters of LaiZhou Bay and JiaoZhou Bay, Shandong Peninsula, China. *Chemosphere* **68**, 126-139

Xue N, Xu X (2005) Screening 31 endocrine-disrupting pesticides in water and surface sediment samples from Beijing Guanting reservoir *Chemosphere* **61**, 1594-1606

Yamamoto M, Nutahara M (1974) Sampling error in monitoring of dieldrin residue in farmland. *Bulletin of the Kochi Institute of Agriculture and Forestry Science* **6**, 57-58

Yoshioke A (1974) Diagnosis of environment seen from soil-pollution. The present state of soil pollution by heavy metals and pesticides. *Environmental Creation* **4**, 84-89

Zhou R, Zhu L, Yang K, Chen Y (2006) Distribution of organochloride pesticides in surface water and sediments from Qiantang River, East China. *Journal of Hazardous Materials* **137**, 68-75

Zimmerman LR, Thurman EM, Bastian KC (2000) Detection of persistent organic pollutants in the Mississippi Delta using semipermeable membrane devices. *The Science of the Total Environment* **248**, 169-179

JAPANESE ABSTRACT

ドリン剤として総称されるディルドリン、アルドリン、エンドリンはいずれも有機塩素系殺虫剤であり、土壌残留性および作物残留性が高いことから、残留性有機汚染物質（POPｓ）に指定されている。これらの殺虫剤は、日本においては、1954 年に最初の農薬登録がされ、1975 年に登録失効するまでの 22 年間、農業生産の場で使用されていた。登録失効以降、使用されることはなかったが、今日においても、東京都内で生産されたキュウリ果実から残留基準値を超えるディルドリンが検出される事例がある。また、2002 年に行われた東京都における土壌残留実態調査の結果では、東京都内の 814 農地のうち、85 農地の土壌に 10 ng/g—2600 ng/g 乾燥重量の濃度で残留ディルドリンが検出されている。この総説では、今日の日本におけるドリン剤の土壌における残留濃度を諸外国の最近の残留実態と比較し、さらに、日本における土壌汚染物質に対する法制度について概説する。