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Distribution Characteristics of Organochlorine Pesticides in Soil and Groundwater of Different Irrigation Farmlands in Southeast Suburb of Beijing

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1. Introduction

Wastewater has been widely used in agriculture irrigation for nearly a century in the world, due to the scarcity and quality declination of water resources. The wastewater irrigation technologies in USA, Japan, Israel and some developed countries are more mature than other developing countries (Angelakis A N et al., 1999; U Pinto et al., 2010; US EPA, 2004). Although wastewater irrigation can supply a lot of nutrients to crop, such as Nitrogen and Phosphor sources, it can cause soil pollution, like heavy metals, organic pollution and so on. Therefore, reclaimed water, instead of wastewater, as agriculture irrigation water resources became more popular now, because not only can reclaimed water relieve the tensions of agriculture water resource, but also relieve the pressure of wastewater on environment (T.Asano et al., 1996; D.Levine et al., 2004; I.K.Kalavirouziotis et al., 2008; V.Reboll et al., 2000). However, the nature of reclaimed water is still a poor-quality water, the amounts of contaminants in reclaimed water are changed with treatment, still a lot of pollutants can not be effectively removed. Some matters in reclaimed water may benefit crops, such as N, P, K, some else may be harmful to the surrounding environment, crop, operator and consumer of agriculture products, like heavy metals, salts, and various carcinogenic, teratogenic, or mutagenic organic compounds, the long-term irrigation will bring harmful to soil and groundwater, causing a new source pollution (Francisco P et al., 2009; Friedler E et al., 2004; Gross A et al., 2007).

As a large agricultural country, the agricultural irrigation water consumption accounts for more than 70% of the total water consumption in China. Due to water shortage, wastewater irrigated area expanded rapidly from 1970s to the 1990s. By 1998, wastewater irrigated area has reached 361.8×10^4 hm^2, which accounting for 7.3% of the total irrigated area. The city of Beijing in China began to use wastewater irrigation since the early 50s of the 20th century, so far, the wastewater irrigation area in Beijing are nearly 80,000 hm^2 and the yearly wastewater irrigation is about 220 million m^3 which accounting for 20% of the city's wastewater emissions, 87% of the wastewater irrigation area are mainly in Tongzhou District, Daxing District and Chaoyang District (Zhang H Y et al., 2006; Hua X M et al., 1996). The quality and fertility of domestic sewage are better and beneficial to rice, but the industrial effluents are not as well, it contains some heavy metal salts, such as lead, chromium, arsenic, mercury, and some harmful ingredients, take chlorine, sulfur, phenol,

cyanide for example, all of them are bad for rice growing, therefore, long-term use of wastewater irrigation is bound to lead a series of pollution problems.

In recent years, along with urban sewage treatment rate rising, reclaimed water reuse has been strengthened. Various studies and experimental demonstration bases have been performed, which greatly promoted the reclaimed water reuse in agricultural and associated research. As one of the most serious water shortage city in China, Beijing is constantly increasing the intensity use of reclaimed water irrigation. In 2006, about 199 million m^3 reclaimed water were used for agriculture irrigation in Beijing. By 2010, the Beijing center city reclaimed water production capacity will reach 1.4 million m^3 everyday, annual renewable water consumption will reach 600 million m^3, recycled water will become the major water sources for irrigation, industrial cooling, and ecological water use et al (LIU P B, 2007). However, even the reclaimed water quality has been greatly improved, it still has potential effects on crop growth, soil quality, and groundwater quality during the process of irrigation, especially some refractory organics will persist in the environment. These problems have received widely concern of the publics. The use of reclaimed water in China was only at the initial stage. Therefore, a lot of works need to be done to confirm the safety use of reclaimed water.

Organochlorine Pesticides (OCPs) are a kind of synthetic chemical pesticides, which can be divided into two branches, one is take benzene as raw materials, the other is cyclopentadiene, their physical and chemical properties are stability, both the persistence and bioaccumulation are strong, they are also difficult to degrade naturally in environment, but they can be a threat to ecosystems and human healthy through evaporation, migration, food chain transfer and other paths (Tan Z et al., 2008; Katsoyiannis A et al., 2005). Among 12 kinds of Persistent Organic Pollutants (POPs) listed in the Sidegemo Convention, 9 of them are OCPs (Liu C et al., 2008). Once these pollutants were released to the environment, they will exist in environment for many years. Even in some areas OCPs never been applied before, we can also find the existence of them, this indicates that the OCPs have stronger migration ability in space. Although some of the OCPs had been banned in China since 1983, the harm caused to the environment still exists due to their refractory properties.

OCPs are widely distributed in various environment media, such as soil sediments, plant tissues, animal organs and body. In which the soils are usually the main destination and accumulation sites for these pollutants, it often acts as pollutants carrier and offer the natural purification place. Therefore, it is important to study the pollution characteristics of OCPs in soils and assess the potential of groundwater contamination. Based on the above, 3 typical farmlands, i.e. wastewater irrigated farmland, reclaimed water irrigated farmland and groundwater irrigated farmland of Beijing suburb were selected for study. Both soil samples of vertical profiles, irrigation water and groundwater samples of each farmland were collected and analyzed. Through detail comparison of OCPs contents in each soil vertical profile and OCPs concentration in irrigation water and groundwater, the distribution characters of OCPs in soil profiles were summarized, the impact of irrigation water and controlling factors of OCPs migration were also discussed.

2. Materials and methods

2.1 Research area description

Both 3 farmlands selected in this study are all located in the south-east suburb of Beijing. The wastewater irrigated farmland (WIF) makes use of Liangshui River's water for irrigation. Liangshui River is the second-largest drainage river in Beijing. It collects domestic water and industrial wastewater from Beijing centre city. This WIF has a long history of

wastewater irrigation, which began from the 20th century 60s and with an area of about 1.24×10^4 hm^2 so far. Irrigation periods of each year are from March to June and the middle September to December, respectively. The average water use is about 3×10^3 $m^3 \cdot hm^{-2} \cdot a^{-1}$.

The reclaimed water irrigated farmland (RIF) selected in this study is located in Duozi village, Ciqu town, which is about 4 kilometers northwest to the WIF. Irrigation water used in this farmland was came from Tonghui irrigation canal which got through the farmland from north to south. Many branches of Tonghui irrigation canal provide sufficient reclaimed water for agriculture irrigation. This canal was built in 1958, which started from the Gaobeidian Lake, and ended in Liangshui River. The full length is about 17 kilometers. It was sewage canal historically which accepted sewage water from Tonghui River. In 1993, the largest sewage treatment plant in Beijing named Gaobeidian was completed and put into operation. From then, Tonghui irrigation canal began to accept reclaimed water. Therefore, this region was also wastewater irrigated area before 1993. The history of reclaimed water irrigation is no more than 20 years.

Groundwater irrigated farmland (GIF) is located in Yongle town, Tongzhou District of Beijing, which is about 18 kilometers southeast to the WRF. The irrigation water for this farmland is groundwater, extracting from 60m underground through the motor-pumped wells.



Fig. 1. Location of the study areas

2.2 Sample collection

Soil profiles with depth of 6.0 m were drilled using Eijkelkamp soil sampler in farmlands. Each farmland has 3 profiles, and they were distributed at the vertices of equilateral

triangular with a side length of 1.0 m. Therefore, total 9 boreholes were drilled. A, B and C profiles were drilled in the WIF. D, E, and F profiles were drilled in the RIF. G, H, and I profiles were drilled in the GIF. Soil samples were collected from surface to 5.5 m underground every 0.5m. So, there were 12 samples taken from each borehole, and 108 soil samples named from A00 to I55 were collected from three farmlands. Soil samples were stored in brown glass bottles, sealed with film, and kept store no more than 20 days. Physical-chemical properties of soil samples such as Clay content, total amount of clay minerals, Cation exchange capacity (CEC), Total organic compounds (TOC), et al were analyzed. 14 kinds of OCPs including α-HCH, β-HCH, γ-HCH, δ-HCH, 4,4'-DDT, 2,4'-DDT, 4,4'-DDE, 4,4'-DDD, Hexachlorobenzene, Heptachlor, Aldrin, Heptachlor epoxide, Dieldrin and Endrin were also analyzed using Gas chromatography.

The groundwater and surface water samples near the profiles were also taken for analyses in each farmland. Water samples were placed in incubator to refrigeration and avoid light, sent to the laboratory at the same day and then stored in refrigerator at 4 ˚C.

2.3 Sample preparation

Soil sample preparation: According to the EPA 3550, 15g soil sample, 3g sodium sulfate anhydrous and 20 mL extraction solvent (1:1 acetone/ hexane mixed solvent) were added into a 40 mL vial. The soil was ultrasonically extracted at 50 °C in 400 W for 20 min to promote the diffusion of OCPs from the soil into the extractant, and then the vials were centrifuged at 3000 rpm for 3 min. After that, the organic phase was transfered to 100 mL glass bottle. Repeated this extraction twice, combined the extracts. The extracts then were dehydrated with sodium sulfate anhydrous, and concentrated to 1.0 mL using a rotary evaporator in water bath at 35-40 °C. OCPs in extracts were purified by prepared SPE florisil columns, and concentrated to 1.0 mL through nitrogen blow for further chromatographic analysis.

Water sample preparation: According to the EPA 3510, water samples were filtered with APFF glass fiber membrane at first. Then 1 L filtered water was put into separator funnel and added 20 mL cyclohexane. Then Liquid-liquid extracted for 10 min, held for 10 min, and transfered the organic phase. This extraction should be repeated twice, and combined the organic phase. The extracts then were dehydrated with sodium sulfate anhydrous, and concentrated to 1.0 mL using a rotary evaporator in water bath at 35-40 °C. OCPs in extracts were purified by prepared SPE florisil columns, and concentrated to 1.0 mL through nitrogen blow for further chromatographic analysis.

2.4 Analysis methods

The purified sample extracts for OCPs analysis were quantitatively analyzed using a HP-6890 gas chromatograph, equipped with electron capture detector (ECD) and a HP5 capillary column (30 m×0.32 mm×0.25 μm). The oven temperature was programmed begin from 80°C and kept for 2 min, then increased from 80°C to 185°C at the rate of 30 °C/min, and to 215 °C at the rate of 3 °C/min, held for 4 min and then to 225 °C at the rate of 1 °C/min and held for 2 min, finally increased to 290 °C at the rate of 20 °C/min and held for 2 min. Helium was the carrier gas at a flow rate of 1.0 mL/min. The temperatures of the injector and detector were set up at 250 °C and 320 °C, respectively. 1.0 μL of sample was injected under the splitless injection mode.

Instrument detection limits and method detection limits were list in table 1. Separation effect of mixed standard sample which contains 14 kinds of OCPs was shown in figure 2. Table 2 list the testing methods of soil physical-chemical properties.

3. Physical-chemical properties of soil profile in farmlands

3.1 Physical-chemical indexes

Soil Physical-chemical properties are important factors affecting the vertical migration of organic pollutants in vadose zone. 8 indexes of each sample including pH, Eh, soil moisture (%), EC, clay content (%), total clay minerals (%) total organic carbon content (%), and cation exchange capacity (CEC) were tested in this study. The vertical variations of 8 indexes were shown in Figure 3.

ITEM	Method limits (μg/kg)	Instrument limits (μg/L)
α-HCH	0.06	0.60
Hexachlorobenzene	0.07	0.65
β-HCH	0.04	0.40
γ-HCH	0.07	0.65
δ-HCH	0.60	5.75
4,4'-DDE	0.01	0.05
4,4'-DDD	0.04	0.35
2,4'-DDT	0.25	2.45
4,4'-DDT	2.00	19.5
Heptachlor	0.07	0.65
Aldrin	0.04	0.40
Heptachlor expoxide	0.04	0.35
Dieldrin	0.05	0.50
Endrin	1.00	9.65

Table 1. The detection limits of OCPs

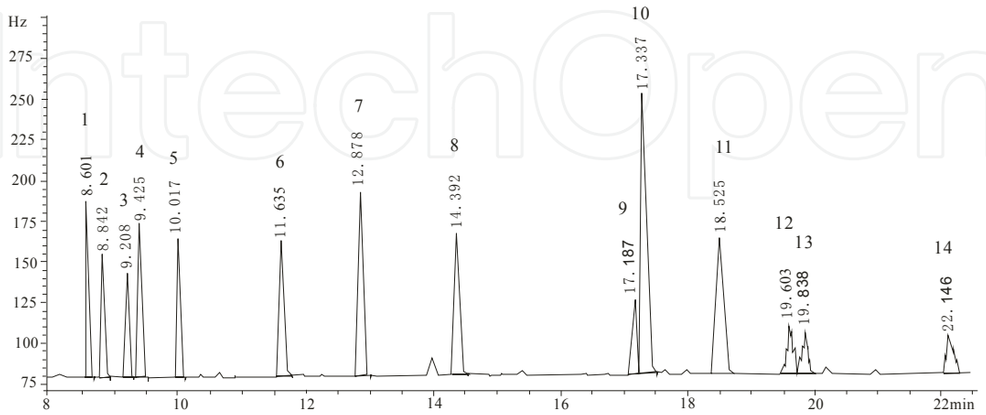


Fig. 2. 1. α-HCH, 2. Hexachlorobenzene, 3. β-HCH, 4. γ-HCH, 5. δ-HCH, 6. Heptachlor, 7. Aldrin, 8. Heptachlor expoxide, 9. 4,4′-DDE, 10. Dieldrin, 11. Endrin, 12. 4,4′-DDD, 13. 2,4′–DDT, 14. 4,4′-DDT.

Testing project	Testing method
Soil moisture	Velocity and mass method
pH	Potentiometric method
CEC	Ammonium chloride-ammonium acetate exchange and ammonium acetate exchange method
TOC	External heating potassium dichromate oxidation method
EC	Conductivity method
Eh	Potentiometric method
Grain composition	Hydrometer method
Clay mineral	X－ray diffraction method

Table 2. Testing projects and methods of physical-chemical properties

As Figure 3 shows, the soil physical-chemical properties of these 3 farmlands had no obvious differences in general. The soil moisture of WIF profiles varied between 18% - 39%, maximum soil moisture all appeared at the 1.5 m in 3 boreholes. The profiles of RIF and GIF basically had the same variation of soil moisture, mostly changed between 10.91% -36.98%, and most lower soil moisture appeared at 0-0 .5 m. The pH values of the three farmlands were basically around 7.7, rendered as a neutral environment, and slightly increased along the profiles. The soil redox potential variations with depth were consistent in 3 farmlands profiles, and oxidation environment played a leading role in the whole profiles. In contrast, RIF has higher soil redox potential values, GIF in the middle, WIF in the last. The CEC variations with depth were consistent in 3 farmlands profiles too. The CEC values of WIF and RIF were 2.13-18.00 cmol/kg and 4.90-16.44 cmol/kg respectively, and maximum value appeared in 1.0-1.5 m both for WIF and RIF. The CEC values of GIF were 3.37-9.74 cmol/kg, and maximum value appeared in 3.0 m. Compared with WIF and RIF, GIF has higher EC, the high values was in 1.5-3.0 m. Clay content and clay minerals contents were almost same, and their changes alone the profiles in 3 farmlands had no obvious differences too. However, we found that clay contents with slightly higher values were in 1.0-1.5m in WIF and RIF, and 2.0-3.0m in GIF. The changes of TOC content alone the profiles in 3 farmlands were more obvious than other indexes, significant reductions were found in almost all soil profiles, the average maximum TOC content occurred at top soils. Under the top layer, higher TOC content layers can be outlined in 1.5 m in WIF profiles and 3-4.5 m in RIF profiles. However, there were no obvious high TOC content layers in GIF profiles.

3.2 Profile textures

The profile soil textures of three farmlands based on soil grain size were shown in figure 4. Accumulated particles percentages were used to help us found out main particle composition. Particle sizes were divided into three classes, i.e. sand (>0.075 mm), silt (0.075-0.005 mm) and clay (<0.005 mm) respectively according soil classification standards.

As shown in figure 4, the main soil particles in three farmlands were silt, followed by clay. Only small percentages were sand. However, profiles in RIF had more sandy particles than GIF and WIF statistically. That to say, pollutants were more easier to migrate in RIF than in GIF and WIF, only from the view of soil textures.

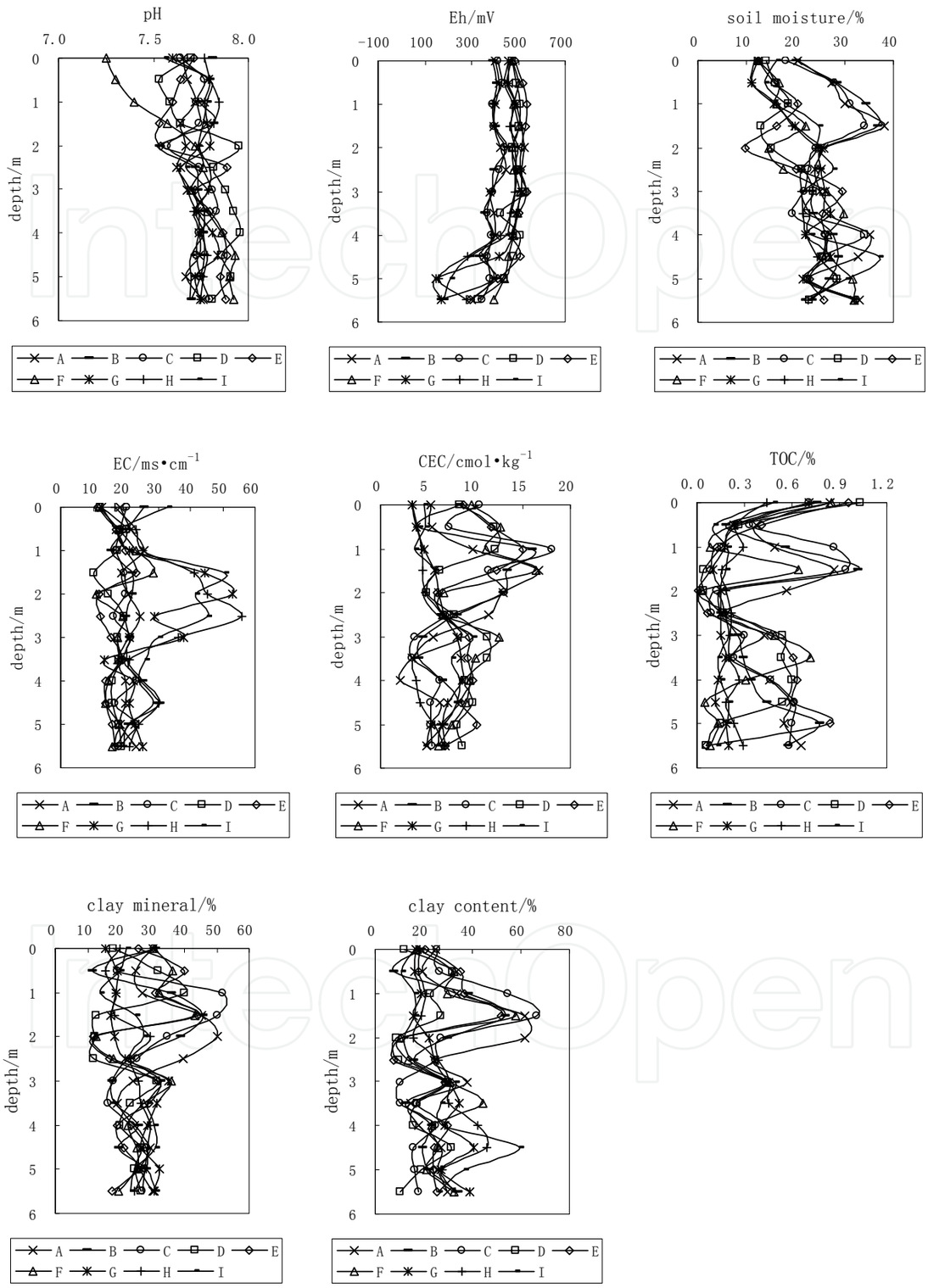


Fig. 3. Variation of soil Physical-chemical indexes with depth in three farmlands

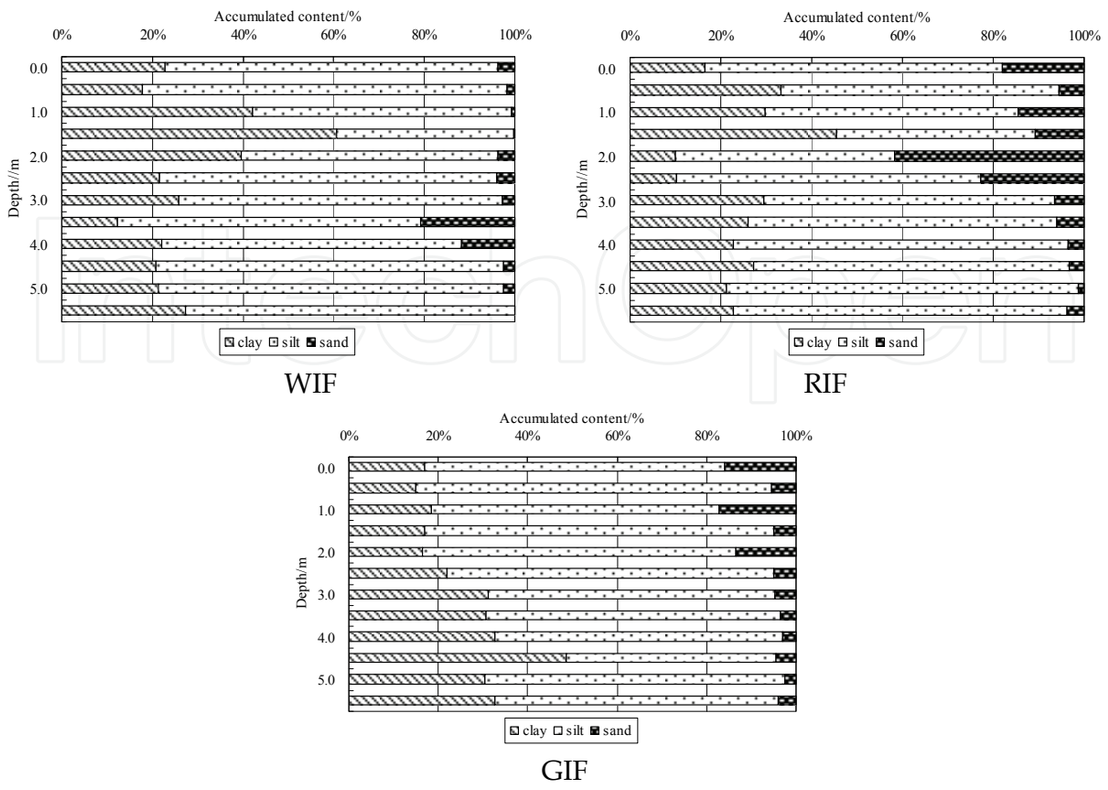


Fig. 4. Soil textures classification of three farmlands

4. The vertical distribution of OCPs in three farmlands

4.1 The distribution of OCPs in soil profiles of WIF

4.1.1 Vertical change of total OCPs

There were 14 kinds of OCPs detected in three boreholes, including α-HCH, β-HCH, γ-HCH, δ-HCH, 4,4'-DDT, 2,4'-DDT, 4,4'-DDE, 4,4'-DDD, Hexachlorobenzene (HCB), Heptachlor, Aldrin, Heptachlor epoxide, Dieldrin and Endrin. The changes of total OCPs along the profiles and the detected OCPs species numbers in 3 profiles were shown in figure 5.

Clearly we can see the highest ∑OCPs content was in top soil layers, and immediate drop in the whole profiles under the top layer. The maximum content was 30.93 μg/kg, which was at least 13 times of other layers. That means most OCPs were not easy to migrate vertically in these profiles. The detected OCPs numbers in surface soil were the most in 3 profiles, and decreased with depth. Under the surface, the greatest detected OCPs number was only 3. That's to say, 7-8 kinds of OCPs were detected in the surface soil, but only 3 kinds of OCPs detected under the surface. In additions, Heptachlor epoxide, Dieldrin and Endrin were not detected in three profiles.

Figure 6 described OCPs' compositions in surface soils. DDTs (mostly 4,4'-DDE) and HCHs (mostly δ-HCH) were the highest contents OCPs, which highest content were 20.03 μg/kg and 10.46 μg/kg, respectively, lower than the first class soil standard (<50 μg/kg) of Chinese soil environmental quality standard (GB15618-1995). Aldrin and HCB were also found in surface soils with lower contents. That's to say, DDTs and HCHs were the main pollutants in surface soils.

However, from the detection frequency of the 3 profiles (figure 7), we can see that Heptachlor had the highest detection frequency (66.7%), followed by DDTs (50.0%) and Aldrin (41.7%) in profile A. In profile B, DDTs had the highest detection frequency (66.7%), followed by Heptachlor (41.7%). Same to profile B, the OCPs which had the highest detection frequency in profile C was DDTs (58.3%), followed by Aldrin (50.0%) and Heptachlor (33.3%). That means except DDTs and HCHs, Heptachlor and Aldrin were the main pollutants in the profiles too.

4.1.2 Vertical variation of single OCP

DDTs, HCHs, Heptachlor, Aldrin were chosen as the main objects to discuss the vertical distribution of OCP, the variation were shown in figure 8.

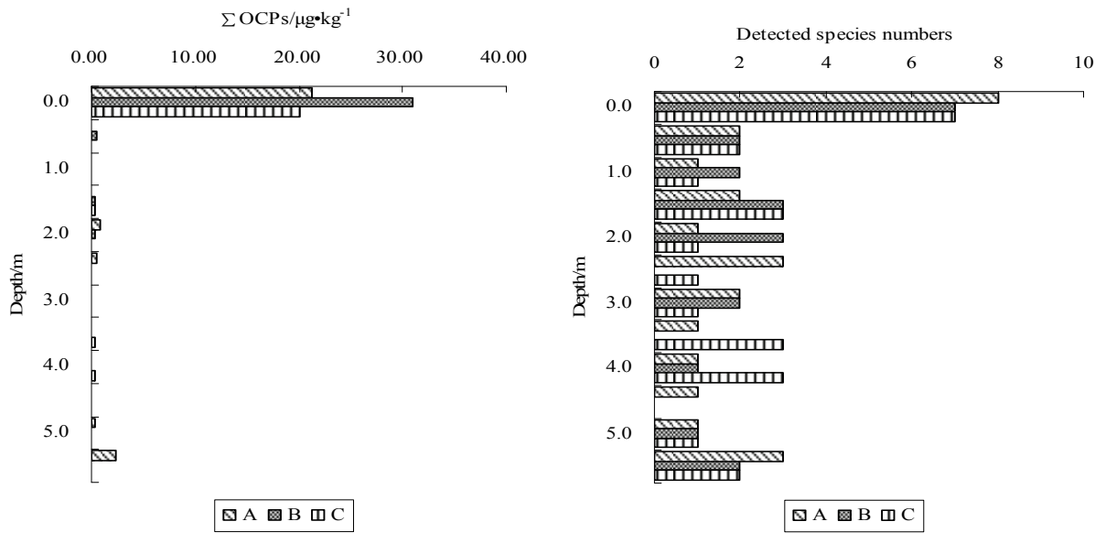


Fig. 5. Vertical distribution of ∑OCPs and detected OCPs species number in WIF

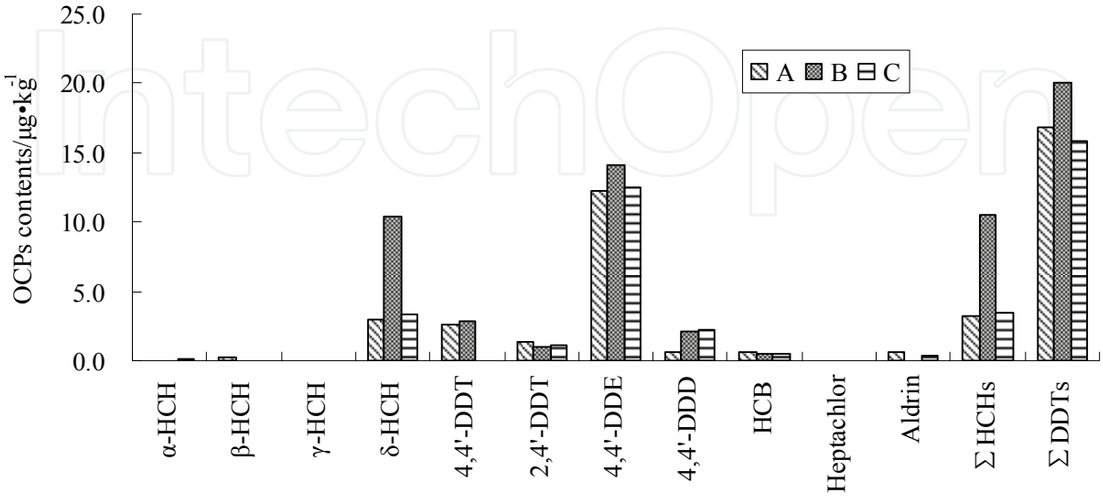


Fig. 6. Distribution of OCPs in surface soils of WIF

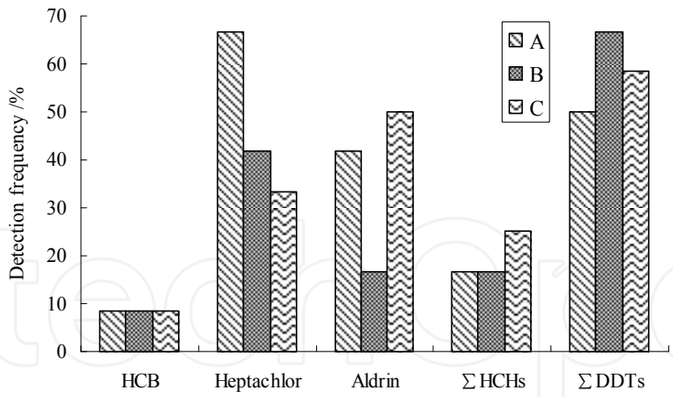


Fig. 7. Detection frequency of OCPs in WIF profiles

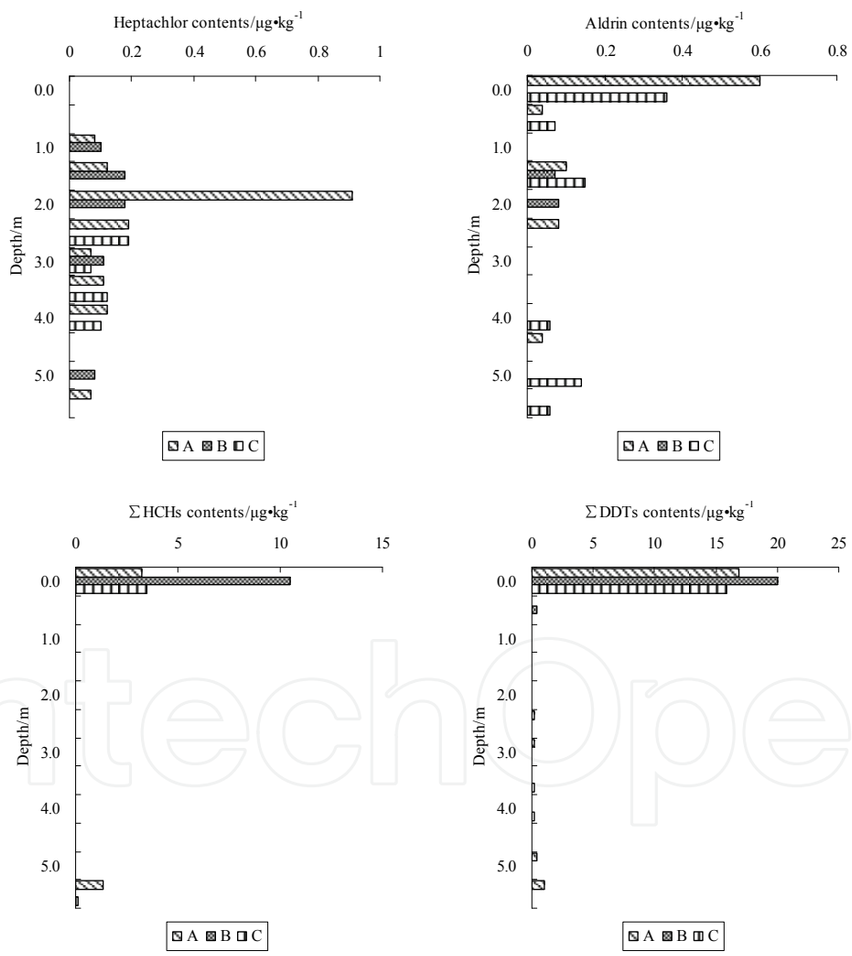


Fig. 8. Vertical distribution of single OCP in profiles of WIF

DDTs and HCHs were the main OCPs in WIF, their vertical distribution in generally took on sharp dropped look with depth and then kept very low content under the surface, which was same to the ∑OCPs. Aldrin contents in the profiles also dropped very fast, but didn't like DDTs and HCHs drop so much.

The changes of Heptachlor in the profiles were quite different from former mentioned OCPs, there was no Heptachlor detected in surface soils, but under the surface, Heptachlor were detected from 1.0 m to 5.5 m. Considered the physical-chemical properties of these OCPs, we can find out that DDTs have higher logK_{OW} and lower Henry's Law Constant, however, except higher logK_{OW}, Heptachlor also have higher Henry's Law Constant (table 3). That means both DDTs and Heptachlor were easy to be absorbed by soil and hard to migrate, but Heptachlor much easier to volatilize and lost in surface soil than DDTs. This can be a reasonable explanation for the difference.

OCPs	Molecular Weight	Water Solubility 25°C	logK_{OW} (octanol-water)	Henry's Law Constant	Vapor Pressure
		mg·L^{-1}		atm-m^3/mole	mm Hg
α-HCH	291	2	3.8	1.22E-05	-
β-HCH	291	0.24	3.78	4.40E-07	-
γ-HCH	291	7.3	3.72	5.14E-06	4.20E-05
δ-HCH	291	10	4.14	4.29E-07	3.52E-05
4,4'-DDT	354.5	0.0055	6.91	8.32E-06	1.60E-07
2,4'-DDT	354.5	0.085	6.79	7.41E-06	3.44E-12
4,4'-DDE	318	0.04	6.51	4.16E-05	7.43E-12
4,4'-DDD	320	0.09	6.02	6.60E-06	1.35E-06
Hexachlorobenzene	285	0.0062	5.73	0.0017	1.80E-05
Heptachlor	373.5	0.18	6.1	2.94E-04	4.00E-04
Aldrin	365	0.017	6.5	4.40E-05	1.20E-04
Heptachlor epoxide	389.2	0.2	4.98	2.10E-05	1.95E-05
Dieldrin	381	0.195	5.4	1.00E-05	5.89E-06
Endrin	381	0.25	5.2	6.36E-06	3.00E-06

Table 3. Physical-chemical properties of 14 kinds OCPs

4.2 The distribution of OCPs in soil profiles of RIF

4.2.1 Vertical change of total OCPs

The changes of total OCPs along the profiles in RIF and the detected OCPs species numbers in 3 profiles were shown in figure 9.

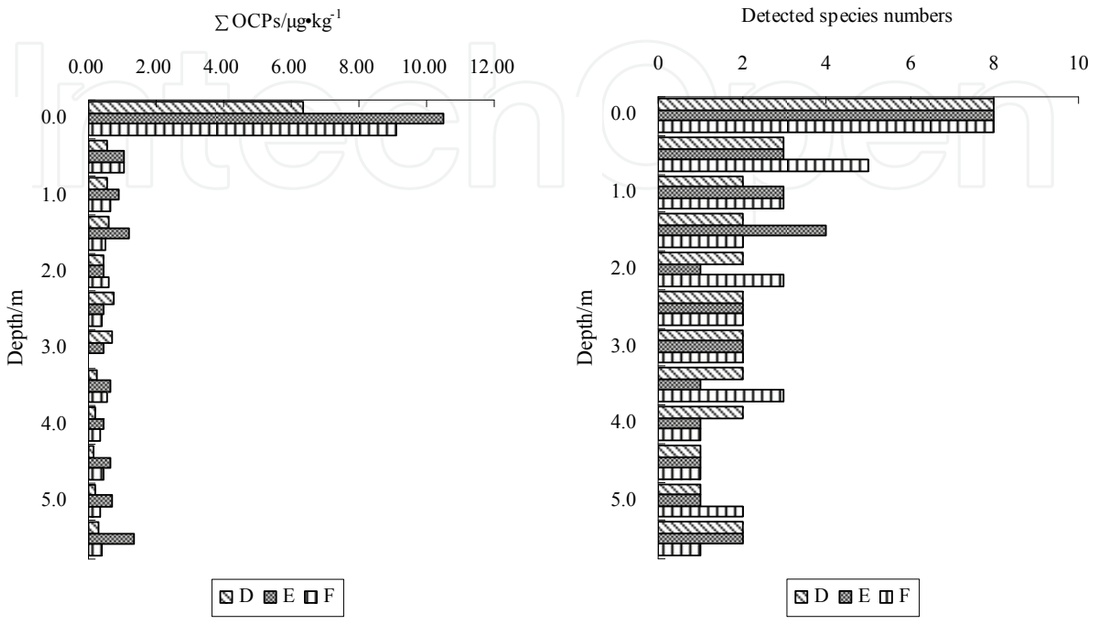


Fig. 9. Vertical distribution of ∑OCPs and the detected OCPs species numbers in RIF

From figure 9, we can see that the vertical variation of OCPs in three profiles were consistent, the contents decreased rapidly with depth, and then changed slowly, ended with consistent. The maximum contents all occurred in surface soils, of which the highest content was in profile E, reached 10.50 μg/kg. Compared the ∑OCPs in surface soil with WIF, the total OCPs in RIF was about 2 times lower. There were no apparent differences under the surface of two farmlands, but the total OCPs in RIF was slightly higher than WIF. The detected OCPs numbers in surface soil were the most in 3 profiles, and the amount decreased with depth. Under the surface, the greatest detected OCPs number were 5. That's to say, 8 kinds of OCPs were detected in the surface soil, but only 1-5 kinds of OCPs detected under the surface. In addition, Heptachlor epoxide, Dieldrin and Endrin were not detected in three profiles.

Figure 10 shows that, HCHs, DDTs, Heptachlor and Hexachlorobenzene were detected in the surface soils, the main pollutants were HCHs and DDTs, their highest contents were 6.82 μg/kg and 1.63 μg/kg, both of them were lower than the WIF, even in an order of magnitude. The lowest detected content was Hexachlorobenzene, and the value was 0.69 μg/kg. 4,4'-DDE was the main composition of detected DDTs and δ-HCH was the main composition of detected HCHs.

Figure 11 shows that, Heptachlor was detected in all three profiles and the detection frequency in profile D and E was 100%, followed by Aldrin and DDTs. The detection frequency of HCB was the lowest. Therefore the main OCPs in RIF were Heptachlor, Aldrin and DDTs.

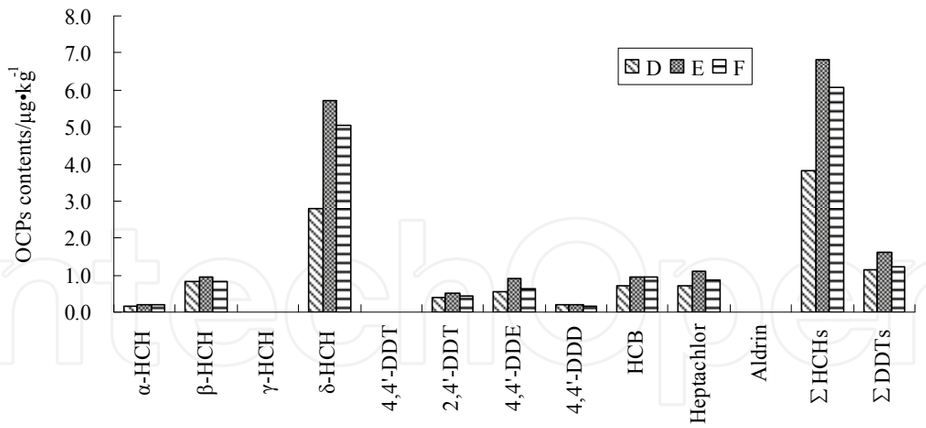


Fig. 10. Distribution of OCPs in surface soil of RIF

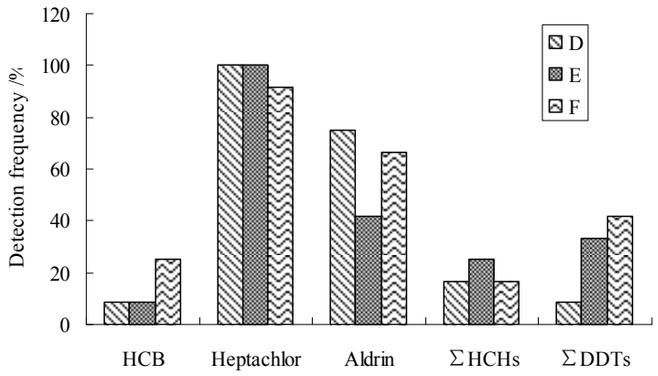


Fig. 11. Detection frequency of OCPs in RIF profiles

4.2.2 Vertical variation of single OCP

Heptachlor's, Aldrin, DDTs and HCHs were chosen as the main objects to discuss the vertical distribution of OCP, the variation were shown in figure 12.

DDTs and HCHs were the main OCPs in RIF, its vertical distribution in generally took on sharp drop look with depth and then kept very low content under the surface, which was same to the ∑OCPs changes, and also to WIF. Although Aldrin was not detected in RIF's surface soil, it was found in most sub-layers of the 3 profiles.

The changes of Heptachlor contents in the profiles were different from other OCPs. There was no apparent difference between top layer and sub layers. As we discussed before, both DDTs and Heptachlor were hard to migrate in vertical profiles. But Heptachlor was easier to volatilize in surface soil layer than DDTs. This supposed that surface soil layers of RIF maybe suffered more serious pollution relatively. That's why Heptachlor still can be detected in the surface soils. As we know, Octanol-water partition coefficient and Henry's Law Constant of Aldrin were close to Heptachlor. There was no Aldrin detected in surface soil may be due to volatilization.

In addition, heptachlor epoxide in all layers of three profiles was not detected which meant that there was no degradation reaction of Heptachlor. Dieldrin was not detected in three profiles which meant that Aldrin had not changed.

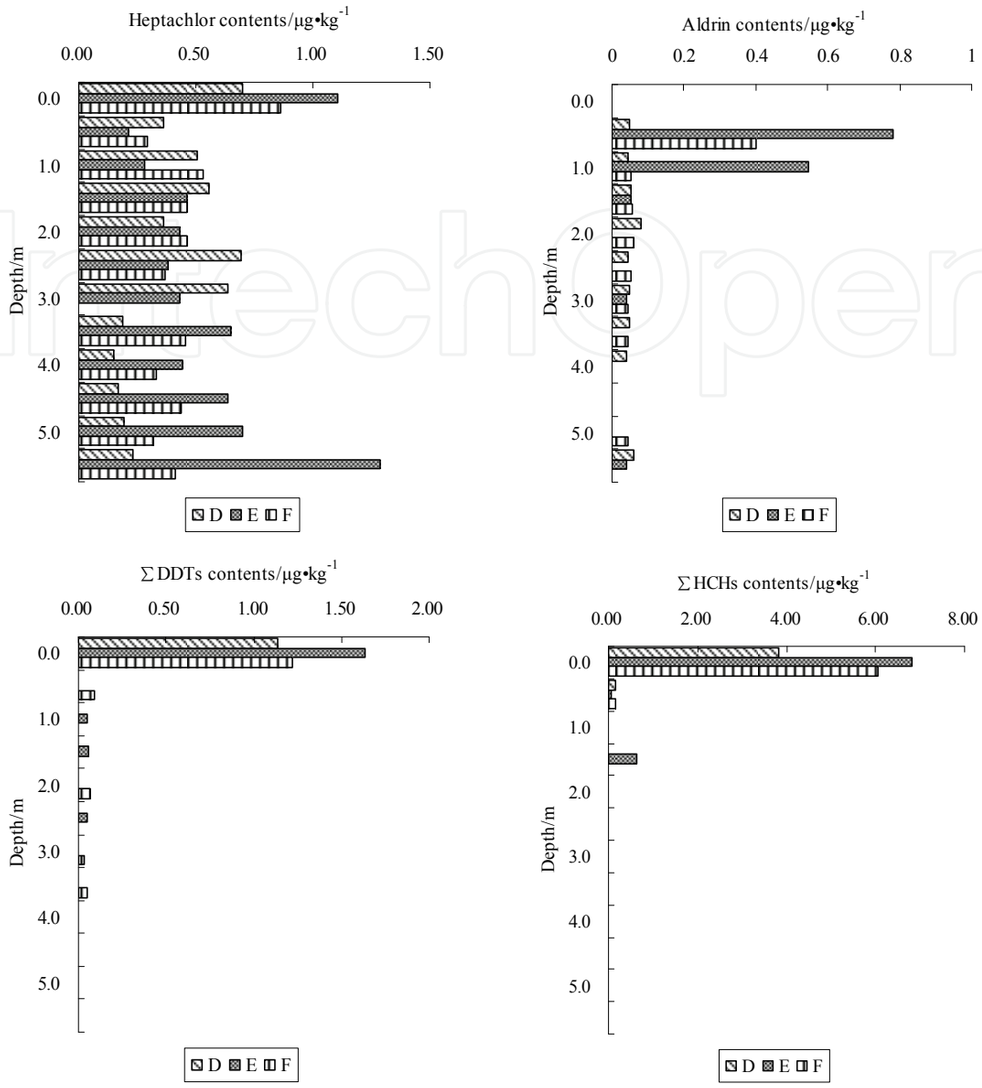


Fig. 12. Vertical distribution of single OCP in profiles of RIF

4.3 The distribution of OCPs in soil profiles of GIF

Groundwater is an ideal irrigation water resource. Using groundwater for irrigation can not only avoid new pollution sources to soil, also can dilute the pollutants in soil. Because the groundwater irrigated farmland has no history of wastewater irrigation, it can be regarded as background.

4.3.1 Vertical change of total OCPs

The distribution of total OCPs in soil profiles of GIF and detected OCPs species number of 3 profiles were shown in figure 13.

The vertical variations of ∑OCPs in boreholes of GIF were different from WIF and RIF. We can not see the typical drop of ∑OCPs contents from surface layer to sub layers. On the contrary, the maximum content was detected in 5.5 m, and the value was 6.77 μg/kg. But

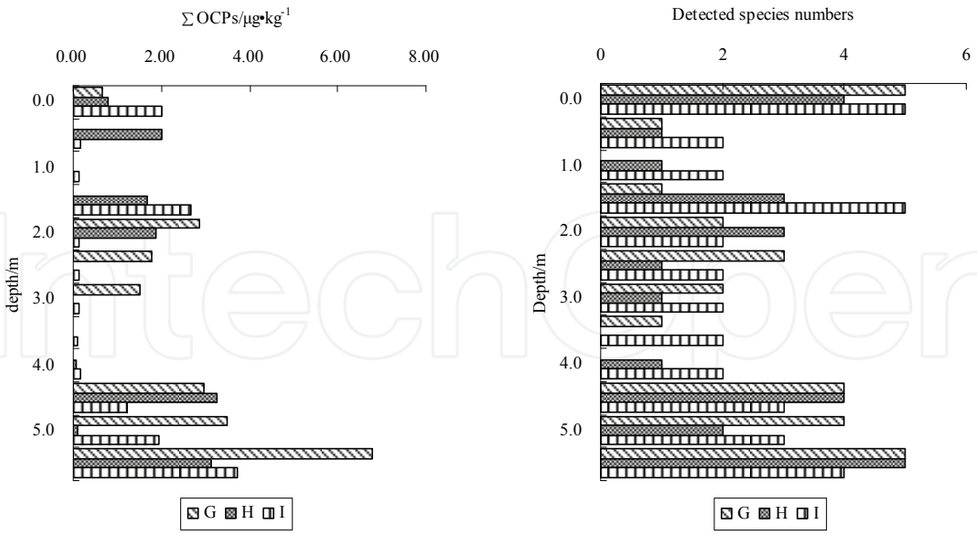


Fig. 13. Vertical distribution of ∑OCPs and detected OCPs species number in GIF

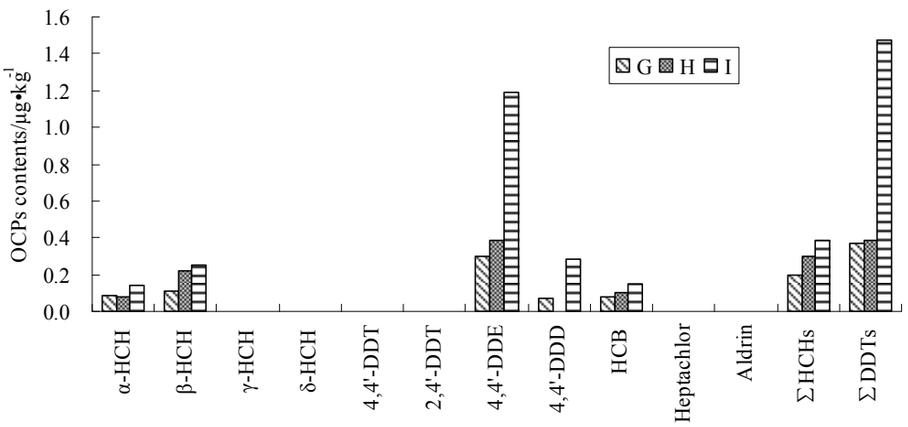


Fig. 14. Distribution of OCPs in surface soil of GIF

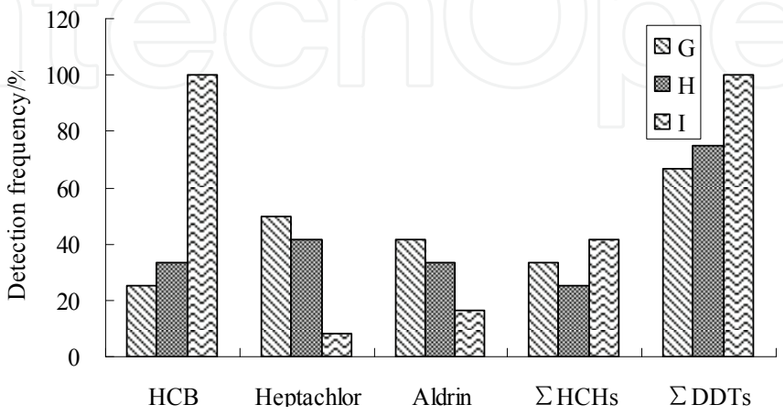


Fig. 15. Detection of frequency of OCPs in GIF profiles

this value was much lower than that of WIF and RIF. The detected OCPs species number's distributions were also different from WIF and RIF. No sharp drop occurred from top layer to sublayers. May be this just was the characters of background.
In figure 14, we can see that DDTs, HCHs and HCB were detected in the surface soil, and 4,4'-DDE was the main composition of DDTs with the maximum value of 1.19 μg/kg and β-HCH was the main composition HCHs with the maximum value of 0.25 μg/kg. However, figure 15 shows that except DDTs, HCHs and HCB, both Heptachlor and Aldrin had been detected in 3 profiles. The sequence of average detection frequency of OCPs was HCB> DDTs> Heptachlor, HCHs> Aldrin.

4.3.2 Vertical variation of single OCP

DDTs, HCHs, HCB, Heptachlor, Aldrin were chosen as the main objects to discuss the vertical distribution of OCP, the variation were shown in figure 16.
As we discussed before, DDTs, Heptachlor and Aldrin had the same distribution characters with WIF and RIF. Compared with DDTs, HCHs have lower Octanol-water partition coefficients, means that HCHs were lower hydrophobic and easier to migrate with infiltration water than DDTs. So, we found more HCHs detected in deep layers. The $\log K_{OW}$ of HCB is 5.73. This value is lower than DDTs' and higher than HCHs'. So HCB was no easy to migrate too. But HCH has highest Henry's Law Constant in 14 kinds OCPs detected in this study, which is 0.0017 atm-m^3/mole. We did not discuss HCB in WIF and RIF, because it was not main OCPs pollutant in that two farmlands. Actually, it was detected in 3 farmlands. The average contents in surface layer of WIF, RIF GIF were 0.49 μg/kg, 0.86 μg/kg, and 0.11 μg/kg respectively. Only because HCB's relative content was higher in GIF, so we discussed here.

4.4 Comparison of OCPs in soil profiles

Firstly, detected OCPs average contents in surface soil of 3 farmlands were compared in figure 17. We can see that there was slightly difference between detected species of OCPs in 3 farmlands. α-HCH, β-HCH, δ-HCH, 4,4'-DDT, 2,4'-DDT, 4,4'-DDE, 4,4'-DDD, HCB and Aldrin were detected in WIF, totally 9 species. α-HCH, β-HCH, δ-HCH, 2,4'-DDT, 4,4'-DDE, 4,4'-DDD, HCB and Heptachlor were detected in RIF, totally 8 species. α-HCH, β-HCH, 4,4'-DDE, 4,4'-DDD and HCB were detected in GIF, only 5 species. We can also see that the ∑OCPs of WIR was much higher than RIF and GIF, the average contents of 3 farmlands were 24.12 μg/kg, 8.64 μg/kg, and 1.15 μg/kg respectively. This indicated the irrigation water sources have great affects on the OCPs pollution in soil, despite that most OCPs came from agricultural pesticides application. Secondly, the average contents of OCPs in sub-layers of 3 farmlands were compared in figure 18. The results of detected species below surface soil of three farmlands showed that Heptachlor and Aldrin were the common detected OCPs, the detection frequency and contents of Heptachlor were much higher than other OCPs, and its downward movement in profiles was the most obvious one of all. The contents of OCPs under surface soil appeared as: GIF >RIF >WIF. That was quite different from surface layer. Since most OCPs were easy to be absorbed in surface layer, though the surface layer suffered different level of pollution, the sub-layers had no distinct difference. To explain this phenomenon, soil texture should be considered too. As we mentioned above, soil profiles textures of these 3 farmlands had no obviously difference. The percentage of clay and silt were more than 24.6% and 63.9% for 3 farmlands. Therefore these kinds of soil textures

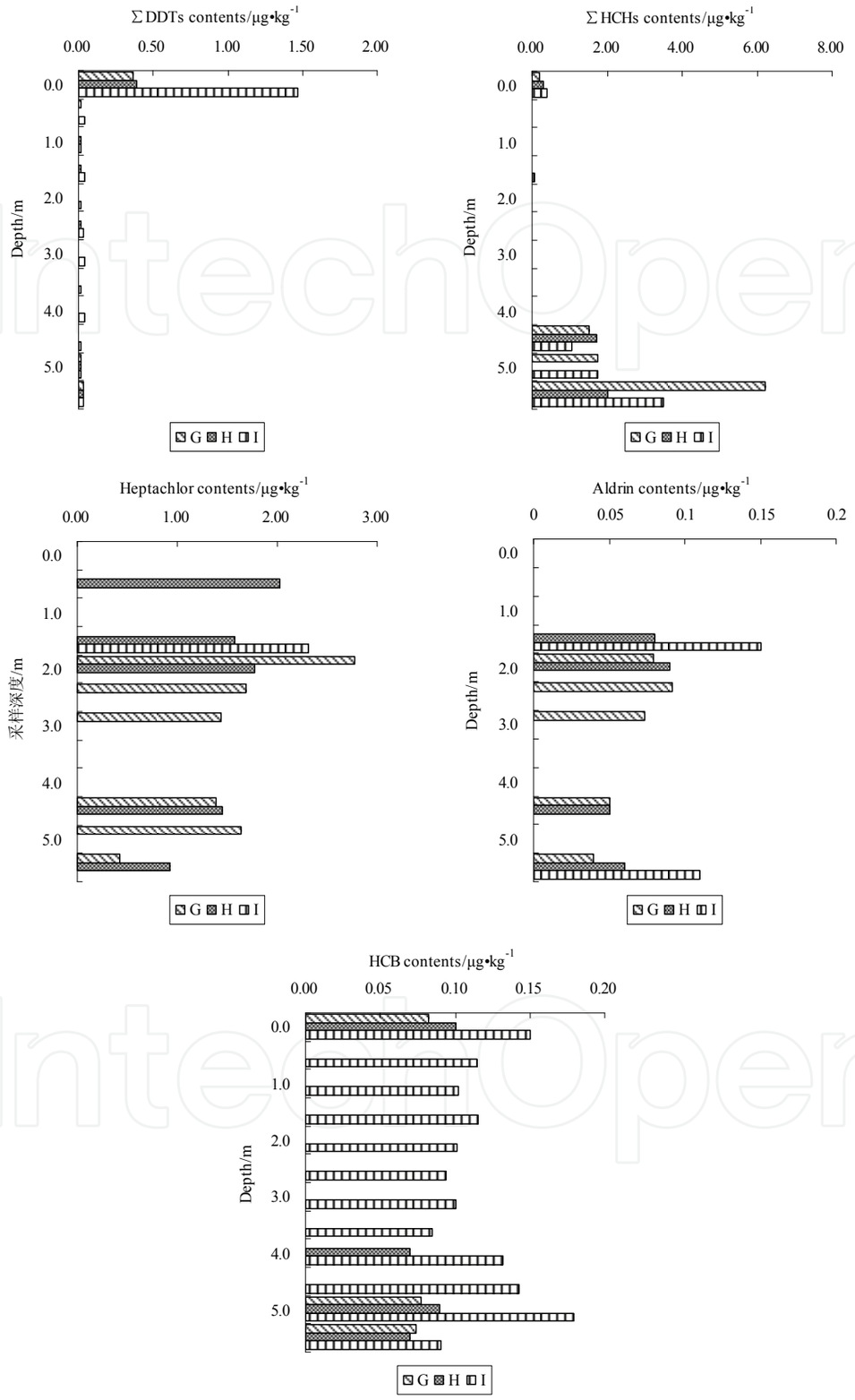


Fig. 16. Vertical distribution of single OCP in GIF

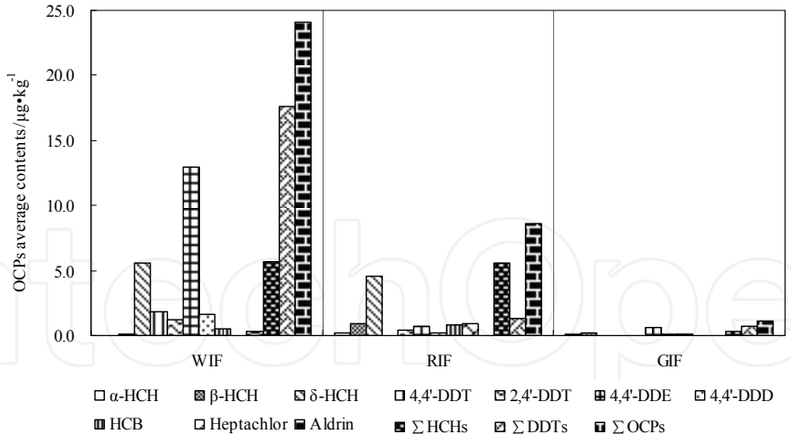


Fig. 17. Comparison of average OCPs contents in surface soil in 3 farmlands

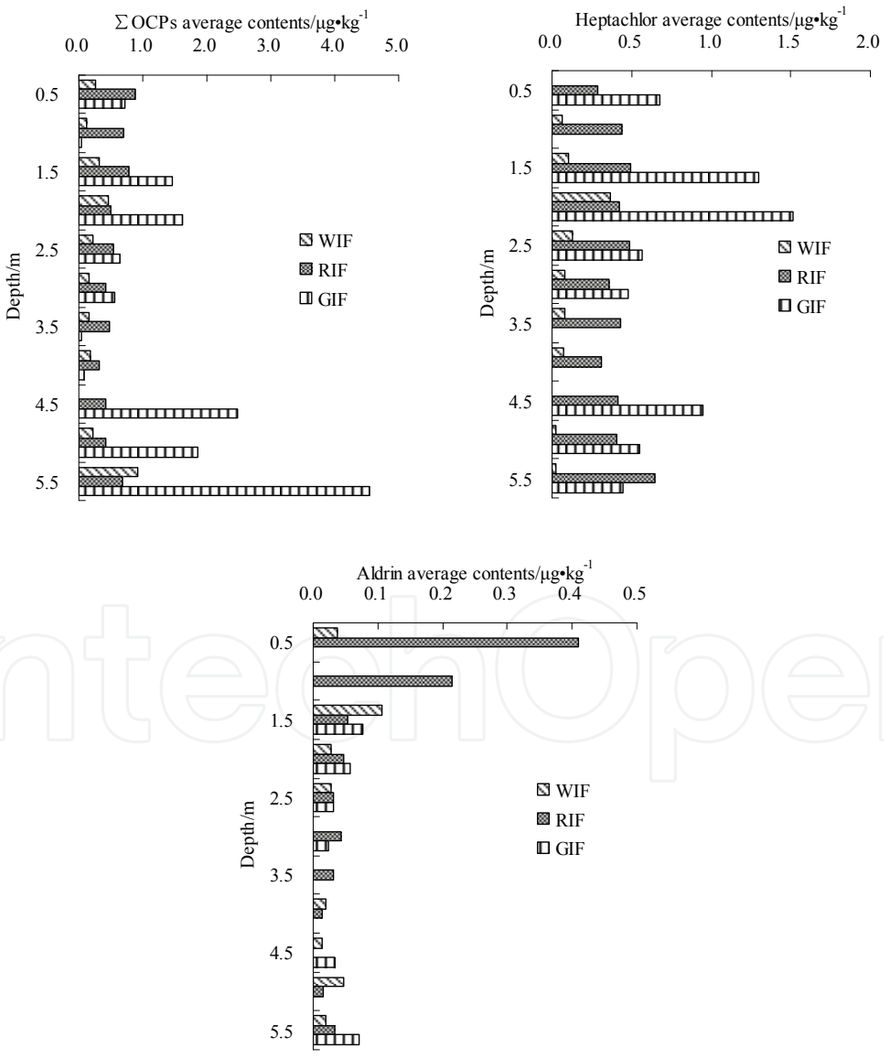


Fig. 18. Comparison of average OCPs contents in sub-layers in 3 farmlands

have strong ability to absorb the OCPs and prevent them from vertical migration. If we must give out the different of the 3 farmlands, we can say, pollutants were a little easier to migrate in RIF and GIF than in WIF, and the vertical changes of soil texture in WIF and RIF were larger than GIF.

On the whole, although none of the detected OCPs' contents in 3 farmlands was over the first class soil standard (<50 μg/kg) of Chinese soil environmental quality standard (GB15618-1995), OCPs had the ability of downward migration in profiles under the long-term irrigation both for wastewater and reclaimed water, and in most conditions, the main accumulation layer of OCPs was the surface soils.

5. OCPs in water samples of three farmlands

5.1 OCPs in water samples of WIF

5.1.1 OCPs in surface water

Table 4 was the concentration of OCPs detected in surface water of WIF. Only 5 species of OCPs including α-HCH, β-HCH, γ-HCH, Heptachlor, and Aldrin were detected in the irrigation water. The detected OCPs also were found in profiles. It's confirmed that irrigation water was a kind of OCPs pollution source, but the contribution degree still under discussion. In addition, DDTs was detected in the soil profiles, but not detected in surface water, indicating there were other sources of pollution, may be related to pesticide application. Other reason for that was pretreatment procedure of water samples before analysis. Since the water samples must be filtered by APFF glass fiber membrane to remove the suspend particles, the OCPs absorbed in the particles were also removed. This part OCPs were not count in the water analysis results. They would go into farmlands accompany with irrigation.

	α-HCH	β-HCH	γ-HCH	Heptachlor	Aldrin
WS-1	＜0.60	1.47	＜0.65	2.50	2.09
WS-2	0.85	3.17	0.82	1.90	1.81
WS-3	0.85	2.76	＜0.65	2.06	2.79

Ps: WS means surface sample of wastewater irrigation farmland

Table 4. Concentration of OCPs in surface water of WIF (ng/L)

5.1.2 OCPs in groundwater

Table 5 shows the results of groundwater detection. β-HCH, Heptachlor, Aldrin and Dieldrin were detected in groundwater. These OCPs also detected in irrigation water and soil profiles except Dieldrin. Since Dieldrin was detected only once in one well, the detection had contingency. The consistency of detected species of OCPs supposed that wastewater irrigation was one of OCPs' pollution sources. These OCPs detected in groundwater were supposed to have relatively higher mobility. In contrast, other OCPs such as DDTs, which had high detection frequency in profiles but undetected in groundwater, were poor in vertical migration and easy to be trapped by soils. Because DDTs were a group of compounds with large molecular, and had strong fat-soluble and poor water-soluble chemical properties.

	β-HCH	Heptachlor	Dieldrin	Aldrin
WG-1	6.08	49.44	< 0.50	1.94
WG-2	2.57	< 0.65	< 0.50	0.96
WG-3	1.27	< 0.65	1.16	2.48
WG-4	1.97	< 0.65	< 0.50	< 0.40
WG-5	2.82	38.96	< 0.50	1.55
WG-6	2.22	< 0.65	< 0.50	0.91
WG-7	2.27	< 0.65	< 0.50	< 0.40

Ps: WG means groundwater of wastewater irrigation farmland

Table 5. Concentrations of OCPs in groundwater of WIF (ng/L)

5.2 OCPs in water samples of RIF

5.2.1 OCPs in surface water

The results of OCPs detected in irrigation water of RIF were listed in table 6. β-HCH, Heptachlor and Aldrin were detected in irrigation water. These three OCPs were also detected in soil profiles. In addition, DDTs, HCB which detected in soil profiles were not detected in surface water. This phenomenon was the same to WIF.

	β-HCH	Heptachlor	Aldrin
RS-1	0.54	1.87	3.40
RS-2	1.41	1.78	5.87
RS-3	1.26	1.80	2.42
RS-4	1.25	1.94	1.96

Ps: RS means surface sample of RIF

Table 6. Concentrations of OCPs in surface water of RIF (ng/L)

5.2.2 OCPs in sewage treatment plants effluent

Reclaimed water of 4 sewage treatment plants were detected several times in this study.

As shown in figure 19, ∑DDTs were detected in all plants with relative high concentration. It was the typical OCPs in wastewater and reclaimed water. The highest concentration of DDTs was appeared in the June from Gaobeidian secondary effluent, which was 97.8 ng/L. The main composition of DDTs was 4,4'-DDE, which accounted for at least 84% of DDTs. 2,4'-DDT was also detected occasionally, but the concentration was very low. Though Heptachlor was only detected in May, its concentration were much higher than other OCPs except in Gaobeidian secondary effluent. ∑HCHs were detected in all plants too, but the concentration were not as high as ∑DDTs, the highest concentration of HCHs was 20.2 ng/L which appeared at Xiaohongmen secondary effluent in May, and 4 kinds of isomers, α-HCH, β-HCH, γ-HCH, δ-HCH were detected occasionally. Despite of the low concentration, HCHs were considered as the typical OCPs of wastewater and reclaimed water for their high detection frequency. Like HCHs, HCB and Aldrin were detected in all plants too, and concentrations were not high. Since they were found in 3 farmlands, the irrigation water should not be excluded from the OCPs sources. Heptachlor epoxide and Dieldrin were only

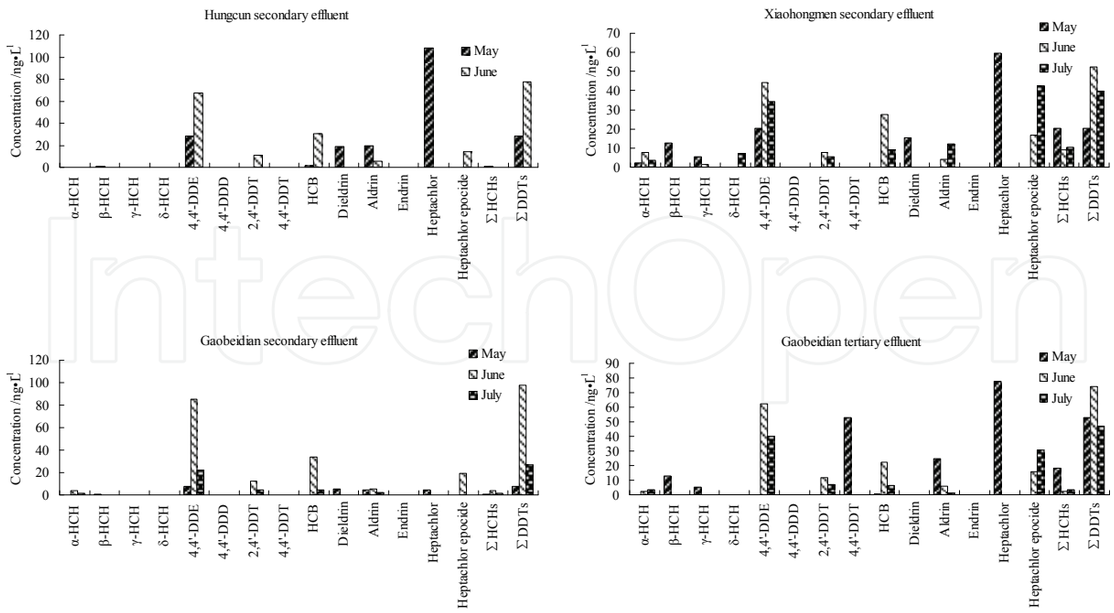


Fig. 19. Concentrations of OCPs in sewage treatment plants

detected occasionally, and were not detected in the 3 farmlands. Endrin was not found both in sewage treatment plants and farmlands.

Through comparing the irrigation water and effluent of sewage treatment plants, we could find that the OCPs detected in irrigation water were all detected in effluent of four plants, but the concentrations were lower than in sewage treatment plants' effluent. Pollutants could be removed effectively through sedimentation and degradation, during the long distance transportation in river channels. And these confirmed that wastewater and reclaimed water were the OCPs pollution sources for farmlands in some degree.

5.2.3 OCPs in groundwater

The concentrations of OCPs in groundwater of RIF were listed in table 7.

	α-HCH	β-HCH	Heptachlor	Aldrin
RG-1	＜0.60	2.71	157.45	2.25
RG-2	2.60	9.63	24.79	1.63
RG-3	＜0.60	<0.40	<0.65	<0.40
RG-4	1.72	12.90	<0.65	<0.40
RG-5	<0.60	<0.40	22.20	<0.40
RG-6	2.09	11.32	22.80	2.02

Ps: RG means groundwater of RIF

Table 7. Concentrations of OCPs in groundwater of RIF (ng/L)

α-HCH, β-HCH, Heptachlor and Aldrin were detected in the groundwater, the other substances were not detected. The difference between soil profiles and groundwater was that DDTs were not detected in groundwater samples, while a higher detection frequency in

HCHs, this meant that HCHs had stronger vertical migration than DDTs. Compared the test results of irrigation water, groundwater and effluent from sewage treatment, common detected pollutants were β-HCH, Heptachlor, and Aldrin.

5.3 OCPs in water samples of GIF

6 wells near the boreholes were selected to gather groundwater samples, but none of 14 kinds of OCPs were detected in them in the GIF. This supported that the OCPs pollution of soil in this farmlands had no relationship with groundwater irrigation. The detection in soil mainly came from pesticide application.

6. The relationship between soil properties and OCPs

The physical and chemical properties of OCPs in soil are mainly controlled by soil characteristics (Cheng H H et al., 1990), so it is important to study the vertical variety of soil properties in profiles. The solubility of OCPs in water is typically under 1 mg·L^{-1}, while the distribution coefficient logK_{OC} is usually over 3, which shows that OCPs have a high fat-soluble and low water solubility, so the TOC will be an important factor in controlling the migration of OCPs. Furthermore, OCPs are mostly weak polar material, according to the rule of similarity, water with strong polar can play a significant inhibitory effect, thus, soil moisture is also the main factor in impacting the migration of OCPs. Montmorillonite is the most content material in soil of various components of clay mineral, it has the feature of double-crystal structure and large CEC, which can enhance the capacity to adsorb organic or inorganic ions. In order to quantitative descriptive the effects of soil properties on the concentration of OCPs, multiple regression analysis was taken in three farmlands, total content of OCPs was the dependent variable and soil properties were the independent variables in stepwise method. The results were in the table 8, 9, 10.

Model Summary				
Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1[a]	0.423	0.179	0.155	6.28979
2[b]	0.693	0.480	0.448	5.08123
3[c]	0.779	0.606	0.569	4.49014

a. Predictors: (Constant), Soil moisture
b. Predictors: (Constant), Soil moisture, TOC
c. Predictors: (Constant), Soil moisture, TOC, EC

Table 8. Multiple regression analysis in WIF

The results in table 8 were used the stepwise removing, we can see the coefficient of determination in model 3 was 0.606 which was relatively high, while the TOC, soil moisture and EC were the independent variables. The regression equation was y=30.541+19.428 x1-0.665x2-0.954x3, where y was the total OCPs, x1 was the TOC, x2 was the soil moisture content, x3 was the EC. These showed that TOC, soil moisture and soluble salt of soil were the impact factors of OCPs in WIF.

In table 9, we can see the coefficient of determination in model 3 was 0.575. The TOC, soil moisture and Eh were the independent variables. The regression equation was

y=9.334+4.812x1-0.196x2-0.012x3, where y was the total OCPs, x1 was the TOC, x2 was the soil moisture content, x3 was the Eh. Showed that TOC and soil moisture of soil were the common impact factors in wastewater and RIF.

Model Summary				
Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1[a]	0.540	0.292	0.271	2.00184
2[b]	0.717	0.514	0.484	1.68367
3[C]	0.759	0.575	0.536	1.59737

a. Predictors: (Constant), TOC
b. Predictors: (Constant), TOC, Soil moisture
c .Predictors: (Constant), TOC, Soil moisture, Eh

Table 9. Multiple regression analysis in RIF

Model Summary				
Model	R	R Square	Adjusted R Square	Std. Error of the Estimate
1	0.583	0.340	0.321	1.27109

Predictors: (Constant), Eh

Table 10. Multiple regression analysis in GIF

In table 10, the coefficient of determination in model 1 was 0.340, while the Eh was the independent variable and the regression equation was y=4.521-0.008x, where y was the total OCPs, x was the Eh. These showed that Eh was the common factor in reclaimed water and GIF.

7. Source analyses of OCPs in three farmlands

Sources of pesticide in soil are mainly the following aspects: using for preventing pest; the input of irrigation water and the deposition of atmospheric particles. In recent years, due to rapid growth of food demand, pesticide application in most cases are excessive. Besides, solid wastes are piled up and dumped to the soil surface continually, thus, hazardous wastewater is continued to infiltrate into the soil, more and more hazardous gases and particulates landed into the soil with rain. When the content of harmful substances in soil exceed the soil's self-purification ability, the composition, structure and function of soil will be changed, microbial activity will be inhibited, and harmful substances or its decomposition products will be accumulated in soil gradually, finally absorbed by human body, when the extent great enough to threaten human health, the soil pollution is formed.

Commercial HCHs is a mixture of several closely–related compounds. The common components were 55%-80% of α-HCH, 5%-14% of β-HCH, 12%-14% of γ-HCH, 2%-10% of δ-HCH and 3%-5% other organochlorine pesticides. The dechlorination rate sequence in HCHs is: α-HCH> γ-HCH>δ-HCH> β-HCH, in which the α-HCH is the most unstable and

has the fastest degradation rate; γ-HCH in agricultural soils can be easily changed into other HCHs by decomposition or biotransformation; β-HCH is a stable HCHs with the lowest solubility and difficult to evaporate(Chen L G et al., 2005; Rekha P N et al., 2004). In the three farmlands, δ-HCH had the highest detected concentration in all soil profiles and the γ-HCH had the lowest, one reason was the migration from surface soil where had the high concentration of δ-HCH, the other reason was the high solubility of δ-HCH(Prakash O et al., 2004), the undetected γ-HCH in profiles showed that there was no new input in recently, so the HCHs residues in soil was a legacy problem which caused by history wastewater irrigation and pesticide application.

Commercial DDTs is a mixture too. The major components include 4,4'-DDT (75%), 2,4'-DDT (15%), 4,4'-DDE (5%), 4,4'-DDD (<5%) and other substances, DDT in anaerobic condition can be degraded to DDD compounds, while degraded to DDE under aerobic conditions, the property of DDE is more stable and its degradation rate is proportional with the increasing of soil moisture, temperature and microbial activity(Hitch R K et al., 1992). In three farmlands, the content of 4,4'-DDE was the highest in DDTs in soil profiles, 4,4'-DDD was the lowest, because the 4,4'-DDE was harder to decompose than 4,4 ' -DDT and 4,4'-DDD, 4,4'-DDE had the highest percentage in DDTs indicating that most residual DDT in soil was the degradation products, which consistent with the degradation in environment after DDTs been banned, also consistent with the Eh test results (soil in three farmlands were the oxidation state). DDT in soil environment undergoes a long period of physical, chemical and biological change, the (DDE + DDD) / DDT ratio should be greater than 1, if the ratio is less than 1, indicating that new sources may input (Qiu X et al., 2004; Jaga K et al., 2003). The ratio in WIF was 5.33 and 2.25 in RIF, DDT was not detected in GIF, all these values were greater than 1, indicating that no new sources input recently.

8. Conclusions

From the analyses and studies of irrigation water, pollutants contents in soil profiles and the physical-chemical properties of soils, combined with statistical analysis software to analyze the relationship between contents of OCPs and the physical-chemical parameters of soil, conclusions are as follows:

The variations of soil physical-chemical indexes with depth in three farmlands were nearly consistent. The main soil particles were silt, followed by clay, and sand. Lithology changes in WIF and RIF were larger than in GIF. The proportion of silt and clay in RIF were less than in WIF, while the sand was greater than in WIF.

Surface soil was the accumulated layer of OCPs in WIF and RIF, both ∑OCPs and single OCP overall decreased with depth, but there was no this phenomenon in GIF. The average contents of ∑OCPs in surface soils of 3 farmlands were 24.11 μg/kg, 8.64 μg/kg, and 1.15μg/kg respectively. Clearly the sequence of total OCPs in soils was: WIF> RIF> GIF, which supposed that the content of OCPs had a great relationship with the quality of irrigation water.

OCPs detected in three farmlands were basically the same, and surface water and groundwater were also in good agreement. DDTs and HCHs were the main OCPs in surface soils which accounted for at least 80% of ∑OCPs. However, Heptachlor and Aldrin were common OCPs under the surface in profiles. This phenomenon supposed that Heptachlor and Aldrin were easier to migrate than DDTs and HCHs in soils profiles, and these two substances were a major threat to quality of groundwater.

DDTs and HCHs were not detected in irrigation water and groundwater, maybe caused by water sample pretreatment of filtration. OCPs detected in effluent of sewage plants confirmed that DDTs and HCHs were the main OCPs in wastewater and reclaimed water. But most OCPs could be removed effectively through sedimentation and degradation, during the long distance transportation in river channels.

Heptachlor and Aldrin were detected both in irrigation water and groundwater except groundwater of GIF. That means except for pesticides application, wastewater and reclaimed water were other important sources for OCPs in farmlands.

Using SPSS to analyze the soil properties and total OCPs, the results were that, TOC and soil moisture were the main impact factors of vertical distributions of OCPs in WIF and RIF, while Eh was the important factor in GIF.

Source analyses supposed that, there were no new application of DDTs and HCHs in 3 farmlands. The accumulations of DDTs and HCHs in surface soils were due to historical pesticides application and wastewater irrigation.

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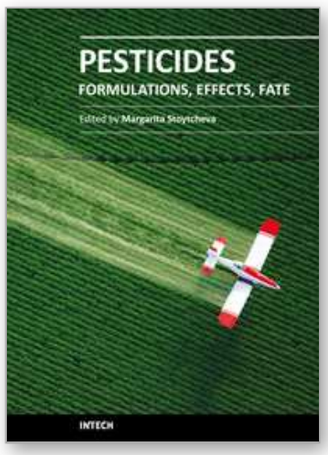
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This book provides an overview on a large variety of pesticide-related topics, organized in three sections. The first part is dedicated to the "safer" pesticides derived from natural materials, the design and the optimization of pesticides formulations, and the techniques for pesticides application. The second part is intended to demonstrate the agricultural products, environmental and biota pesticides contamination and the impacts of the pesticides presence on the ecosystems. The third part presents current investigations of the naturally occurring pesticides degradation phenomena, the environmental effects of the break down products, and different approaches to pesticides residues treatment. Written by leading experts in their respective areas, the book is highly recommended to the professionals, interested in pesticides issues.

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