Residues of Organochlorine Pesticides in Vegetables from Deyang and Yanting Areas of the Chengdu Economic Region, Sichuan Province, China

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Abstract

Residual levels of organochlorine pesticides (OCPs) were determined in 34 samples of 19 varieties of vegetables collected from selected sites around Deyang city and Yanting County, Southwest China, in June and September 2007. The analytical method included soxhlet extraction with a mixture of dichloromethane and acetone (2:1 vv). Clean-up was done on superposed layers of alumina/silica gel (1:2 v/v) impregnated with concentrated sulfuric acid. The determinations were done using a gas chromatograph with electron capture detector (GC-ECD). The compounds targeted are: isomers of hexachlorocyclohexane or HCHs (α-HCH, β-HCH, γ- HCH or lindane and δ-HCH); isomers/metabolites of dichloro-diphenyl-trichloroethane (DDTs) namely p,p^1-dichloro-diphenyl-dichloroethylene (p,p^1-DDE), p,p^1-dichloro-diphenyl-dichloroethane (p,p^1-DDD), p,p^1- and o,p^1-dichloro-diphenyl-trichloroethane (p,p^1-DDT and o,p^1-DDT). The results indicated that all the vegetable samples had some levels of one or more OCPs in them. Residues of DDTs were found in 94.12% while HCHs were in 91.18% of all the samples analyzed indicating high incidence of these xenobiotics in the vegetables from the areas investigated. Among the HCH isomers, γ- HCH was the most prevalent but β-HCH was the most abundant indicating both old and fresh inputs of HCHs. DDT metabolites p,p-DDE and p,p'-DDD were more prevalent than the parent material, p,p-DDT suggesting minimal fresh inputs of DDT. The OCPs residue levels in the vegetables were generally low (≤ 1.3 ng/ g wet weight) except in one sample of green pepper (*Capsicum annum L*) in which the concentrations (ng/g wet weight) of o,p'-DDT (82.59), p,p'-DDE (61.41) and total DDT (148.44), all exceeded the Chinese Extraneous Maximum Residue Limit of 50 ng/ g for DDTs in vegetables according to the guidelines of the Chinese quality standard for food (GB 2763-2005). Considering the industrial and agricultural growth around the areas investigated, a deeper investigation and regular ecological and foodstuff monitoring is recommended.
[Journal of American Science 2009; 5(4):91-100]. (ISSN: 1545-1003).

Key words: Vegetables, Organochlorine pesticides, Deyang, Yanting

1.0 Introduction

Oraganochlorine pesticides (OCPs) are among the xenobiotics that have become constituents of the biosphere due to their great use all over the world, stability in several natural conditions and mobility in the environment. In china, large quantities of OCPs, particularly hexachlorocyclohexane (HCH) and dichlorodiphenyltrichloroethane (DDT) were produced and used in agriculture and public health until 1983. Therefore, the occurrence of excessive OCPs residues in Chinese environments is believed to be serious and widespread and has over the years sustained considerable research interests in elucidating environmental contamination status and human exposure in the country. Sichuan province is one of the leading agricultural regions in China and it is an important Southern China vegetable production base (Chen et al, undated). To keep the pest effects at economical levels, obviously, Sichuan province has been a major consumer of pesticides including OCPs. It was estimated that between 10,000 and 20,000 metric tons (MT) of technical HCH and 16 000 MT of DDT were used in Sichuan during the period 1951-1984 and these were among the highest usages in China (Li et al, 1998; Liu et al, 2006). The extent of usage suggests that environmental contamination could also be widespread; however, the magnitude and distribution of this is only just beginning to be accurately characterized.

A survey recently undertaken in the Chengdu Economical Region (CER) which is the main agricultural and industrial region in Sichuan, found out that OCPs were still detectable in all surface soils despite having been forbidden more than 20 years in China (Xing et al, 2009). The study also revealed that the DDT residual contents of agricultural soils in some areas including Deyang and Yanting, were in excess of 50 ng/g, which is maximum permissible level in agricultural soils in China, according to the Chinese environmental quality standard for soils (GB15618-1995). This called for explanations about the possible occurrence of OCPs in food crops grown in these soils since several crops are known to accumulate OCP in edible parts up to critical levels, which may contribute to dietary intake of the contaminants. In addition, a national dietary intake survey of 1992 showed that Sichuan was one of the regions in China with high dietary intakes of DDT (Chen and Gao, 1993). Already, Li et al (2006) have reported a high prevalence of β-HCH (91.2%), p,p'-DDE (92.1%) and p,p'-DDT (91.2%) as well as positive correlation between high levels of these compounds in the subjects' sera on one hand and breast cancer on the other, particularly among premenopausal women in the province. Therefore, information on OCPs residue levels is of paramount importance to the consumers of agricultural produce from CER, Sichuan province. This paper presents the results of a preliminary survey carried out on the residue levels of orgnochlorine pesticides (DDTs and HCHs) in vegetables produced in Deyang and Yanting areas of the agricultural and industrial CER. Vegetables are important components of the Chinese diet both in terms of quantities consumed and nutritional value. Also, vegetables are consumed directly without much processing or storage. Therefore, even low residual levels of toxic contaminants in vegetables spell high danger to the consumers. These make information on residue levels of xenebiotics (such as DDT and HCHs) in vegetables very important for the protection of human health. Vegetables were also selected for this survey because, at the time of the survey, they were the most widely distributed food plants in the study area.

2.0 Materials and Methods

2.1 Description of the Study Areas

The sampling spots are located within two areas, Deyang and Yanting in the CER, Sichuan Province. The names of the areas were for the purposes of this report and are not restricted to administrative boundaries with same names.

Deyang sampling area is located at the central part of the CER, western edge of Sichuan Basin between the latitudes: 30^0 57' 56 N and 31^0 21' 00" N and between longitudes 104^0 05' 40" E and 104^0 15' 32"E. It includes selected sites in Pengzhou County of Chengdu City prefecture; Shifang, Mianzhu and Guanghan counties in Deyang City. This is typically a hilly area that falls within a subtropical humid climate zone with a continental monsoon climate. The average annual temperature is about 15.7^0C (which means no extreme temperatures in summer and winter) while annual precipitation average here is about 1053mm. The soil types are varied but the main ones are paddy soil, purple soil, and yellow earth. The soils are acidic to neutral (pH: 5.5-7.5) with organic matter content 0.6-4.0%. (Xing et al, 2009). Arable land forest and non-arable land evenly share the total land area. The Deyang area is an important vegetable production area in Sichuan. The main vegetables grown are: Pepper, celery, eggplant, tomato, lettuce, four season beans, cowpea, Chinese leaf, cucumber, bitter gourd, rape shoots (Chen et al., undated).

Yanting area in Yanting County of the Mianyang City prefecture falls within the Northeastern part of the CER, in central Sichuan Basin or northern part of Sichuan Province between the latitudes 31^0 13' 04" N and 31^0 19' 58"N and longitudes 105^0 19' 53"E and 105^0 57' 56"E. The annual average temperature in Yanting is 17-18^0C and precipitation 800 -1000 mm, with most of the precipitation occurring during the rainy season from May through August. The soil is classified as purple soil- a lithologic soil (Regosols in FAO Taxonomy) (CERN, 2003). The soil organic matter contents are low (0.6-2.0%) but pH high (8.0-8.6) (Xing et al., 2009). Steeper hills are forested, flat or gentle slopes are used for crops and lowlands used for irrigated paddy. The main crops are rice, wheat, corn, rape and sweet potato.

2.2 Site Selection and Sampling

The sampling sites were selected following the results of our soil OCPs survey (Xing et al, 2009) that covered the entire Chengdu economic region. Crop samples were taken from the farmlands whose soils had been found with relatively elevated levels of OCPs (≥ 10 ng g^{-1} dry weights). A global positioning system (GPS) instrument was used to locate sampling locations. Vegetables found growing on the sites or within the immediate vicinity were eligible for sampling. More samples were collected from the Deyang area (N=24) than from the Yanting area (N=10), since during the soil survey, a larger area in Deyang than in Yanting had been found to contain relatively elevated soil OCP residue levels (≥ 10 ng g^{-1} dry weight). Vegetables were collected directly from the fields to ensure that they originated within the study areas. The samples were collected on June 11-12 (spring vegetables) and September 22-26 (autumn vegetables) in the year 2007. Each sample was sealed in a clean polythene sampling bag, and immediately transported to the laboratory where they were kept refrigerated at 4^0C until analysis.

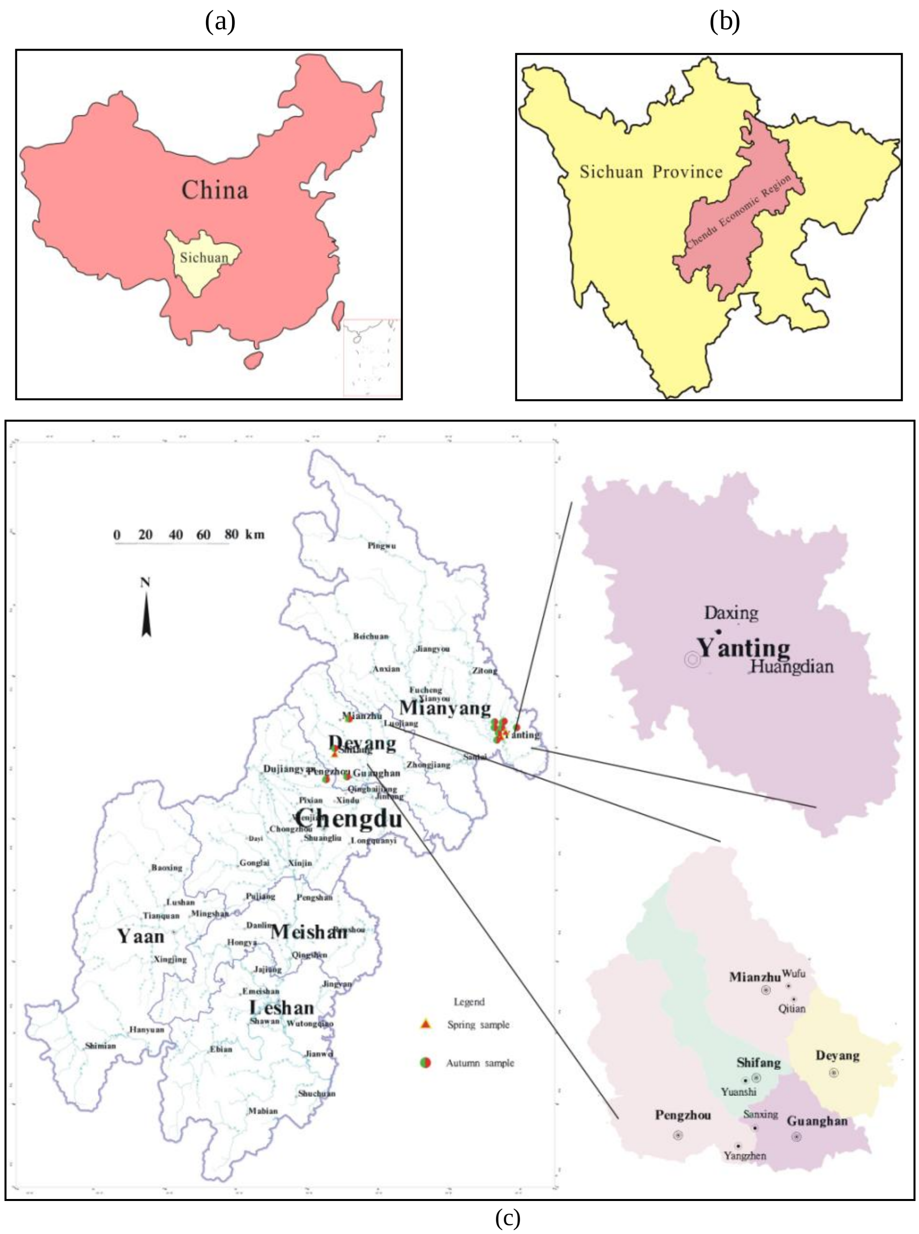

Figure 1. Sampling locations in Deyang and Yanting areas of the Chengdu Economic Region, Sichuan Province, China; (a) Location of Sichuan Province in china (b) Location of Chengdu Economic Region in Sichuan and (c) the location of sampling spots

2.3 Experimental Procedures

2.3.1 Extraction and Clean Up

About 50g of thawed edible portion of each vegetable sample was chopped (where possible) and blended for 3-5 minutes. Ten grams (10 g) of the homogenate was carefully weighed and transferred into a well tucked filter paper containing about 20 g of anhydrous sodium sulfate. The sealed mixture was spiked with 20ng of 2,4,5,6- tetrachloro-m-xylene (TCMX) and decachlorobiphenyl (PCB 209) as recovery surrogates. The mixture was then soxhlet extracted for 48 hours with 120 mL of dichloromethane and acetone mixture (2:1 vv). The extract was then concentrated to about 2 mL using a rotary evaporator at 40^0C, solvent-exchanged to n-hexane and transferred into a separatory funnel where about 3 mL of concentrated sulfuric acid (98%) was added with shaking (to remove lipids). The mixture was allowed to stand until two distinct layers were observed. The lower turbid layer (lipids in acid) was then removed and discarded. The procedure was repeated three times or until the lower acid layer appeared clear. The remaining organic layer (containing OCP extracts) was passed through alumina/silica gel (1:2 v/v) clean-up column (8 mm i.d.). The sample extract was eluted three times with 30mL dichloromethane/hexane (2:3) mixture to yield the OCPs fraction. The eluate was concentrated to about 0.5 mL before being transferred (using n-hexane) into a 2 ml screw cap cell bottle (Agilent Technologies USA) where it was concentrated further to 0.2ml under gentle stream of purified nitrogen gas at 40-45^0C. The extracts were stored at -20^0C, prior to GC-ECD analyses. Pentachloronitrobenzene (PCNB) was added as an internal standard before the analyses.

2.3.2 Instrumental Analysis

Concentrations of organochlorine pesticide residues were determined with HP 6890 gas Chromatograph (GC) system equipped with a micro-cell ^{63}Ni electron capture detector (ECD). The sample volume injected was 2 µl. The separation was performed on a fused silica capillary column (HP-5, 30 m x 0.32 mm i.d. and film thickness of 0.25µm). The carrier gas was nitrogen at a flow rate of 2.5 mL/min in a constant flow mode. Injector and the electron capture detector temperatures were set at 290^0C and 300 ^{0}C respectively. The oven temperature was programmed as follows: Initial temperature 100^0C held for 4 minutes; increased to 200^0C at a rate of 4^0C/min then increased to 230^0C at a rate of 2 ^{0}C/min followed by 8 ^{0}C/min till the final temperature of 280^0C at which it was held for 15 minutes. A six points response factor calibration was established to quantify the target analytes. System control was by HP-3365 Chem-station software.

2.3.3 Quality Assurance and Control

All analytical procedures were monitored using strict quality assurance and control measures. Chemicals used in the sample preparation and analyses were of high grade analytical standards obtained from Tedia Company, USA and J.T Baker, USA. Before use, neutral alumina, neutral silica gel and anhydrous sodium sulfate were soxhlet-extracted for 48 hours with dichloromethane and then baked for 12 hours in 250 ^{0}C, 180 ^{0}C and 450^0C respectively. Laboratory glassware were washed with concentrated sulfuric acid/potassium dichromate mixture, rinsed with distilled water and n-hexane then dried in the oven at 110^0C overnight prior to use. With each set of samples analyzed, field and procedural blanks were also analyzed to check for cross contamination and interferences. None of the target compounds were detected in the blank samples. The method of detection limits (MDLs) of OCPs ranged from 0.005-0.01 ng/g. The recovery rates of the surrogates; TMCX and PCB209 were 65±4% and 74±5% respectively. Each sample was analysed in duplicate and the average pesticides residue levels in the duplicates recorded as the residue level in the candidate sample. The relative standard deviation (RSD) for duplicate samples averaged 4%. Results of the analysis are reported in ng/g on wet-weight (wet wt) basis. A reporting limit of >0.01 ng/g wet wt was taken for calculation purposes since some duplicate averages were below the MDLs. Levels below the reporting limit or below MDL were taken as zero (0) in the calculations.

3. Results and Discussion

3.1 Occurrence and Levels of OCPs in Vegetables

A total of 34 samples of different species of vegetables (leafy, fruiting, root/bulbs and beans) were analysed for OCPs. The monitored OCPs are α-HCH, β-HCH, γ-HCH and δ-HCH (HCHs); p,p'-DDE , p,p'-DDD, o,p'-DDT and p,p'-DDT (DDTs). Concentration and detection rates of detected OCPs residues are given in Table 1. Quantifiable residues of DDTs were found in virtually all the vegetable samples from Deyang and in all but two samples from Yanting, indicating a widespread contamination of vegetables by OCPs in the study areas. The DDT metabolites (p,p'-DDE and p,p'-DDD) were in general more prevalent than the parent materials (o,p'-DDT and p,p'-DDT) suggesting either efficient biotransformation of the parent materials in the plant systems or old sources of

DDT contamination. Among the HCH isomers, lindane was the most frequently quantified both in the Deyang and Yanting vegetable samples (Table 1).

Without the single sample of green pepper (*Capsicum annuum* L.), the average residue level of total HCH (∑HCH) was slightly higher than that of total DDT (∑DDTs) in both Deyang (0.25 vs 0.24 ng/g) and Yanting (0.17 vs 0.11 ng/g). The pepper was not included in the calculations because DDT residue levels in it (totaling 148.44 ng/g wet wt.) appeared anomalous and since there was no other pepper sample to compare, the results were considered as outlier. Previous surveys had also reported ∑HCHs dominance over ∑DDTs but with higher concentrations than found in the present study, which indicates the use limitations of the chemicals are effective. For example, a national dietary survey undertaken in 1992 found average residue levels of ∑HCHs and ∑DDTs in vegetables to be 7.1 and 1.00 ng/g respectively (Liu et al, 1995) while in Shanghai (in the year 2000) the average concentrations were 4.9 and 2.9 respectively (Nakata et al, 2002). The slightly higher concentrations of ∑HCHs than ∑DDTs is probably because more technical HCH (approximately 4.9 million tons) than technical DDTs (0.4 million tons) were produced and used in China before the agricultural uses of these insecticides were banned in 1983 (Hua and Shan, 1996). Moreover, lindane replaced technical HCH in 1991 and about 3200 tons were used between 1991 and 2000 (Li et al, 2001). It should however, be noted that in a soil survey of the present study areas, Xing et al (2009) reported much higher mean residue levels of DDT (31.00 ng/g) than HCH (1.96 ng/g) in the agricultural soils. The discrepancy between the residue levels of DDT and HCHs in soil and their accumulative levels in the edible portions of vegetables could be because DDT is more hydrophobic than HCH. Hydrophobic compounds are strongly bound to root and soil organic colloid surfaces resulting in less absorption and/or translocation (Pereira et al, 2006).

Average HCH residue levels in the vegetables from Deyang were in the range, 0.06 -0.56 ng/g wet wt with bean (*Phaseolus vulgaris*) as the least contaminated vegetables and radish (*Raphanus sativus*) as the most, respectively. The range in Yanting was from <0.01 ng/g wet wt in spinach (*Spinacia oleracea*) to 0.30 ng/g wet wt in Pumpkin (*Cucurbita spp*). As for the ∑DDTs the average residue concentrations ranged from 0.03 ng/g to 0.64 ng/g in Deyang and from <0.01 to 148.44 ng/g in Yanting. Bean (*Phaseolus vulgari*) was again the least contaminated crop in Deyang where potato (*Solanum tuberosum*) had the highest mean ∑DDTs value. In addition to the pepper, residue levels of DDT in Deyang vegetables were notable for cabbages (*Brassica spp.*; 0.39 ng/g) and radish (*Raphanus sativus*; 0.33). However, apart from the pepper sample, vegetables in this study had residue levels far much below the recommended extraneous maximum residue limits (EMRLs) of 50 ng/ g wet wt (for lindane, ∑HCH and DDT), as set forth by the Chinese ministry of public health (Chinese food standard, GB 2763-2005), indicating minimal risk to the consumers.

Our results are not easy to compare with those of many other studies, because the differences in sampled species, analytical method employed and to some extent, differences in expression of analytical results (wet weight vs lipid content basis). However, it can be seen in Table 2 that our results for ∑DDTs and ∑HCHs or lindaneare in agreement with those reported in Shanghai (Nakata et al, 2002) but in most cases much lower than reported in Tianjin, China where the ∑DDT residue level in spinach (102 ng/g) exceeded the EMRL of 50 ng/g for vegetables as recommended by the Chinese laws (Tao et al, 2005). In comparison with results obtained in other countries (Table 2), the residue levels of ∑DDT and ∑HCH in vegetables found in this study were comparable to found in vegetables from Gambia and Senegal (Manirakiza et al., 2003) but lower than found in Agra, India (Bhanti and Taneja, 2005), Debrecen, Hungary (Hovánszki et al, 2007) and Nigerian markets (Adeyeye and Osibanjo, 1999). A much more serious vegetable contamination was found in the urban markets of Ghana (Amoah et al, 2006) where residue levels of lindane and ∑DDTs upto 300 ng/g wet wt and 400 wet wt respectively, were found.

Table 1. Edible tissue concentration (ng/g wet wt) of organochlorine residues in vegetables from Deyang and Yanting, Sichuan

Vegetable (N)	α-HCH	β-HCH	γ-HCH	δ-HCH	∑ HCHs	p,p'-DDE	p,p'-DDD	o,p'-DDT	p,p'-DDT	∑DDTs
					Samples from Deyang					
Amaranth (1)	0.07	0.10	0.05	<0.01	0.22	0.06	0.02	<0.01	<0.01	0.08
Beans (3)	0.04	<0.01	0.02	<0.01	0.06	0.03	<0.01	<0.01	<0.01	0.03
	(0.10)a		(0.05)		(0.16)	(0.04)				(0.04)
Cabbages (4)	0.01	0.12	0.05	<0.01	0.17	0.25	0.02	0.04	0.07	0.3 9
	(0.02)	(0.37)	(0.10)		(0.46)	(0.92)		(0.15)	(0.29)	(1.27)
Celery (1)	0.01	0.09	0.06	0.01	0.17	0.03	<0.01	0.03	<0.01	0.06
Eggplant (1)	0.40	0.08	<0.01	<0.01	0.48	0.14	<0.01	<0.01	<0.01	0.14
Lettuce (6)	0.04	0.10	0.03	0.18	0.35	0.08	0.01	<0.01	0.09	0.24
	(0.09)	(0.26)	(0.08)	(0.47)	(0.57)	(0.31)	(0.05)		(0.30)	(0.85)
Onion (1)	<0.01	0.14	0.02	<0.01	0.16	0.05	0.01	<0.01	<0.01	0.06
Potato (2)	<0.01	0.02	0.04	<0.01	0.06	0.52	0.01	<0.01	0.10	0.64
		(0.03)	(0.08)		(0.11)	(1.02)	(0.02)		(0.21)	(1.22)
Pumpkin (1)	0.12	<0.01	0.08	<0.01	0.20	0.01	<0.01	<0.01	0.19	0.20
Radish (2)	0.05	0.36	0.11	0.04	0.56	0.12	0.04	<0.01	0.17	0.33
	(0.09)	(0.62)	(0.19)	(0.08)	(0.98)	(0.22)	(0.09)		(0.34)	(0.64)
Spinach (2)	0.01	0.09	0.08	0.06	0.24	0.04	0.03	0.03	0.03	0.12
	(0.01)	(0.12)	(0.09)	(0.08)	(0.26)	(0.05)	(0.04)	(0.04)	(0.05)	(0.15)
All Samples (24)	**0.05 (0.40)**	**0.09 (0.62)**	**0.05 (0.19)**	**0.05 (0.47)**	**0.25 (0.98)**	**0.13 (1.02)**	**0.01 (0.09)**	**0.03 (0.19)**	**0.06 (0.34)**	**0.24 (1.27)**
Positive samples (%)	**62.50**	**66.67**	**70.83**	**37.50**	**91.67**	**91.67**	**45.83**	**29.17**	**33.33**	**100.0**
					Samples from Yanting					
Cabbages (1)	0.02	0.23	0.02	<0.01	0.29	0.04	<0.01	0.11	<0.01	0.15
Pepper (1)	0.13	0.12	0.02	<0.01	0.27	61.41	0.41	82.59	4.04	148.44
Eggplant (1)	0.02	0.00	0.00	<0.01	0.02	<0.01	<0.01	<0.01	<0.01	<0.01
Lettuce (2)	<0.01	0.07	0.00	<0.01	0.09	0.01	<0.01	<0.01	<0.01	0.01
		(0.07)	(0.02)		(0.09)	(0.01)				(0.01)
Pumpkin (2)	0.03	0.07	0.02	0.17	0.30	0.05	<0.01	<0.01	<0.01	0.05
	(0.06)	(0.15)	(0.04)	(0.29)	(0.48)	(0.10)				(0.10)
Spinach (1)	<0.01	<0.01	<0.01	<0.01	<0.01	0.07	0.02	0.08	0.17	0.34
Rape stalk (2)	0.08	0.05	<0.01	<0.01	0.21	0.02	<0.01	<0.01	0.06	0.19
	(0.15)	(0.10)			(0.23)	(0.03)			(0.11)	(0.34)
All samples (9)[b]	**0.03 (0.15)**	**0.07 (0.23)**	**0.03 (0.08)**	**0.04 (0.29)**	**0.17 (0.48)**	**0.03 (0.10)**	**0.00 (0.02)**	**0.03 (0.11)**	**0.04 (0.23)**	**0.11 (0.34)**
Positive samples (%)	**50.00**	**60.00**	**70.00**	**20.00**	**90.00**	**70.00**	**20.00**	**40.00**	**30.00**	**80.00**
EMRL[c]			**50**		**50**					**50**

Note: [a] Maximum residue value
[b]Residues in pepper (*Capsicum annuum* L.) not included in the calculations but included in frequency counts.
ΣHCHs= α-HCH + *β*-HCH + *δ*-HCH + *γ*-HCH; ΣDDTs = *p, p'*-DDE + *p, p'*-DDD + *o, p'*-DDT + *p, p'*-DDT
[c] The Chinese statutory Extraneous Maximum Residue Limit according to food standard (GB 2763-2005)
N= number of samples analysed

3.2 Compositional Analysis for Possible Sources of OCPs

Relative abundances of the HCH congeners to he total HCHs (∑HCHs) were quite similar in all crops samples and when all samples were considered together the order β-HCH >α-HCH ≈γ-HCH>δ-HCH (in Deyang samples) and β-HCH >δ-HCH > α-HCH ≈γ-HCH (in Yanting samples). The predominance of β-HCH was probably due to its high stability, low water solubility and resistance to microbial degradation and because α-HCH can be converted to β-HCH in the environment (Willet et al, 1998). Surprisingly, the most prevalent isomer in the vegetables of this study, γ-HCH (lindane), was not the overall most abundant HCH isomer. This is attributable to the instability of γ-HCH, which may be converted to α-HCH in the presence of solar radiation (Willet et al, 1998).

The composition pattern in terms of the ratio of α -HCH to γ -HCH (lindane) can be used to monitor whether the source was technical HCHs or lindane The ratio α –HCH/γ-HCH should be 4–7 for technical HCH, and nearly zero (0) for technical lindane (Iwata et al., 1995). In the present study, the ratio α –HCH/γ-HCH varied from 0.00 to 0.00-0.70 in vegetables from Deyang and from 0.00-1.88 in vegetables from Yanting. It is important to note that the soils from the CER, Sichuan (including the present study areas) were also found with predominantly lower α-HCH/γ–HCH ratio than in technical HCH with 95% of the ratios being in the range 0.00 – 0.14 (Xing et al, 2009). Accordingly, it can be concluded that lindane was still being used in the areas studied.

The average concentration levels of DDT isomers/metabolites in the vegetables were observed in the following order: p,p'-DDE > p,p'-DDT > o,p'-DDT > p,p'-DDD in Deyang and p,p'-DDT > p,p'-DDE >o,p'-DDT >p,p'-DDD in Yanting. DDT is known to biodegrade to DDE under aerobic and to DDD in anaerobic conditions. The predominance of p,p-DDE and low levels of p,p-DDD in Deyang show that either the DDT compounds were from the historical sources or that the biotransformation of DDT in the vegetables is very efficacious. Conversely, the fact that p,p-DDT was the main contributor in vegetables from Yanting, indicates recent inputs of DDT in the area. Furthermore, ratios of (DDD + DDE)/ DDTs of >0.5 shows DDTs have been subjected to long term weathering (Doong et al, 2007). In the present study, 58.33% of the samples from Deyang and 40.00% of samples from Yanting had the ratio (DDD + DDE)/ DDTs >0.5 showing that both weathered and fresh DDT could be contaminating crops in the areas.

Table 2 Global comparison of Organochlorine pesticide residue levels in some vegetables (ng/g wet wt)

Vegetable	Location	n	Year	∑HCHs	∑DDTs	Reference
Cabbage	Deyang, China	4	2007	0.17	0.39	Present study
	Yanting, China	1	2007	0.29	0.15	Present study
	Tianjin, China	-	2002	38.00	34.00	Tao, et.al. (2005)
	Debrecen, Hungary	-	-	-	19.30	Hovánszki et al. (2007)
	Agra, India	-	2002-03	2.49 [a]	2.33	Bhanti & Taneja (2005)
	Gambia/Senegal	-	2002	1.10	0.12	Manirakiza et al. (2003)
Celery	Deyang, China	1	2007	0.17	0.06	Present study
	Tianjin, China	-	2002	40.00	32.00	Tao et al. (2005)
	Shanghai, China	1	2000	1.70	2.50	Nakata et al., 2002
	Debrecen, Hungary	-	2004	-	10.60[b]	Hovánszki et al. (2007)
Lettuce	Deyang, China	6	2007	0.35	0.24	Present study
	Yanting , China	2	2007	0.09	0.01	Present study
	Urban markets, Ghana	60	2002	300 [a]	400	Amoah et al (2006)
	Gambia/Senegal	-	2002	0.30	0.45	Manirakiza et al. (2003)
Amaranth	Deyang, China	1	2007	0.22	0.08	Present study
	Nigeria	11	1991-92	5.40	22.60	Adeyeye & Osibanjo (1999)
Spinach	Deyang, China	2	2007	0.24	0.15	Present study
	Yanting , China	1	2007	<0.01	0.34	Present study
	Tianjin, China	-	2002	43.00	102.00	Tao, et al (2005)
	Shanghai, China	1	2000	<0.03	0.14	Nakata et al. (2002)
	Agra, India	-	2002-03	14.17	6.61	Bhanti & Taneja (2005)
Eggplant	Deyang, China	1	2007	0.49	0.14	Present study
	Yanting , China	1	2007	0.02	<0.01	Present study
	Shanghai, China	1	2000	0.09	<0.03	Nakata et al.(2002)
	Agra, India	-	2002-03	6.40 [a]	8.44	Bhanti & Taneja (2005)
	Gambia/Senegal	-	2002	0.30	5.10	Manirakiza et al. (2003)
	Nigeria	11	1991-92	3.40	21.50	Adeyeye & Osibanjo (1999)
Pumpkin	Deyang, China	1	2007	0.20	0.20	Present study
	Yanting	2	2007	0.30	0.05	Present study
	Agra, India	-	2002-03	0.04 [a]	ND	Bhanti & Taneja (2005)
	Debrecen, Hungary	-	2004	-	19.3 [b]	Hovánszki et al. (2007)
Radish	Deyang, China	2	2007	0.56	0.33	Present study
	Agra, India	-	2002-03	6.15	8.44	Bhanti & Taneja (2005)
	Gambia/Senegal	-	2002	0.30	1.44	Manirakiza et al. (2003)
Onion	Deyang, China	1	2007	0.16	0.06	Present study
	Shanghai, China	1	2000	0.15	0.07	Nakata et al (2002)

Note: Where more than one site was sampled, the site with highest level was taken for this comparison.
[*] ng/g dw
[a] mean level of γ-YCH; [b] mean value for DDE; [c] composite samples; ND=not detected; - Not available

3.3 Seasonal Variation

The residue levels and the detection rates of the OCPs are given in Table 3. The autumn vegetables had higher OCPs levels than spring vegetables. This may be because more pests were active during summer when the vegetables were growing – a situation which might have necessitated the illegal use of some of the OCPs. It is also possible that the high temperatures in summer volatilized the OCPs from their reservoirs such as soil or vegetation and that the edible parts of the crops may have trapped some of the evaporating pesticides.

It can also be seen in Table 3 that residues of β-HCH, γ-HCH (lindane) and hence ∑HCHs were more prevalent in spring samples than in autumn ones. In particular, the insecticide lindane was quantifiable in all the spring vegetables, which suggests that the vegetables might have been contaminated at the beginning of spring or earlier when they were planted. In the southern China region, lindane was mainly used in summer as a seed dressing or a general insecticide (Yang et. al., 2007).

The distribution of the DDTs was similar in both seasons, with degradation products p,p'-DDE and p,p'-DDD leading in prevalence especially in the winter crops. This further suggests historical rather than current sources of DDTs in the areas studied.

Table 3 Residue levels (ng/g wet wt) and incidence (%) of organochlorine pesticides in vegetables from Deyang and Yanting , Sichuan

Compound	Spring Vegetables (N=18)			Autumn Vegetables (N=16)
	Incidence (%)	Average (min-max)[a]	Incidence (%)	Average (min-max)[a]
α-HCH	50.00	0.02 (<0.01-0.15)	75.00	0.07 (<0.01-0.40)
β-HCH	94.44	0.13 (0.01-0.37)	25.00	0.06 (<0.01-0.62)
γ-HCH	100.00	0.06 (0.02-0.10)	50.00	0.03 (<0.01-0.19)
δ-HCH	27.77	0.01 (<0.01-0.0.08)	41.67	0.08 (0.06-0.47)
∑HCHs	100	0.19 (0.02-0.46)	81.25	0.24 (<0.01-0.98)
p,p'-DDE	88.89	0.03 (<0.01-0.06)	87.5	0.20* (<0.01-61.41)
p,p'-DDD	38.89	0.01 (<0.01-0.04)	31.25	0.01* (<0.01-0.41)
o,p'-DDT	38.89	0.05 (<0.01-0.18)	18.75	0.02*(<0.01-82.59)
p,p'-DDT	11.11	0.02 (<0.01-0.23)	50.00	0.11* (<0.01-4.04)
∑DDTs	100.00	0.09 (0.01-0.34)	87.50	0.34* (<0.01-148.44)

∑DDTs = p,p'-DDT + o,p'-DDT + p,p'-DDD + p,p'-DDE; ∑HCHs= α- + β- + γ- + δ-HCH
* DDT values for one sample, pepper not included in the calculation of the mean

4.0 Conclusions

This survey revealed high occurrence rates but low residue levels of OCPs in vegetables produced in Deyang and Yanting farmlands Southwest China. Since most of the determined residue levels are far below the prescribed national and international residue limits, it can be concluded that there was a good observance of limitations in agricultural application of the chlorinated pesticides. However, the high prevalence of contamination was worrisome considering the cumulative nature, level of persistence of these pesticides and, especially, the high amounts of vegetables in the Chinese diets. Consumption of pesticide free vegetables and elimination of the vegetable OCP contamination are recommended.

Acknowledgement

This study was part of the project financed by the Natural Science Foundation of China under Grant No. 40473043 and Geological Survey of Sichuan Province, People's Republic of China. We thank Su Qiuke and the environmental organic Chemistry post graduate students of China University of Geosciences (Wuhan), who helped with the collection and /or extraction of the samples.

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