Residues of Organic Chlorinated Pesticides in Agricultural Soils of Beijing, China

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Abstract. Concentration of organic chlorinated pesticides (OCPs) were measured in topsoils of a selected farm (NK farm) in Beijing in 1993 and 2003. The results indicated that OCPs, mainly 1,2,3,4,5,6-hexachlorocyclohexane (HCH) and 1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane (DDT), degraded greatly in the 20 years after the prohibition of their use. DDT was the major contributors of pollution on the farm with 92.23% and 81.28% contributions of total OCP load in soils in 2003 and 1993 respectively. The levels of total DDT and HCH in the old orchard group, in which cultivation began in 1962, were significantly higher ($p < 0.05$) than those in the new orchard group in which cultivation began in 1991, and all data were higher than the level of barren land ($p < 0.05$). Studying the isomeric and parent substance metabolite ratios indicated there was more application and accumulation of DDT in the old orchard and that DDT in the new orchard had undergone a different degradation time period or perhaps had been applied more recently, but no new input of HCH was detected. Pollution potential was assessed on the basis of China Soil Environmental Quality Standard.

Introduction

Because of their low cost and high degree of effectiveness, Organic chlorinated pesticides (OCPs) have been used globally since the 1940s. Taking China as an example, until 1983 the use of OCPs, with 1,2,3,4,5,6-hexachlorocyclohexane (HCH) and 1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane (DDT) as representatives, has made up 50% of total pesticide use (Hua and Shan 1996). Most OCPs are persistent, bioaccumulated, semivolatile, and highly-toxic, which has led to growing concerns regarding the potential for contamination of the environment and associated effects on human health and wildlife (Hua and Shan 1996; Dou and Zhao 1998; Ma *et al.* 2001). In the 12 priority persistent organic pollutants listed in

the 2001 Stockholm Convention on Persistent Organic Pollutants, 9 were OCPs (i.e., aldrin, dieldrin, endrin, DDT, chlordane, heptachlor, mirex, toxaphene, and hexachlorobenzene).

In China, the use and production of aldrin, dieldrin, endrin, toxaphene, chlordane, and heptachlor have been either limited or are nonexistent. The abuse of DDT and HCH in agricultural application during the 1960s and 1970s was the main contribution to the high levels of their residues detected in the environment and their consequent adverse effects on humans and wildlife (Liu *et al.* 2001; Wong *et al.* 2002). In 1983, the use of DDT and HCH was formally banned in China, yet small amounts of DDT and HCH (γ -HCH, lindane) are still being produced as raw materials for other chemicals or for export.

The remnant levels of OCPs in water, sediments, soils, crops, vegetables, and fruits decreased greatly after the prohibition of these substances. According to a nationwide investigation conducted by the Ministry of Agriculture in 1988 and 1989, the HCH remnant levels in crops decreased by a factor of 10 compared with those in the early 1980s. The same decrease happened with DDT (Lin *et al.* 2000).

Although the remnant levels of OCPs in China have decreased greatly since the 1980s, they are still higher than those in developed countries (Zhong *et al.* 2000; Wong *et al.* 2002), and their influence has still not disappeared; very low concentrations still exist in the human body and the environment. What is the current status of OCP residues in China? After a survey of OCP residues in food was conducted by the Ministry of Health in 1992 (Zhang and Zhang 2001), there has been no national investigation.

In this article NK farm, in which DDT and HCH were used in large quantities during 1962 to 1982, was selected for a case study of OCP remnant levels in agricultural soils. The OCP residues in soils of the old orchard group, in which apple trees started being cultivated in 1962, and the new orchard group, in which cultivation began in 1991, in NK farm were measured in samples taken in 1993 and 2003, 10 years and 20 years, respectively, after their prohibition. By analyzing factors such as cultivation history, history of OCP use, and isomeric and parent substance metabolite ratios, current OCPs status and time trend were determined. Potential risk was assessed using environmental guidelines established in China.

Table 1. Group, sample description, and selected soil properties

Group	Barren land	New orchard	Old orchard	
			Orchard	New fallow
Sample no.	N12	N1, N15, N16, N17	N2, N3, N4, N6, N7, N8, N9, N10, N13	N5, N11, N14
Land use	Barren	Orchard	Orchard	Orchard, but changed to fallow recently
Cultivation history	No cultivation	Apple trees began to be cultivated in 1991	Apple trees began to be cultivated in 1962	Apple trees began to be cultivated in 1962 and were fallowed recently: N5 was fallowed in 2000, N11 in 1999, and N14 in 1997
Soil texture	Sandy	Sandy loam	Sandy loam	Sandy loam
Organic C (%) (in 2003)	1.104	1.734	1.798	1.794
Organic C (%) (in 1993)	0.73	1.625	2.53	2.22
pH (in 2003)	7.8	6.6	7.2	6.5
pH (in 1993)	8.1	7.5	7.6	7.4

Materials and Methods

Study Area and Soil Sampling

NK farm is located at E 116.094°, N 40.177° in a northwestern suburb of Beijing, China. The climate is typically cool monsoon weather with an average annual temperature of 11.6°C and an average annual rainfall of 584 mm.

NK farm is dedicated for the cultivation of apple trees. The total orchard area is approximately 140 ha including an area of new orchards cultivated in 1991 that makes up one fourth of the farm. The remaining three fourths of the farm is old orchard in which cultivation began in 1962, with a small part of that field lying fallow since 1997. DDT and HCH were used at NK farm in large quantities during 1962 to 1982; carbamate, pyrethroids, and organophosphates have been the primary pesticides in use since 1982. The group, sample description, and selected soil properties are listed in Table 1.

To investigate the contents of OCPs in the ploughed layer of the 17 orchard sites, grid pattern sampling was carried out in 1993 and 2003. Four subsamples from each of the sampling areas were collected at soil depth (0 to 20cm) with a soil corer. All subsamples were thereafter pooled to obtain one representative sample for each site. Samples were homogenized by sifting through a steel mesh (2-mm grid size). The samples were freeze dried at -20°C and then stored at 4°C until analyzed.

Standards, Materials, and Solvents

The organochlorines were quantified using *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, α -HCH, β -HCH, γ -HCH, δ -HCH, dieldrin, endrin, heptachlor, and chlordane. These standards at 100 mg/l were supplied by the National Research Center for Certified Reference Materials of China and then diluted to the desired concentration. All solvents obtained from the Beijing Chemical Factory were of analytical grade and were redistilled in an all-glass system before use. Florisil (Beijing Yizhong Chemical Plant) in 100 to 200 mesh was activated for 16 hours at 130°C; silica gel (Shanghai Changquan Chemical Plant) in 80 to 120 mesh was activated for 4 hours at 110°C; and anhydrous Na₂SO₄ (Beijing Chemical Factory) was dried at 300°C for 4 hours. All of the materials were analytical grade, and the activated ones were kept in a desiccator before use.

Sample Preparation and Cleanup

Samples, 10 g, of freeze-dried soil were mixed with 25 ml hexane-acetone (1:1) and then shaken in an ultrasonic bath for 20 minutes and centrifuged. The extracts obtained were concentrated for subsequent cleanup, transferred to a Florisil column (a glass column [30 cm * 1.5 i.d.] packed with 9 g Florisil, 1 g silica gel, and 2 g anhydrous Na₂SO₄ and pre-eluted with 30 ml hexane) and eluted with a 50-ml eluting mixture (15% acetone and hexane). The eluates were then concentrated and made into a final volume of 1 ml with hexane for gas-chromatographic analysis.

Analysis

The levels of HCH and DDT were measured using a Hewlett-packard 6890 gas chromatograph equipped with an Ni⁶³ electron-capture detector. An HP Ultra-1 capillary column (50 m * 0.32 mm i.d. * 0.17 μ m film thickness) was used. The carrier gas was helium at a flow rate of 1 ml/min. Nitrogen at a flow rate of 0.6 ml/min was used as the make-up gas for the electron-capture detector. A 1- μ l sample of extract was injected in the splitless mode. The temperature program was as follows: the initial temperature was held at 150°C for 2 minutes, then increased at 5°C/min to 200°C, then maintained for 2 minutes, then increased at 8°C/min to 270°C and held for 5 minutes.

A procedural reagent blank was included with each batch of sample, and none of the target compounds were detected in the procedural blanks. OCP residues were quantitatively determined by comparing the area under each peak with the area under the standard peak. The correlation coefficients of OCP calibration curves were all >0.998. Matrix spike recovery experiments were also undertaken. The recoveries of α -, β -, γ -, and δ -HCH with spiked samples were 92.6%, 93.5%, 94.7%, and 97.6%, respectively, and those *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD were 95.8%, 99.2%, 96.4%, and 95.4%, respectively. The limits of detection of OCPs were described as three of signal-to-noise ratio. The detection limits of α -, β -, γ - and δ -HCH were 0.16, 0.059, 0.099, 0.15 ng/g d.w., respectively, and those of *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, and *p,p'*-DDD were 0.37, 0.056, 0.094, and 0.096 ng/g d.w., respectively. The relative standard deviation varied from 2.2% to 6.0%.

Table 2. Descriptive statistics of total HCH and DDT in NK farm soils

Statistics	$\sum$ HCH		$\sum$ DDT	
	In 2003	In 1993	In 2003	In 1993
No. of samples	17	17	17	17
Geometric mean (mg/kg d.w.)	0.0320	0.3270	0.3813	1.4197
95% confidence interval	0.0127–0.0805	0.1123–0.8783	0.1514–0.9594	0.5737–3.5135
Geometric SD	6.0183	7.9930	6.0302	5.8262
Minimum(mg/kg d.w.)	0.0020	0.012	0.0072	0.044
Maximum(mg/kg d.w.)	0.7603	9.486	2.9100	24.615

DDT = 1,1,1-trichloro-2,2-bis (p-chlorophenyl) ethane.

HCH = 1,2,3,4,5,6-hexachlorocyclohexane.

Statistics

OCP data and their logarithmic transformations were subjected to Shapiro-Wilk test to define their distribution. Because of the normal distribution of the logarithmic transformation data, the values presented are geometric means together with their 95% confidence intervals (lower and upper bound). Homogeneity of variance was tested using Levene's test. A paired-sample Student *t*-test was used to examine the difference between OCP levels in 1993 and 2003. Analysis of variance (ANOVA) was used to compare the difference among the barren land, the old orchard group, and the new orchard group. After ANOVA, analysis was conducted, Student independent samples *t* test was used to compare the difference between old and new orchard groups and between the orchard and the new fallow subgroups. All statistical analysis was conducted using SPSS software.

Results and Discussion

OCP Levels in the Studied Area

Many of the chlorinated pesticides commonly detected in other regions of the world—such as dieldrin, endrin, toxaphene, and heptachlor—were undetectable in our samples. The major chlorinated pesticides detected in these samples were HCH and DDT families. Table 2 lists the descriptive statistics of total HCH and DDT levels in NK farm soils detected in 1993 and 2003. Total soil HCH content in 2003 ranged from 0.0020 to 0.7603 mg/kg d.w. with a geometric mean of 0.0320 mg/kg d.w. and a low range of 95% confidence interval in the geometric mean (0.0127 to 0.0805). Total soil DDT content in 2003 ranged between 0.0072 and 2.9100 mg/kg d.w. with a geometric mean of 0.3813 mg/kg d.w. and a larger range of 95% confidence interval for geometric mean (0.1514 to 0.9594). Total HCH and DDT presented at higher levels in 1993.

DDT was a major concern of OCP pollution on the farm with 92.23% and 81.28% contribution to total pesticide concentration in 2003 and 1993, respectively. DDT is resistant to biodegradation and is absorbed strongly by soil particles (Keith 1997). The relatively high levels of DDT indicated that it was highly persistent in the top layers of the study area.

Current and historic HCH and DDT levels in NK farm soils were generally comparable with literature data from other foreign and domestic orchards in the corresponding survey year (Table 3). For example, HCH and DDT residues in NK farm surface soils in 2003 were on the same order with con-

centrations reported from orchard soils of Zhejiang Province in China, and DDT levels were lower than those in orchard soils of New Zealand in 2002. NK farm also showed contaminant levels of DDT in 1993 similar to those in Canadian orchard soils in 1994.

Orchard fields are dynamic systems of particular environmental concern because they exhibit a high capacity to concentrate and deliver pollutants. This study proved that orchard soils have much higher levels of HCH and DDT, than reported in from other agricultural soils (Table 3). Compared with rice fields in Jiaying City, NK farm in 2003 had rather large deviations of HCH and DDT remnant levels and much higher maximum HCH and DDT levels. Current HCH and DDT residue levels were even 1 to 2 orders of magnitude higher than those in agricultural soils of the Taihu region of China and cotton fields in Georgia, which were surveyed recently. NK farm in 1993 showed contaminant levels of DDT and HCH some 10-fold higher than those in cotton and rice fields in Jiangsu in 1992 and 1 to 2 orders higher than those detected in German agricultural soils during 1995 and 1996.

Comparison Between Present and Historic OCP Data

Statistics showed there was a significant difference between total HCH level in 1993 and the level in 2003 (paired-samples Student *t* test, $t = -6.175$, $p < 0.05$). For each of the soil samples, the total HCH concentration had decreased greatly in 2003 compare with 1993 (Fig. 1a). The geometric mean level in mg/kg of total HCH dry weight (0.0320) in 2003 was considerably lower than that in 1993 (0.3270).

The 2003 results showed that most soils, with the exception of N11, had lower contents compared with 1993 data (Fig. 1b). Total DDT concentration in 2003 was significantly lower than that in 1993 (paired-samples Student *t* test, $t = -4.917$, $p < 0.05$). The geometric mean level of DDT (0.3813 mg/kg.d.w.) in 2003 was considerably lower than that in 1993 (1.4197 mg/kg.d.w.).

As in the whole of China, the use of HCH and DDT has been banned at NK farm since 1982. Therefore, the farm has been undergoing degradation, and there should have been no new input during the next 2 decades. This may explain the overall decrease in all of the selected OCPs detected in 1993 and 2003.

Comparisons between the 1993 and 2003 data revealed a higher decrease in HCH (90.21%) than DDT (73.14%) during

Table 3. Comparison of OCP levels (mg/kg d.w.) in surface soils from other geographic areas (domestic and abroad)

Area	Year	Land use	HCH		DDT		References
			Range	Mean	Range	Mean	
Beijing, China	2003	Orchard	0.0020–0.7603	0.032	0.00721–2.9100	0.3813	This work
Zhejiang, China	1999	Orchard	0.0011–0.0763	0.015	0.0015–5.6425	0.7282	Zhao and Ma 2001
Auckland, New Zealand	2003	Orchard			<0.03–24.41	Median 1.17	Gaw <i>et al.</i> 2003
Jiaxing, China	2001	Rice field	0.0186–0.0554	0.0347	0.0286–0.0718	0.0489	Zhao <i>et al.</i> 2002
Taihu, China	2001	Agricultural	0–0.0034		0–0.0053		Feng <i>et al.</i> 2003
Georgia, USA	1999	Cotton field	<0.0001–0.00049		0.00034–0.034	0.013	Kannan 2003
Beijing, China	1993	Orchard	0.012–9.486	0.327	0.044–24.615	1.419	This work
Simcoe, Canada	1994	Orchard				7.1	Harris <i>et al.</i> 2000
Jiangsu, China	1992	Cotton field		0.032		1.230	Lin <i>et al.</i> 2000
Jiangsu, China	1992	Rice field		0.045		0.053	Lin <i>et al.</i> 2000
Germany	1995 to 1996	Agricultural	0.0046–0.0115		0.0279–0.173		Manz <i>et al.</i> 2001

DDT = 1,1,1-trichloro-2,2-bis (p-chlorophenyl) ethane.

HCH = 1,2,3,4,5,6-hexachlorocyclohexane.

OCP = Organic chlorinated pesticide.

the 10 years. This may be related to the differences in their chemical structures, which was illustrated by specific parameters.

HCH has much higher water solubility, higher vapor pressure, lower octanol-water coefficient, and lower Henry's Law constant than DDT (*e.g.*, the water solubility of HCH isomers ranges between 0.24 and 31.4 mg/l, two or three orders of magnitude higher than that of the DDT family, which ranges between 0.0055 and 0.1 mg/l [Mackay *et al.* 1997]). In this regard, HCH presents an easier ability to degrade and transfer from soil to liquid and atmosphere and thus less persistent in the environment than DDT.

Comparison Between OCP Levels in the Old Orchard and New Orchard Groups

Statistics showed there is significant difference among OCPs in the soils of the old orchard group, the new orchard group and the barren land (ANOVA $F = 4.928$, $p < 0.05$ for $\sum$ HCH in 2003; $F = 20.257$, $p < 0.05$ for $\sum$ HCH in 1993; $F = 21.522$, $p < 0.05$ for $\sum$ DDT in 2003; and $F = 14.012$, $p < 0.05$ for $\sum$ DDT in 1993). The OCP levels in barren land (N12) were lower than all other data, and the levels of total DDT and HCH in the old orchard group were significantly higher than those in the new orchard group (Table 4).

DDT and HCH were extensively used in the old orchard group during the period 1962 to 1982, whereas few OCPs were applied in the new orchard group since it began cultivation in 1991, 9 years after the prohibition of the two pesticides; therefore, there was significant difference in OCP application between the two orchard groups. The intensity of OCP degradation depends on several environmental factors such as soil type, pH, redox potential, organic carbon content, and microbial biomass (Hitch and Day 1992) as well as anthropogenic activities (*e.g.*, land use, fertilization, and pesticide application). Because all other factors remain similar between the two groups, the different OCP residue levels may be related to the differences in their application.

In the old orchard group, no significant difference between the orchard subgroup and the new fallow subgroup (Table 5) was noted (independent-samples Student *t* test, $t = 1.817$, $p > 0.05$ for $\sum$ HCH in 2003; $t = 1.363$, $p > 0.05$ for $\sum$ HCH in 1993; $t = 1.150$, $p > 0.05$ for $\sum$ DDT in 2003; and $t = 1.824$, $p > 0.05$ for $\sum$ DDT in 1993). This is reasonable because there should have been no new input of DDT and HCH Since 1997, the 15th year after prohibition and when the soils started to become fallow (see Table 1 for the cultivation history of the soils). In theory, the two subgroups were exposed to the same OCP load and degradation period.

Previous and Current OCP Input

Studying the isomeric and parent substance metabolite ratios enables past and present pollutant emission sources to be distinguished. The absolute measurement of individual isomers within the DDT and HCH in 2003 (Fig. 2) showed differences in the concentrations between the groups.

Technical HCH is the mixture of 55% to 80% α -HCH, 5% to 14% β -HCH, 12% to 14% γ -HCH (lindane), 2% to 10% δ -HCH, and 3% to 5% ϵ -HCH. Lindane has the most potent insecticidal properties. Lindane can be completely decomposed or biotransformed into other HCH isomers in agricultural soils (Liu *et al.* 2001) and in upper soils has an average half-life of approximately 2 months (Turnbull 1997), although its exact half-life depends closely on the soil type. If the γ -HCH isomer predominates in relative terms, new inputs must be present in the soil. In all the soil samples from NK farm, the highest concentration of HCH isomers was measured for β -HCH (Fig. 2a), indicating no new input of HCH on the farm.

The ratio of α -HCH to γ -HCH ranged from 1.000 to 2.636 with a mean of 1.753 in the old orchard group and from 0.520 to 1.000 with a mean of 0.807 in the new orchard group. The ratio of α -HCH to β -HCH ranged between 0.003 and 0.223 with a mean of 0.040 in the old orchard group and from 0.034 to 0.177 with a mean of 0.093 in the new orchard group. Compared with the new orchard group, the higher ratio of α -HCH to γ -HCH and lower ratio of α -HCH to β -HCH in the old orchard group indicated the higher percentage of β -HCH

a) HCHs

b) DDTs

Fig. 1. Concentration of total (a) HCH and (b) DDT in 2003 and 1993. DDT = 1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane; HCH = 1, 2, 3, 4, 5, 6-hexachlorocyclohexane.

and can be explained by the isomerization of α - to β -HCH and of γ - via α - to the more stable β -HCH, which is energetically more favorable (Wu *et al.* 1997) and thus the higher HCH residue level in soils.

In the DDT group, the highest levels recorded were between approximately 0.0012 and 1.4480 mg/kg d.w. for *p,p'*-DDE; between 0.0009 and 0.245 mg/kg d.w. for *p,p'*-DDD; between 0.0013 and 0.4030 mg/kg d.w. for *o,p'*-DDT, and between

Table 4. Comparison between concentrations of OCP levels in the old orchard and new orchard groups (mg/kg d.w.)

Survey year	Pollutants	Barren land	Old orchard			New orchard		
			Geometric mean	95% Confidence interval	Geometric SD	Geometric mean	95% Confidence interval	Geometric SD
2003	$\sum$ HCH	0.002	0.0642	0.0234–0.1764	4.9084	0.0079	0.0019–0.0329	0.8955
1993	$\sum$ HCH	0.020	1.006	0.4689–2.1590	3.3259	0.0226	0.0068–0.0746	0.7512
2003	$\sum$ DDT	0.013	0.9936	0.6338–1.5576	2.0291	0.0509	0.0043–0.5932	1.5459
1993	$\sum$ DDT	0.044	3.381	1.6777–6.8135	3.0128	0.2505	0.0487–1.2881	1.0290

DDT = 1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane.

HCH = 1,2,3,4,5,6-hexachlorocyclohexane.

OCP = Organic chlorinated pesticide.

Table 5. Comparison between OCP concentrations in the orchard and new fallow subgroups (mg/kg d.w.)

Survey year	Pollutants	Orchard		New fallow	
		Geometric mean	95% Confidence interval	Geometric mean	95% Confidence interval
2003	$\sum$ HCH	0.0856	0.0229–0.3187	0.0271	0.0032–0.2277
1993	$\sum$ HCH	1.0942	0.4228–2.8319	0.7822	0.0308–19.842
2003	$\sum$ DDT	0.9498	0.5261–1.7147	1.1373	0.2588–4.9977
1993	$\sum$ DDT	3.9689	1.9209–8.2009	2.0900	0.0363–12.0267

DDT = 1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane.

HCH = 1,2,3,4,5,6-hexachlorocyclohexane.

OCP = Organic chlorinated Pesticide.

Table 6. Probability that the sampling group exceeded the guide value of the specific grade according to the China Soil Environmental Standard (GB15618-95)

Year	Pollutants	Old orchard	New orchard	Total farm
2003	$\sum$ HCH	I >50%, II <16%, III <2.5%	I <2.5%	I <50%, II <16%
1993	$\sum$ HCH	I >97.5%	I <2.5%	I >50%, II <50%, III <50%
2003	$\sum$ DDT	I >97.5%, II > 50%, III >50%	I >50%, II <2.5%	I >84%, II <50%, III <50%
1993	$\sum$ DDT	III >84%	I >97.5%, II <2.5%	I >84%, II >50%, III >50%

Grade I: HCHs $\leq$ 0.05 mg/kg d.w. and DDT = 0.05 mg/kg d.w.; grade II: HCHs $\leq$ 0.5mg/kg d.w. and DDT = 0.5 mg/kg d.w.; Grade III:

HCH $\leq$ 1.0 mg/kg d.w. and DDT = 1.0 mg/kg d.w.

DDT = 1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane.

HCH = 1,2,3,4,5,6-hexachlorocyclohexane.

0.0024 and 0.814 mg/kg d.w. for *p,p'*-DDT. DDE was the dominant isomer in the soils (Fig. 2b).

The ratio of *p,p'*-DDE to *p,p'*-DDT which is an indication of the historic accumulation of *p,p'*-DDT, was approximately 1.927 in the old orchard. This was higher than the 1.266 obtained in the new orchard, thus indicating the greater accumulation of DDT in the old orchard group.

The DDT-to-(DDD+DDE) ratio can be used to evaluate the degree of DDT degradation (Tavares *et al.* 1999). The high value of this ratio indicates slow degradation of DDT or addition of unaltered DDT contamination to the samples. The higher ratio of DDT to (DDD+DDE) (mean of 0.890) in the new orchard soils than the old orchard soils (mean of 0.751) indicated that DDT had undergone a different degradation period or suggested a more recent application of *p,p'*-DDT in the new orchard. The large amount of DDT residue disposed of in the past might be an explanation. It also may be related to the production and use of dicofol around China, in which 3.54% to 10.8% of DDT exists as major impurity (Yu *et al.*

2001). Until now, the continuing input of DDT caused by dicofol should be an important aspect for the analysis of DDT residues and elimination strategy in China.

Pollutant Potential Evaluation of the Soil Findings

An OCP pollutant potential was assessed on the basis of the China Soil Environmental Standard. The assessment was made by estimating the Probability to Exceed the Guide value. The probability is estimated by the distribution of the ratio of the estimation of the sampling group to the guide value (Shen 1995). The ratio is computed as such:

$$R = \frac{M_g \times S_g^{(k)}}{G} \quad (1)$$

where, R is the ratio of the estimation of the sampling group to the guide value; M_g is the geometric mean; S_g is

a)HCHs

b)DDTs

Fig. 2. Concentration of OCP isomers in 2003. OCP = organic chlorinated pesticides.

the geometric SD; K is an integer ($-2 \leq K \leq 2$); and G is the guide value to be compared.

The probability that the sampling group exceeded the guide value of the specific grade of China Soil Environmental Standard (Table 6). If the pollution level was less than grade I (background value), it was defined as no pollution; if the pollution level was between grade I and grade II, it was defined as low pollution; if the pollution level was between grade

II and III, it was defined as moderate pollution; and if the pollution level exceeded grade III, it was defined as high pollution.

For the new orchard group, HCH pollution in soils from the 1993 and 2003 samples was categorized as no pollution; in 1993, DDT pollution exceeded grade I but was less than grade II and was categorized as low pollution; in 2003, the pollution was lower than in the 10 years before but still in low-pollution

range. At present, the new orchard can be regarded as having slight HCH pollution and low DDT Pollution.

In the old orchard group, the HCH and DDT pollution in soils in 2003 was decreased from that in 1993. The HCH pollution levels in both of the sampling years are at a moderate level, whereas the DDT pollution level decreased from a high pollution level in 1993 to a moderate pollution level in 2003. The total farm can be regarded as having had low HCH pollution in both 1993 and 2003 and as having had high DDT pollution in 1993 but a low DDT level in 2003.

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