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Multi-Media Exposure, Selective Accumulation and Health Risks of Organochlorine Pesticides in Human Serum under Post-Ban Conditions

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Abstract: Organochlorine pesticides (OCPs), despite being banned, persist in the environment and continue to pose potential risks to human health. This exploratory study investigated the concentrations of multiple OCPs in serum samples from 10 local residents and 52 surrounding environmental samples, including soil ($n = 10$), air ($n = 12$), indoor dust ($n = 10$), and food ($n = 20$), collected in Taizhou, Zhejiang Province, in 2022. Multiple exposure pathways were quantified, and associated health risks were assessed. The results showed evidence of selective accumulation in humans, with serum Σ OCP concentrations ranging from 107.01 to 552.41 ng/g lipid weight (lw). p,p' -DDE was the predominant compound, accounting for 67.81% of total serum OCPs, while β -HCH showed relatively higher enrichment among HCH congeners. Lower proportions of p,p' -DDT and α - γ -HCH were observed. Source contributions varied across media: food was the primary contributor to p,p' -DDE exposure, indoor dust and soil contributed more to β -HCH and certain DDT isomers, and the atmosphere showed a higher contribution of methoxychlor. Differences between integrated exposure profiles and serum composition highlighted the potential roles of metabolic transformation and lipid-associated accumulation in shaping internal OCP burdens. Risk assessment indicated that DDT metabolites and β -HCH were the major contributors to potential exposure risks. Overall, this study provides preliminary insights into the environmental–human distribution patterns of OCPs observed in Taizhou under post-ban conditions and highlights that environmental concentrations alone may not fully represent chronic internal exposure. These findings improve the understanding



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of potential OCP exposure pathways and emphasize the need for larger-scale studies incorporating population characteristics and refined exposure parameters for more comprehensive risk assessment.

Keywords: organochlorine pesticides; serum; multi-media exposure; health risk

1. Introduction

Organochlorine pesticides (OCPs) are a class of synthetic chlorinated compounds that have been widely used in agriculture and public health applications owing to their high insecticidal efficacy and environmental persistence [1]. Since the mid-20th century, OCPs have been widely used worldwide for agricultural production, vector control, and public health purposes. However, these compounds are highly persistent in the environment, can undergo long-range transport, and tend to bioaccumulate. Several OCPs have been classified by the International Agency for Research on Cancer (IARC) as confirmed or potential human carcinogens [2]. Chronic or high-level exposure to OCPs has been linked to various adverse health outcomes, including neurotoxic, endocrine, and immune dysfunctions, as well as hepatic and renal impairments [3]. China was historically among the world's largest producers and consumers of OCPs, with cumulative production estimated at approximately 460,000 tons of dichlorodiphenyltrichloroethane (DDT) and 4 million tons of hexachlorocyclohexanes (HCHs) [4]. Despite being listed under the Stockholm Convention on Persistent Organic Pollutants in 2001 for global restriction and elimination, OCPs remain ubiquitous in environmental media and continue to threaten ecosystem integrity and human health [5].

As legacy pesticides, organochlorine pesticides (OCPs) remain widely distributed across multiple environmental media in China. Hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs) are commonly detected in soils nationwide, with concentrations in some regions exceeding relevant soil environmental quality standards [6]. HCHs, DDTs, and their metabolites (DDE and DDD) are continuously detected in the atmosphere, with HCHs generally dominating and DDE accounting for a relatively high proportion of DDT-related compounds. This pattern indicates that atmospheric OCPs primarily originate from the re-volatilization of historical residues and can be transported over long distances, as evidenced by detections in remote areas such as the Tibetan Plateau [7,8]. OCPs are also commonly detected in indoor dust in urban environments, representing an important exposure pathway via inhalation, ingestion, and dermal contact [9]. Furthermore, OCPs are frequently detected in food items, including cereals, vegetables, and animal-derived products, with higher concentrations in lipid-rich animal tissues, suggesting ongoing contributions from soil residues and biomagnification along the food chain to human exposure [10]. Overall, despite regulatory bans and restrictions, OCPs remain widespread in the environment and continue to pose long-term exposure risks through food-chain transfer and environmental accumulation [11]. However, current studies remain largely limited to single environmental compartments or isolated exposure pathways, while integrated assessments linking multimedia occurrence to human exposure, as well as the mechanisms underlying selective enrichment, are still insufficiently explored.

Multiple studies have demonstrated substantial differences among various types of organochlorine pesticides (OCPs) in terms of environmental residual levels, human exposure pathways, and toxicological effects. Hexachlorocyclohexanes (HCHs) are characterized by relatively high volatility and strong long-range transport potential, leading to their higher proportions in the atmosphere and remote background regions [12]. In contrast, dichlorodiphenyltrichloroethanes (DDTs) and their metabolites exhibit stronger lipophilicity and bioaccumulation capacity, facilitating biomagnification along food chains and resulting in higher detection frequencies in human adipose tissue and serum [13]. Due to differences in toxicological thresholds, carcinogenic potential, and endocrine-disrupting activity among OCP compounds, their associated health risks are highly heterogeneous [14]. For instance, certain DDT metabolites have been reported to exhibit strong endocrine-disrupting effects [15], whereas β -HCH is more prone to accumulation in the human body due to its chemical stability and prolonged biological half-life [16]. Therefore, risk assessments based solely on environmental concentrations may underestimate or overestimate the actual health impacts of specific compounds. Integrated multi-media exposure quantification combined with internal human biomonitoring data is thus essential for identifying key contributing pollutants and dominant exposure pathways.

This study selected Taizhou, Zhejiang Province, a historically OCP-contaminated area, as the study site. Taizhou experienced extensive application of OCPs before their prohibition, and previous studies have reported persistent OCP residues in local environmental compartments. However, previous investigations have mainly focused on individual environmental matrices, leaving the linkage between multi-media exposure pathways and human internal accumulation unclear. To address this knowledge gap, soil, air, indoor dust, and food samples were

collected simultaneously, and multiple OCPs were measured in residents' serum. A multi-media, multi-pathway exposure framework was established to quantify exposure contributions and evaluate compound-specific health risks. By integrating environmental measurements with human biomonitoring, this study aimed to characterize OCP distribution across environmental media and serum, identify key exposure pathways and priority contaminants, and improve the understanding of human exposure to legacy OCPs under long-term ban conditions.

2. Materials and Methods

2.1. Study Area and Sample Collection

Taizhou, located in Zhejiang Province, eastern China, was selected as the study area. This region has experienced intensive agricultural activities and historical application of organochlorine pesticides (OCPs) before their prohibition. According to satellite-derived land use and land cover (LULC) information from 2022, the study area is characterized by a mixture of urban areas, agricultural land, forests, and water bodies, reflecting diverse human activities and environmental conditions that may influence OCP distribution and exposure pathways. In 2022, serum samples were collected from 10 volunteers in Study area. Considering potential individual factors such as sex, age, and body weight, detailed demographic information for all participants is provided in Table S1. All serum samples were stored at -20°C prior to analysis. Corresponding indoor dust samples ($n = 10$) were collected from the participants' residences, and food samples were collected over two consecutive days ($n = 20$). Additionally, environmental samples, including surrounding soil ($n = 10$) and atmospheric samples ($n = 12$, collected from three sites over four consecutive days), were obtained from the participants' residential areas. In total, 62 samples were collected. Detailed sampling locations for soil and air samples are shown in Figure 1.

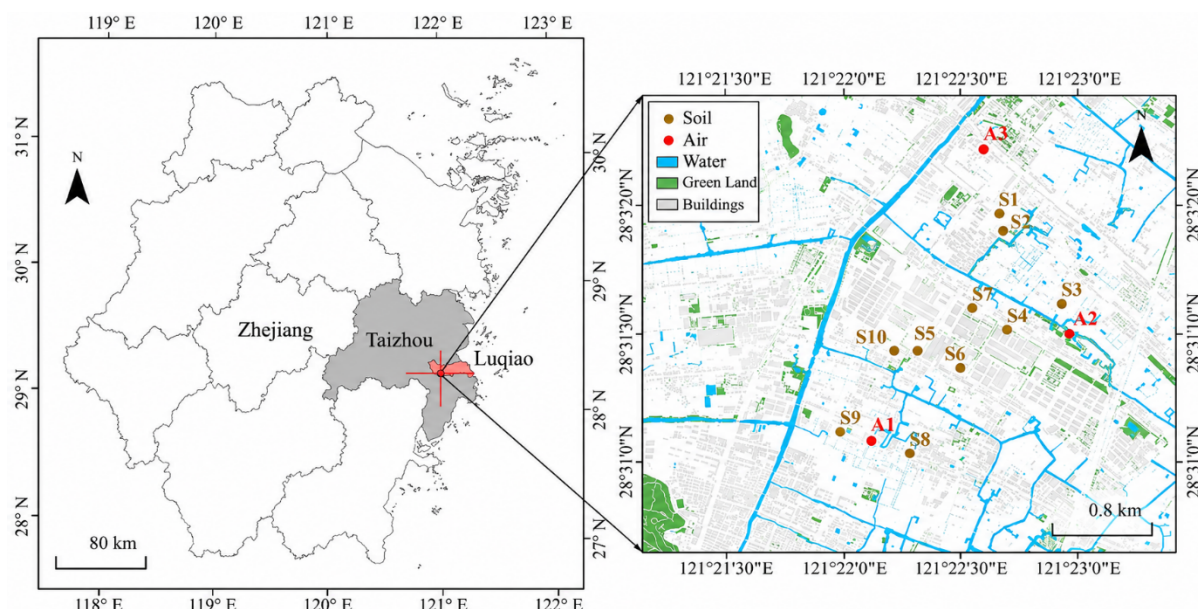


Figure 1. Distribution map of soil and air sampling sites. S1–S10 represent the soil sampling sites, and A1–A3 represent the air sampling sites.

2.2. Materials and Reagents

Major instruments and materials used in this study included deionized and ultrapure water produced by a Milli-Q water purification system (Millipore, Burlington, MA, USA). Pipettes were supplied by Eppendorf (Hamburg, Germany). Sample processing and preparation employed a TDL-40B centrifuge (Shanghai Anting Scientific Instrument Factory, Shanghai, China) and a BF2000 nitrogen evaporation system (Beijing Bafang Century Technology Co., Beijing, China). Analytical procedures utilized a Shimadzu precision electronic balance (Kyoto, Japan) and an N-1300D-WB rotary evaporator (EYELA, Tokyo, Japan). Gas chromatographic separation was performed on a J&W DB-5-MS capillary column ($30\text{ m} \times 0.25\text{ mm} \times 0.25\text{ }\mu\text{m}$; Agilent Technologies, Santa Clara, CA, USA), and target compounds were analyzed using a Trace 1310-TSQ 8000 GC–MS system (Thermo Fisher Scientific, Waltham, MA, USA). For quality control, ^{13}C -Endosulfan I (Cambridge Isotope Laboratories, Tewksbury, MA, USA) was used as an internal standard, while α -HCH, β -HCH, γ -HCH, δ -HCH, o,p'-DDD, p,p'-DDD, o,p'-DDE, p,p'-DDE, p,p'-DDT and Methoxychlor (AccuStandard, New Haven, CT, USA) served as

external standards. All solvents, including dichloromethane, n-hexane, and methanol (chromatographic grade), were purchased from J.T. Baker (Phillipsburg, NJ, USA).

2.3. Sample Preparation

Serum samples were thawed at 4 °C one day prior to extraction. A 2 mL aliquot of serum was transferred into a 15 mL centrifuge tube. Then, 40 µL of 50 ppb ¹³C-labeled OCP internal standards was added. Procedural blanks were prepared using ultrapure water in place of serum; one blank was spiked with internal standards, while another was unspiked.

Each sample was acidified with 1 mL of 6 mol/L HCl and vortexed, followed by extraction with 3 mL isopropanol. This extraction step was repeated once. Subsequently, 3 mL of n-hexane/methyl tert-butyl ether (1:1, v/v) was added for liquid–liquid extraction, and the partitioning step was repeated once. Samples were left to stand overnight and then centrifuged at 2000 rpm for 5 min. The upper organic phase was collected, and the lower phase was re-extracted twice using the same solvent system.

The combined extracts were concentrated to approximately 7 mL under a gentle stream of nitrogen. To remove polar interferences, 4 mL of 1% KCl aqueous solution was added, followed by overnight equilibration and centrifugation (2000 rpm, 5 min). The organic phase was collected, and lipid content was determined gravimetrically after near-dry nitrogen evaporation.

Lipid removal was performed using 4 mL n-hexane and 2 mL 0.5 mol/L KOH in 50% ethanol, followed by repeated extraction and centrifugation. The cleaned extracts were concentrated to 1 mL and purified by gel permeation chromatography (GPC). The eluate was further concentrated by rotary evaporation and subjected to acid silica gel column cleanup (cotton, neutral silica gel, acid silica gel, and anhydrous Na₂SO₄).

Finally, the purified extract was reduced under nitrogen and reconstituted in 100 µL n-nonane for GC–MS analysis.

2.4. Instrumental Analysis

Pretreated samples were analyzed using an Agilent 6890 gas chromatograph coupled with an Agilent 5975 mass spectrometer (Agilent Technologies, Santa Clara, CA, USA). The mass spectrometer operated in negative chemical ionization (NCI) mode with selected ion monitoring (SIM). Helium was used as the carrier gas at a flow rate of 1 mL/min, and methane served as the reagent gas at 1 mL/min. The temperatures of the injector, mass spectrometer source, and quadrupole were set to 250 °C, 150 °C, and 150 °C, respectively. Chromatographic separation was performed on a J&W DB-5-MS column (30 m × 0.25 mm × 0.25 µm; Agilent Technologies). The oven temperature program was set as follows: initial temperature 80 °C, held for 3 min, then increased at 6 °C/min to 260 °C. Quantification was performed using the following m/z ion pairs: 406/408 for ¹²C-Endosulfan I, 406/408 for ¹³C-Endosulfan II, and 453/455 for ¹³C-Endosulfan (internal standard).

2.5. Quality Control and Assurance

Target compounds were quantified using an isotope dilution method with ¹³C-labeled internal standards. Limits of quantification (LOQs) were defined as the concentrations corresponding to a signal-to-noise ratio (S/N) of 10. All calibration curves showed excellent linearity, with correlation coefficients (R²) greater than 0.999. The recovery of ¹³C-Endosulfan was 107.0% ± 24.8%, and the LOQ was 0.08 ng/g lipid weight (lw). Method blanks were included in each analytical batch. Concentrations of target analytes in blanks were low (generally <10% of those in samples) and the corresponding S/N values were <10; therefore, no blank correction was applied to the sample data. Mean and median concentrations reported in this study were calculated based on the individual concentrations measured in all samples collected for each matrix.

2.6. Data Analysis

All statistical analyses were performed using SPSS 22.0 software (IBM, Armonk, NY, USA). Values below the limit of quantification (LOQ) were assigned a value equal to half the LOQ. Differences in concentrations between two groups were assessed using independent-samples *t*-tests. Statistical significance was defined at *p* < 0.05.

2.7. Exposure Assessment

Endosulfan present in environmental media represents a potential source of human exposure, entering the body via three main pathways: oral ingestion, inhalation, and dermal contact [17–19]. The exposure assessment models for different pathways are detailed in Table 1, and the parameters used in these models are listed in Table 2.

Table 1. Exposure Assessment Models for Different Pathways.

Environmental Media	Pathway	Model	Reference	Equation No.
Soil	Intake Dose (S_{intake})	$S_{intake} = S_c \times S_{IRS} \times T_{out}/BW$	[20,21]	1
	Inhalation Dose (S_{inhale})	$S_{inhale} = S_c \times S_{IR} \times \frac{1}{PEF} \times T_{out}/BW$	[20,21]	2
	Dermal Absorbed Dose ($S_{dermal\ contact}$)	$S_{dermal\ contact} = S_c \times S_{SA} \times S_{SAF} \times S_{DAF} \times T_{out}/BW$	[20,21]	3
	Daily Total Exposure Dose (S_{Total})	$S_{Total} = S_{intake} + S_{inhale} + S_{dermal\ contact}$	[20,21]	4
Air	Daily Total Exposure Dose (A_{Total})	$A_{Total} = A_c \times A_{IR}/BW$	[20,21]	5
Dust	Intake Dose (D_{intake})	$D_{intake} = D_c \times D_{IRS} \times T_{in}/BW$	[20,21]	6
	Inhalation Dose (D_{inhale})	$D_{inhale} = D_c \times D_{IR} \times T_{in}/BW$	[20,21]	7
	Dermal Absorbed Dose ($D_{dermal\ contact}$)	$D_{dermal\ contact} = D_c \times D_{SA} \times D_{SAF} \times D_{DAF} \times T_{in}/BW$	[20,21]	8
	Daily Total Exposure Dose (D_{Total})	$D_{Total} = D_{intake} + D_{inhale} + D_{dermal\ contact}$	[20,21]	9
Food	Daily Total Exposure Dose (F_{Total})	$F_{Total} = F_c \times F_{IRS}/BW$	[20,21]	10
-	Hazard Quotient (HQ)	$HQ_i = E_{Total}/RfD$	[20,21]	11

Table 2. Parameters Used in the Exposure Assessment Models.

Equation No.	Abbreviations	Name	Reference Value	Source
1	S_{IRS}	Daily Soil Ingestion Rate	0.05 g/day	[22]
1	T_{out}	Fraction of Daily Time Spent by the Population in Outdoor Soil Contact	0.140	
2	S_{IR}	Soil Environment Air Inhalation Rate	16.1 m ³ /day	
3	S_{SA}	Dermal Surface Area in Contact with Soil Contaminants	1 m ²	
2	PEF	Release Factor of Soil Contaminants	1.36×10^9 m ³ /kg	[17]
3	S_{SAF}	Soil-to-Skin Adhesion Factor	0.0096 g/m ²	[23,24]
3	S_{DAF}	Dermal Absorption Efficiency of Soil Contaminants	0.03	[23]
	S_c			-
5	A_{IR}	Inhalation Rate	16.1 m ³ /day	[22]
5	A_c			-
6	D_{IRS}	Daily Dust Intake	0.05 g/day	[22]
	T_{in}	Proportion of Time Spent in Indoor Dust Exposure During a Day	0.830	-
7	D_{IR}	Inhalation Rate of Airborne Dust	16.1 m ³ /day	-
8	D_{SA}	Dermal Surface Area Exposed to Dust	1 m ² /day	-
8	D_{SAF}	Dust-to-Skin Adhesion Factor	0.096 mg/cm ²	[23,24]
8	D_{DAF}	Dermal Absorption Efficiency of Dust-bound Contaminants	0.03	[23]
	D_c	Concentration of Contaminants in Dust	-	-
10	F_{IRS}	Daily Food Intake	1056.6 g/day	[22]
10	F_c	Concentration of Contaminants in Food	-	-
	BW	Body Weight of Population Groups	-	-
11	E_{Total}	total exposure dose calculated from different exposure pathways	ng/(kg bw day)	

3. Result and Discussion

3.1. Concentration Levels of Organochlorine Pesticides (OCPs) in Human Serum from Taizhou, China in 2022

In this study, ten organochlorine pesticides (OCPs) were quantified in serum samples collected from adults in Taizhou, China, in 2022, revealing significant inter-compound differences in concentrations (Table 3). The mean concentration of Σ OCPs in serum was 242.49 ± 128.63 ng/g lw, with a median concentration of 196.71 ng/g

lw, indicating considerable variability in serum OCP burdens among participants. DDE-related metabolites dominated the OCP profile, with concentrations substantially exceeding those of the parent compound DDT, while β -HCH was the predominant HCH isomer. This compositional pattern is consistent with previous biomonitoring studies worldwide, which have consistently shown that DDE is the dominant DDT-related compound in human serum [25,26]. This is primarily attributed to the preferential biotransformation of DDT to DDE and the longer biological half-life of DDE, leading to its greater accumulation relative to the parent compound [27]. The predominance of p,p'-DDE in serum also reflects the metabolic fate of DDT after human exposure. Following absorption, p,p'-DDT can undergo enzymatic dehydrochlorination mediated by microsomal enzymes, resulting in the formation of p,p'-DDE. Compared with the parent compound, p,p'-DDE exhibits higher environmental persistence and a longer biological half-life, leading to preferential accumulation in lipid-rich tissues. Therefore, the elevated p,p'-DDE proportion observed in serum likely represents both historical exposure and selective retention of stable DDT metabolites. For instance, studies in Chinese populations have consistently reported higher concentrations of p,p'-DDE than p,p'-DDT [28], and similar patterns have also been documented in the U.S. National Health and Nutrition Examination Survey (NHANES) [29]. Among HCH isomers, β -HCH generally dominates due to its higher lipophilicity and longer biological half-life relative to α -HCH and γ -HCH [30]. This pattern is consistent with the present study and aligns with observations reported in Shanghai populations [29]. Among HCH isomers, β -HCH generally dominates due to its stronger lipophilicity and longer persistence in the human body compared with α -HCH and γ -HCH [30], which is consistent with the results of this study and comparable to observations reported in Shanghai populations [31]. Overall, these findings indicate that OCPs remain prevalent in human serum in Taizhou, with DDE, methoxychlor, and β -HCH as the major contributors, highlighting persistent human exposure and accumulation. These results underscore the need for continued monitoring and refined exposure assessment to better evaluate potential public health risks.

Table 3. Concentrations of Organochlorine Pesticides (OCPs) in Human Serum (ng/g lw).

Category	Concentration Range	Mean Concentration $\pm$ Standard Deviation (SD)	Median Concentration
α -HCH	n.d.–10.80	4.30 $\pm$ 3.85	3.63
β -HCH	3.63–52.82	18.43 $\pm$ 15.28	16.76
γ -HCH	0.22–51.96	8.89 $\pm$ 15.50	4.07
Δ -HCH	n.d.–7.44	1.84 $\pm$ 2.60	0.75
o,p'-DDE	n.d.–1.01	0.27 $\pm$ 0.31	0.18
p,p'-DDE	84.49–349.25	164.44 $\pm$ 79.66	130.58
o,p'-DDD	0.12–1.29	0.46 $\pm$ 0.35	0.36
p,p'-DDD	0.40–2.81	1.31 $\pm$ 0.75	1.17
p,p'-DDT	n.d.–21.50	7.31 $\pm$ 7.26	4.48
Methoxychlor	3.10–85.11	35.24 $\pm$ 27.62	34.75
Total	107.01–552.41	242.49 $\pm$ 128.63	196.71

3.2. Concentrations of Organochlorine Pesticides (OCPs) in Different Environmental Media and Food

3.2.1. In Soil

In this study, ten organochlorine pesticides (OCPs) were quantitatively analyzed in representative soil samples from Taizhou (Table S2). The results indicated that the target compounds were generally present at low concentration but remained detectable. The mean concentration of Σ OCPs was 7.22 ng/g, with a median of 3.27 ng/g. Median values showed that p,p'-DDE and p,p'-DDT were the dominant compounds in most soil samples, whereas HCH congeners occurred at relatively low levels. This compositional pattern reflects the historical residues and degradation behavior of OCPs in the study area soils. The higher abundance of p,p'-DDE relative to its parent compound p,p'-DDT can be attributed to biotic and abiotic transformation processes in soil, leading to the more persistent accumulation of DDE [27]. Soil environments provide diverse redox conditions that regulate DDT transformation pathways. Under aerobic conditions, p,p'-DDT can undergo dehydrochlorination reactions, producing p,p'-DDE through the elimination of hydrogen chloride. In contrast, anaerobic environments favor reductive dechlorination, in which chlorine atoms are sequentially removed from DDT molecules, resulting in the formation of p,p'-DDD. These transformation pathways are consistent with the anaerobic/aerobic DDT degradation pathways reported in the University of Minnesota Biocatalysis/Biodegradation Database. Therefore, the higher proportion of DDE than DDT observed in Taizhou soils suggests that long-term aerobic transformation has contributed substantially to DDT attenuation, whereas the relatively lower abundance of DDD indicates limited anaerobic degradation under current soil conditions. Similar compositional patterns have been observed in other

regional soil studies. In agricultural soils across China, DDT-related compounds are typically dominated by p,p'-DDE, with p,p'-DDE/p,p'-DDT ratios generally exceeding 1, reflecting the preferential degradation of DDT to the more persistent DDE [32]. In contrast, HCH congeners exhibited relatively low background levels in this study, suggesting that the historical use of technical HCH occurred earlier and that environmental residues have largely entered a degradation phase. This pattern is consistent with previous soil monitoring studies, where β -HCH, owing to its greater stability and slower degradation rate, tends to persist at higher levels than γ -HCH [30]. Compared with soils from regions with intensive agricultural use abroad, HCH concentrations in this study were substantially lower, although they remained detectable in the soil environment.

3.2.2. In Air

In this study, the concentrations of ten organochlorine pesticides (OCPs) were determined in atmospheric samples from Taizhou (Table S3). All target compounds were detectable, with a mean Σ OCPs concentration of 123.04 ng/m³ and a median of 95.72 ng/m³. DDT and its metabolites were the dominant pollutants in the atmosphere, whereas HCH congeners occurred at comparatively lower levels. This compositional pattern suggests that DDT-related compounds contribute more substantially to atmospheric OCP burdens in Taizhou than HCHs. Historically, DDTs were widely used and are characterized by semi-volatility and environmental persistence, allowing both parent compounds and metabolites to persist in the atmosphere via re-volatilization and long-range atmospheric transport processes [32]. The atmospheric occurrence of DDT metabolites is controlled not only by their historical residues but also by physicochemical properties. Compared with p,p'-DDT, degradation products such as DDE and DDD have lower molecular weights and fewer chlorine substitutions, which may increase their vapor pressures and facilitate atmospheric redistribution. However, their strong hydrophobicity promotes association with atmospheric particles, allowing subsequent deposition to terrestrial and aquatic environments. Although β -HCH tends to accumulate more readily in environmental media due to its stability, its relatively low abundance in the atmosphere suggests that historical HCH inputs in the Taizhou region have largely declined. The observed atmospheric OCP profile is generally consistent with previous studies; for instance, in eastern coastal China and the Yangtze River Delta, DDTs and their metabolites typically dominate atmospheric OCPs [33], with a higher proportion of DDE often regarded as an indicator of historical residues. In low-background regions such as the Tibetan Plateau and Inner Mongolian grasslands [8], HCHs are detectable but occur at significantly lower levels than DDTs, with β -HCH showing relatively higher enrichment among isomers. Methoxychlor exhibited the highest concentration among individual compounds in this study, consistent with observations from some industrial–agricultural mixed regions. Its elevated atmospheric levels may result from a combination of relatively high volatility and slow environmental degradation.

3.2.3. In Dust

In this study, ten OCPs were quantified in indoor dust samples from Taizhou (Table S4). The mean Σ OCPs concentration was 14.09 ng/g, with a median of 10.45 ng/g, indicating that OCPs persist in indoor environments long after their ban. The dominant compounds—o,p'-DDE, p,p'-DDD, and methoxychlor—highlight the notable enrichment of DDT-related metabolites. As a primary environmental metabolite of DDT, p,p'-DDE has been consistently reported at high concentrations in indoor dust worldwide. The enrichment of DDT-related metabolites in indoor dust may result from both historical atmospheric deposition and transformation processes occurring after deposition. DDT deposited onto indoor surfaces can undergo degradation through aerobic and anaerobic pathways, producing DDE and DDD as major transformation products. Under oxygenated conditions, DDT is preferentially converted to DDE through dehydrochlorination, whereas reducing environments favor reductive dechlorination and formation of DDD. Therefore, the predominance of DDE and DDD in indoor dust samples suggests that long-term transformation of historical DDT residues has contributed substantially to the current congener profiles. The mean o,p'-DDE concentration observed here is comparable to other domestic and international urban dust studies; in China, o,p'-DDE concentrations are frequently higher than those of other OCPs, reflecting the persistence and preferential accumulation of DDT metabolites in indoor environments [33]. Methoxychlor exhibited the highest mean concentration among individual compounds in this study, consistent with observations from dust samples in regions characterized by mixed industrial and agricultural activities. The occurrence of methoxychlor in indoor dust may be related to its relatively higher volatility compared with highly chlorinated DDT compounds, facilitating redistribution between air and indoor particles. Although HCH congeners were detected, their concentration remained relatively low. Compared with outdoor media such as soil and air, indoor dust—an important human exposure pathways showed evident enrichment of OCPs, highlighting its role as a key reservoir for legacy pollutants.

3.2.4. In Food

In this study, residues of ten organochlorine pesticides (OCPs) were quantified in common food samples from Taizhou (Table S5). Contamination levels were generally low, with a mean Σ OCPs concentration of 0.91 ng/g and a median of 0.41 ng/g. The similarity between mean and median values indicates that DDE and DDT were the dominant OCPs in food, while HCH congeners occurred at comparatively low levels. This pattern reflects the bioaccumulation and trophic transfer behavior of OCPs in the food chain. As the primary metabolite of DDT, p,p'-DDE is more frequently detected in food owing to its higher environmental stability and longer biological half-life [27]. Compared with the parent DDT, DDE has greater persistence and stronger lipid affinity, which facilitates accumulation in animal-derived foods and subsequent transfer to humans through dietary intake. This mechanism also explains the higher proportion of p,p'-DDE observed in human serum compared with environmental matrices. In contrast, HCH congeners occurred at relatively low levels, consistent with recent monitoring studies, suggesting that historical inputs of technical HCH have largely declined and that their transfer and accumulation along the food chain are lower than those of DDT-related compounds. Similar food residue surveys across multiple Chinese cities have reported that p,p'-DDE and p,p'-DDT dominate in cereals, vegetables, and animal-derived products, with concentrations generally ranging from 0.1 to 5 ng/g, in line with the levels observed in this study [34,35]. Overall, OCP contamination in food from Taizhou remains low; however, DDT and its metabolites continue to dominate, reflecting the enduring impact of historical environmental inputs and the persistent bioaccumulation of these compounds along the food chain on dietary exposure.

3.3. Multi-Pathway Human Exposure to Organochlorine Pesticides

OCPs in environmental media are potential sources of human exposure and can enter the human body via three primary pathways: oral ingestion, inhalation, and dermal contact [17–19]. To comprehensively evaluate the potential exposure routes of OCPs in the Taizhou population, relative contributions of individual compounds were estimated based on concentrations measured in soil, air, indoor dust, and food (Figure 2). The results showed that dietary intake was the predominant exposure pathway, particularly for DDT and its metabolites (e.g., p,p'-DDE), which accounted for the largest proportion of total exposure. The dominance of dietary exposure is closely related to the physicochemical properties of OCPs. Compounds with high hydrophobicity, particularly DDT and DDE, preferentially partition into lipid-rich matrices and accumulate through trophic transfer. Consequently, although environmental concentrations in food may be lower than those in soil or air, their higher bioavailability and ingestion rate result in greater contributions to human exposure. This was followed by indoor dust ingestion and atmospheric inhalation, whereas soil contact contributed minimally. HCH isomers contributed relatively little to total exposure (0.23–0.65), with β -HCH exhibiting slightly higher contributions from air and food compared with other isomers (Table S6). Overall, DDT-related compounds and their metabolites were the major contributors to human exposure, with p,p'-DDE alone accounting for the highest total exposure level (9.17 ng/kg bw/day) across all media, highlighting its persistent accumulation and dominant role in dietary and environmental exposure. Previous studies have indicated that dietary intake constitutes the predominant exposure pathway for OCPs [35], reflecting the preferential accumulation of DDE and DDT in lipid-rich food items relative to other environmental media. Moreover, indoor dust is increasingly recognized as a significant exposure medium in residential environments, as studies indicate that hand-to-mouth ingestion and inhalation of dust can contribute more to OCP exposure than soil, especially in children due to their higher contact rates and behavioral patterns [36]. The contribution of indoor dust may also reflect the role of residential environments as long-term reservoirs of legacy OCPs. Due to their strong sorption to organic matter and particles, DDT-related compounds deposited indoors can persist for decades and continuously contribute to human exposure through dust ingestion and inhalation. Long-term monitoring studies suggest that while HCHs and DDTs can undergo long-range atmospheric transport and contribute to human exposure, the inhalation pathway generally accounts for a smaller fraction of total exposure compared with dietary intake [37].

3.4. Health Risk Assessment of Multi-Media Exposure to OCPs in the Population of Taizhou, China

This study applied the hazard quotient (HQ) approach to assess the potential health risks of human exposure to OCPs via different environmental media in Taizhou. The HQ was calculated using the following equation, and an HQ value > 1 indicates a potential adverse health effect on human health [38]. Oral reference doses (RfDs) recommended by the U.S. Environmental Protection Agency (EPA) were used as benchmark values, as follows: α -HCH: 8000 ng/(kg bw·day); β -HCH: 300 ng/(kg bw·day); γ -HCH: 300 ng/(kg bw·day); δ -HCH: 300 ng/(kg bw·day); o,p'-DDE: 500 ng/(kg bw·day); p,p'-DDE: 500 ng/(kg bw·day); o,p'-DDD: 500 ng/(kg bw·day); p,p'-DDD: 500 ng/(kg bw·day); p,p'-DDT: 500 ng/(kg bw·day); and methoxychlor: 5000 ng/(kg bw·day) [39]. By

comparing HQ values across different compounds, the relative magnitude of health risks associated with OCP exposure can be systematically evaluated.

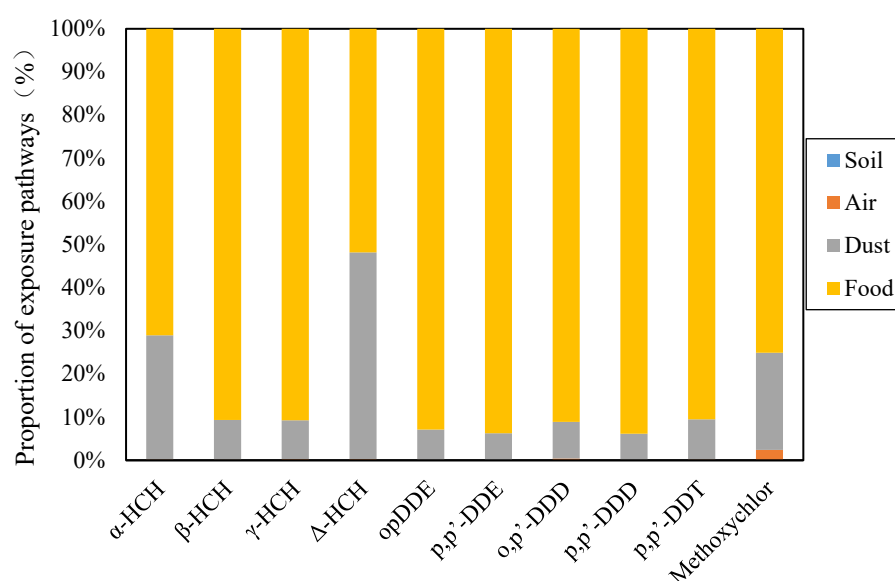


Figure 2. Multi-Media Contributions of OCPs to External Human Exposure. Note that the blue component is present in the figure but may not be visually discernible because of its relatively low contribution.

This study systematically evaluated the multi-media exposure risk of OCPs in the population of Taizhou using the hazard quotient (HQ) approach (Table 4). In addition to compound-specific HQ evaluation, the cumulative risk of OCP mixtures was assessed using the hazard index (HI), calculated as the sum of individual HQ values. The calculated HI value for total OCP exposure was 3.75×10^{-2} , which remained substantially below the threshold value of 1, suggesting that the combined exposure risk from the investigated OCP mixture was low under the current exposure scenario. Although the cumulative risk remained low, distinct differences were observed among individual compounds and exposure pathways. Overall, dietary intake represented the dominant exposure route, with the highest contributions from DDT and its metabolites (p,p'-DDE and p,p'-DDT) as well as β-HCH, followed by indoor dust ingestion and inhalation, while atmospheric inhalation and soil contact contributed minimally to total risk. Among DDT-related compounds, p,p'-DDE accounted for the largest contribution (1.83×10^{-2}), followed by p,p'-DDT (1.10×10^{-2}), consistent with previous studies reporting widespread dietary exposure to DDT residues in food products across China [10]. The predominance of p,p'-DDE in risk contribution is consistent with its environmental persistence and bioaccumulation potential. Although DDT application has been restricted for decades, continuous transformation of residual DDT and accumulation of stable metabolites maintain the exposure contribution of DDE. For HCH isomers, β-HCH exhibited the highest dietary contribution (1.97×10^{-3}), followed by γ-HCH (8.52×10^{-4}), likely due to its higher lipophilicity and longer biological half-life [30]. Overall, dietary intake remained the dominant exposure pathway for OCPs in Taizhou, with DDT-related compounds contributing more substantially than HCHs. Indoor dust represented a secondary but non-negligible exposure pathway, particularly for lipophilic metabolites such as DDE and DDT, highlighting its role as an important reservoir for persistent organic pollutants in residential environments. Nevertheless, several uncertainties should be considered when interpreting the exposure estimates. The exposure parameters used in this study were obtained from standardized exposure factor databases and previous studies, which may not fully capture regional differences in dietary habits, lifestyle behaviors, and environmental contact patterns in Taizhou. In particular, coastal dietary characteristics, including seafood consumption, may influence dietary exposure to OCPs. Furthermore, some dermal exposure parameters were adopted from international studies due to the limited availability of China-specific data, which may introduce additional uncertainty. Future studies integrating locally measured exposure factors, individual dietary information, larger sample sizes, and uncertainty analyses are warranted to improve the precision of OCP exposure assessment.

Table 4. Multi-Media Exposure Risk Assessment of OCPs in Environmental Media and Food.

	α -HCH	β -HCH	γ -HCH	Δ -HCH	opDDE	p,p'-DDE	o,p'-DDD	p,p'-DDD	p,p'-DDT	Methoxychlor
Soil	1.57×10^{-9}	3.78×10^{-8}	1.68×10^{-8}	2.94×10^{-8}	3.27×10^{-8}	6.32×10^{-7}	1.01×10^{-7}	1.94×10^{-7}	7.40×10^{-7}	4.78×10^{-9}
Air	5.97×10^{-8}	3.04×10^{-6}	2.28×10^{-6}	1.81×10^{-6}	5.70×10^{-7}	1.29×10^{-5}	3.62×10^{-6}	2.01×10^{-6}	1.98×10^{-5}	2.67×10^{-6}
Dust	1.16×10^{-5}	1.99×10^{-4}	8.60×10^{-5}	3.70×10^{-4}	4.38×10^{-5}	1.13×10^{-3}	7.82×10^{-5}	1.59×10^{-4}	1.02×10^{-3}	2.51×10^{-5}
Food	2.81×10^{-5}	1.97×10^{-3}	8.52×10^{-4}	3.99×10^{-4}	5.85×10^{-4}	1.72×10^{-2}	8.42×10^{-4}	2.45×10^{-3}	9.99×10^{-3}	8.40×10^{-5}
Total	3.99×10^{-5}	2.17×10^{-3}	9.40×10^{-4}	7.71×10^{-4}	6.30×10^{-4}	1.83×10^{-2}	9.24×10^{-4}	2.61×10^{-3}	1.10×10^{-2}	1.12×10^{-4}

3.5. Comparison of External OCP Exposure Profiles between Human Samples and Environmental Media and Food

To explore the potential relationship between external environmental occurrence and internal human body burden, this study compared the compositional profiles of OCPs in serum with those in soil, air, indoor dust, food, and integrated exposure estimates (Figure 3). Overall, DDT-related compounds dominated across all environmental media and in human serum; however, notable differences in congener composition were observed. These differences should be interpreted cautiously because serum OCP concentrations represent integrated internal burdens accumulated over extended periods, whereas environmental measurements reflect contamination levels at the sampling time. Therefore, serum profiles may reflect historical exposure, biological persistence, and metabolic processes in addition to recent environmental inputs. In the integrated exposure profile, p,p'-DDE (48.46%) and p,p'-DDT (29.57%) were the major contributors, jointly accounting for nearly 80% of total OCP exposure, indicating that current exposure is largely driven by DDT-related compounds. In food, p,p'-DDE accounted for 50.16%, closely matching the integrated exposure pattern and suggesting that dietary intake is a key pathway for DDT-related exposure. In contrast, p,p'-DDT dominated in soil and indoor dust, accounting for 40.61% and 31.36%, respectively, while methoxychlor showed the highest proportion in air (37.70%), indicating distinct source contributions and transformation behaviors across environmental media.

Compared with the external exposure profile, the composition in human serum showed notable differences. p,p'-DDE dominated in serum (67.82%), significantly higher than its proportion in integrated exposure (48.46%), whereas p,p'-DDT decreased markedly to 3.01% compared with 29.57% in external exposure. These differences may be partly associated with metabolic transformation after exposure, in which p,p'-DDT can be converted to the more stable p,p'-DDE through dehydrochlorination, as well as preferential retention of DDE in lipid-rich tissues. However, this discrepancy should not be interpreted solely as a direct response to current environmental exposure conditions. Due to the long biological half-lives of DDE and other OCPs, serum concentrations integrate exposure histories over years or even decades. Therefore, the higher proportion of p,p'-DDE in serum likely reflects the combined effects of historical DDT contamination, ongoing low-level exposure, metabolic conversion, and selective biological accumulation [27]. Similarly, β -HCH showed enrichment in serum (7.60%) compared with integrated exposure (3.49%), consistent with its greater stability and persistence in the human body [30], whereas α -HCH and γ -HCH exhibited relatively minor changes, suggesting isomer-specific differences in metabolism and elimination. This enrichment may also reflect the long-term persistence of β -HCH, which has a relatively longer biological half-life and stronger retention capacity compared with other HCH isomers. This discrepancy highlights that atmospheric abundance does not necessarily directly determine human body burden. Compound-specific properties, including volatility, deposition behavior, metabolic transformation, and bioavailability, jointly regulate the transfer efficiency from environmental media to humans. In contrast, methoxychlor, although dominant in the atmosphere (37.70%), contributed only 2.98% to integrated exposure but increased to 14.53% in serum, suggesting that internal exposure may be influenced by differences in exposure efficiency and bioavailability across pathways. These observations highlight that atmospheric abundance or current environmental concentration alone cannot directly predict human body burden. Instead, compound-specific physicochemical properties, biological persistence, metabolic transformation, bioavailability, and exposure history jointly determine internal accumulation. Nevertheless, the interpretation of relationships between external environmental concentrations and internal serum burdens remains subject to uncertainty. Due to the limited sample size and the lack of repeated measurements, statistical associations between individual environmental matrices and serum OCP concentrations were not evaluated in the present study. Moreover, considering the long biological half-lives of legacy OCPs, current serum concentrations may represent cumulative historical exposure rather than only contemporaneous environmental contamination. Future studies with larger cohorts, longitudinal sampling, and individual-specific exposure information are required to better quantify the contribution of specific environmental sources to internal OCP accumulation.

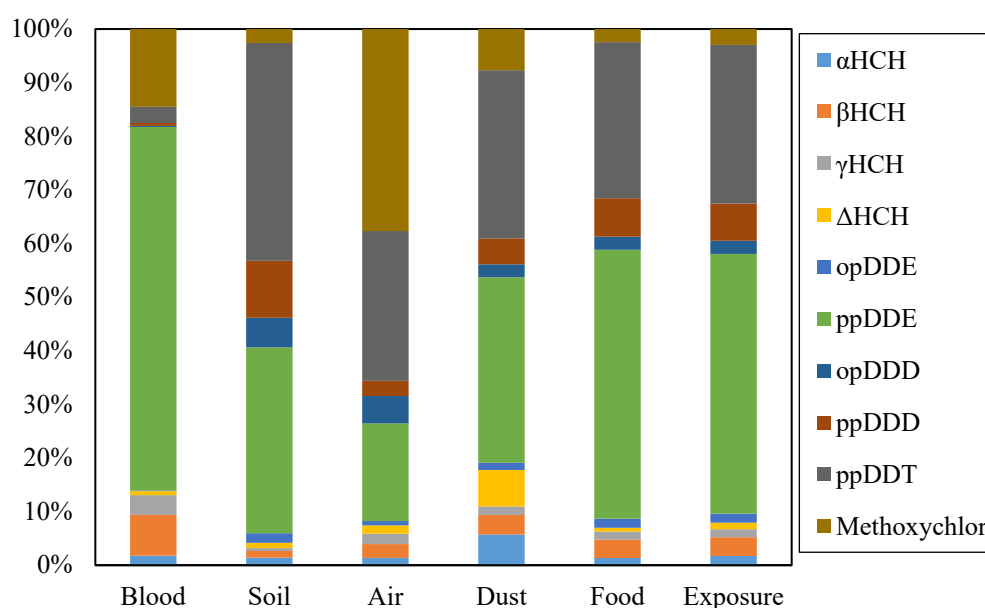


Figure 3. OCP Compositional Proportions in Serum and External Exposure Media.

4. Conclusions

This exploratory study investigated the occurrence patterns and preliminary distribution characteristics of multiple OCPs in human serum and surrounding environmental media (soil, air, indoor dust, and food) in Study area, and evaluated multi-pathway exposure and associated health risks by integrating external environmental measurements with internal human biomonitoring. The results indicated that OCPs, despite long-term regulatory restrictions, remain detectable across environmental compartments and may continue to contribute to human exposure. DDT-related compounds, particularly p,p'-DDE, dominated the serum profile, while β-HCH showed preferential accumulation among HCH congeners, reflecting the persistence, lipophilicity, and biological retention characteristics of these legacy pollutants. The observed differences among environmental media suggested that OCP fate may be influenced by the combined effects of environmental persistence, transformation, inter-media transport, and bioaccumulation. Food was estimated to be the dominant exposure pathway for highly lipophilic DDT-related compounds under the applied exposure assumptions, likely due to their preferential accumulation in lipid-rich food matrices and previously reported biomagnification potential, whereas atmospheric occurrence and indoor dust accumulation reflected their potential for volatilization, long-range transport, and redistribution among environmental compartments. Differences between external exposure profiles and serum compositions further suggested that environmental concentration alone may not fully represent human internal burdens, as historical exposure, compound-specific metabolism, biological half-life, and lipid-associated accumulation jointly influence serum concentrations. Although both individual compound risks and cumulative mixture risks remained low under the current exposure scenario, the continued detection of DDT metabolites and β-HCH highlights the persistent environmental legacy of OCPs. Their persistence, potential for long-range transport, bioaccumulation capacity, and previously reported biomagnification potential allow these compounds to remain environmentally relevant decades after restriction. Overall, this exploratory study provides preliminary insights into the environmental fate and potential human exposure pathways of legacy OCPs and highlights the value of integrating environmental monitoring with biological measurements for improved assessment of persistent organic pollutants.

Supplementary Materials

The additional data and information can be downloaded at: <https://media.scilit.com/articles/others/2608251654010548/ECCS-26060187-SI.pdf>. Refs. [40,41] are cited in the supplementary materials.

Author Contributions

Author Contributions: L.H.: conceptualization, methodology, investigation, data curation, formal analysis, visualization, writing—original draft preparation; Y.F.: investigation, data curation, formal analysis, visualization, writing—original draft preparation; K.G.: methodology, investigation, formal analysis; Y.W.: investigation, data curation; C.G.: investigation, data curation, formal analysis; X.L.: investigation, data curation; D.C.: methodology,

formal analysis, validation; J.J. (Junjie Jiang): methodology, validation, supervision; M.D.: investigation, data curation, formal analysis; Y.W.: supervision, methodology, writing—review and editing; J.J. (Jun Jin): conceptualization, supervision, project administration, writing—review and editing. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement

The study was conducted in accordance with the Declaration of Helsinki and was approved by the Biological and Medical Ethics Committee of Minzu University of China (protocol code ECMUC2020026CA, approved on 13 April 2020).

Informed Consent Statement

Written informed consent was obtained from all participants involved in the study prior to sample collection. No personally identifiable information was disclosed in this manuscript; therefore, consent for publication was not applicable.

Data Availability Statement

The data generated and/or analyzed during this study are available from the corresponding author upon reasonable request, subject to ethical and privacy restrictions. The data cannot be made publicly available because they involve human serum samples and may contain sensitive information that could compromise participant confidentiality.

Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

During the preparation of this work, the authors used ChatGPT (OpenAI) solely for language editing, including improving English grammar, wording, and readability. The authors reviewed and revised all AI-assisted edits and take full responsibility for the content of the published article.

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