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Risk Assessment on Organochlorine Pesticides in Agricultural Soils of Eastern City, China

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Abstract: To explore how organochlorine pesticides (OCPs) are perpetual in soils and the risk they may bring, Ningbo, a city with an extensive usage history of OCPs, was selected as a case to investigate. Sixty-nine agriculture soils were taken from 0–20 cm layers, then OCPs were analyzed, and a risk assessment was performed. Results indicate five OCPs were detected in agricultural soils, with total concentrations ranging from below detection limits to 43.08 μg·kg^{-1} and an average value of 15.58 μg·kg^{-1}. Among them, δ-Hexachlorocyclohexane (δ-HCH) and p, p′-Dichlorodiphenyltrichloroethane (p, p′-DDT) were the primary contributors to the residual contamination levels. The health risk assessment indicates that even at maximum exposure levels, the non-carcinogenic risk (1.71×10^{-4}) and carcinogenic risk (5.97×10^{-8}) of OCPs in the study area are significantly below the risk thresholds of 1 and 10^{-6}, respectively. Monte Carlo simulation further confirms that the 95th percentile values for non-carcinogenic and carcinogenic risks (3.39×10^{-4} and 1.23×10^{-7}) remain well below these limits, suggesting that the health risks posed by OCPs to adults are negligible. Subsequent ecological risk assessment revealed that the vast majority (73.91%) of soil samples exhibited medium-low ecological risk, with dichlorodiphenyltrichloroethane (DDTs) being the primary contributor to ecological risk. Our findings strengthen the view that although OCPs have been banned for a long time, the ecological risks of residuals in the soil remain a concern, and more effective control methods should be used to mitigate them.

Keywords: organochlorine pesticides (OCPs); hexachlorocyclohexanes (HCHs); dichlorodiphenyltrichloroethane (DDTs); health risk assessment; ecological risk assessment

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1. Introduction

Organochlorine pesticides (OCPs) are well-known persistent pollutants with high toxicity and bioaccumulation [1,2]. Owing to their high efficiency and cost-effectiveness, OCPs have been used extensively in agricultural processes to control pests and enhance crop quality and yield [3,4]. Hexachlorocyclohexane (HCHs) and dichlorodiphenyltrichloroethane (DDTs) are the main varieties of OCPs, which were widely produced and used in China before being banned in 1983 [5,6]. Their stable chemical properties make them highly resistant to degradation, allowing them to remain in various environmental media for extended periods and continuously migrate and transform, ultimately causing multiple negative impacts on ecosystems and human health [7–9].

In China, regulatory thresholds for OCPs in agricultural soils have been established to mitigate these risks. For instance, the risk screening values for HCHs and DDTs in agricultural soils are set at 0.10 mg·kg^{-1} according to the national soil environmental quality

standards (GB 15618-2018) [10]. These thresholds are crucial for assessing and managing the potential contamination risks associated with OCPs. The specific ecological risks of OCPs include bioaccumulation in food chains, which can lead to toxic effects on higher trophic levels and disrupt ecosystem balance [11,12]. For human health, exposure to OCPs through contaminated food and water can cause acute poisoning, cancer, and neurological disorders [13]. Studies have also shown that OCPs can pose risks to soil microorganisms and affect soil health, further influencing agricultural productivity [14]. Therefore, risk assessment of residual OCPs is crucial for protecting the ecological environment and human health.

The health risk assessment (HRA) model recommended by USEPA is a robust tool for evaluating agricultural soil contamination risks, widely applied in studies of OCPs in farmland soils [14–16]. Conventional HRA relies on fixed parameters (e.g., ingestion rates, exposure frequency) to generate single-point risk estimates, yet fails to account for soil pollutant heterogeneity and population variability. Monte Carlo simulation addresses these limitations by integrating probability distributions of key variables to quantify risk likelihoods within confidence intervals (e.g., 5th–95th percentile) [17–19]. However, there is currently no unified standard for evaluating soil environmental ecological risk. Given the bio-amplification effects of OCPs in the food chain, food chain-related models are widely used both domestically and internationally to evaluate ecological risks. For instance, Urzelai et al. [20] established HCHs risk thresholds for soil invertebrates using toxicological benchmarks, while Long et al.'s [21] effect-range thresholds (ERL: effects range low; ERM: effects range median) delineate pollutant impacts on soil biota. These methodologies have become cornerstone approaches for evaluating OCPs' ecological risks across soil ecosystems [22].

Ningbo, a major agricultural base in Zhejiang Province and a highly urbanized region has seen rapid development in intensive agriculture driven by the increasing demand for agricultural products. Historically, from 1980 to 1984, Ningbo used approximately 25,000 tons of OCPs to boost agricultural yields [23,24]. This extensive historical use has likely led to significant OCPs residue problems in the region. A study in 2012 reported that the concentrations of OCPs in suburban agricultural soils of Ningbo ranged from n.d. to 41.3 $\mu g \cdot kg^{-1}$ [25], indicating that high levels of OCPs residues persist in soils even decades after the implementation of bans. Moreover, residues of multiple OCPs have been detected in economic seafood from the Ningbo area [26]. This "high historical input-strong environmental persistence-multi-medium diffusion" characteristic poses a serious threat to regional soil health and human safety.

Therefore, the contamination status of OCPs (HCHs and DDTs), their spatial distribution, and potential risks of human health and ecosystem in soils collected from farmland of the Ningbo region, China, were comprehensively investigated in this study. By achieving these objectives, this study aimed to contribute to a deeper understanding of the environmental hazards associated with OCPs and support sustainable agricultural practices and environmental management in the region.

2. Materials and Methods

2.1. Study Area and Sample Collection

Ningbo, situated in the eastern coastal region of Zhejiang Province and at the southern wing of the Yangtze River Delta, is characterized by a subtropical monsoon climate with mild temperatures, ample humidity, and distinct seasons. The annual mean temperature ranges from 16.4 °C to 17.5 °C, and the average annual precipitation is approximately 1480 mm, concentrated mainly from May to September. The region's land use is diverse, dominated by arable land, forestland, and water bodies, with arable land primarily used for

cultivating rice and vegetables. Soil types varied, including red soil, yellow soil, paddy soil, and coastal saline soil, with textures predominantly consisting of clay and loam. Ningbo, with its long-standing agricultural history, remains one of the key agricultural production areas in Zhejiang Province.

To systematically locate sampling points within the agricultural lands of the study area, a 2 km × 2 km grid was initially established based on remote sensing images and land-use maps. The positions of these points were subsequently optimized by integrating factors such as industrial and agricultural layout, crop planting areas, and cultivation history. From September to November 2022, a total of 69 agricultural soil samples were collected at a depth of 0–20 cm using clean stainless steel shovels (Delixi Electric Co., Ltd., Leqing, China). Each sample was composed of five closely located subsamples collected in a diagonal pattern to account for spatial variability and ensure representativeness. The quartering method was then employed to reduce the bulk to approximately 1 kg of mixed soil, which was stored in polyethene bags away from light. Detailed information, including GPS coordinates, land-use type, and crop type, was meticulously recorded for each sample. During the sampling process, areas with recent disturbances (e.g., recent cultivation or construction) were avoided to maintain sample integrity. The distribution of sampling points is illustrated in Figure 1.

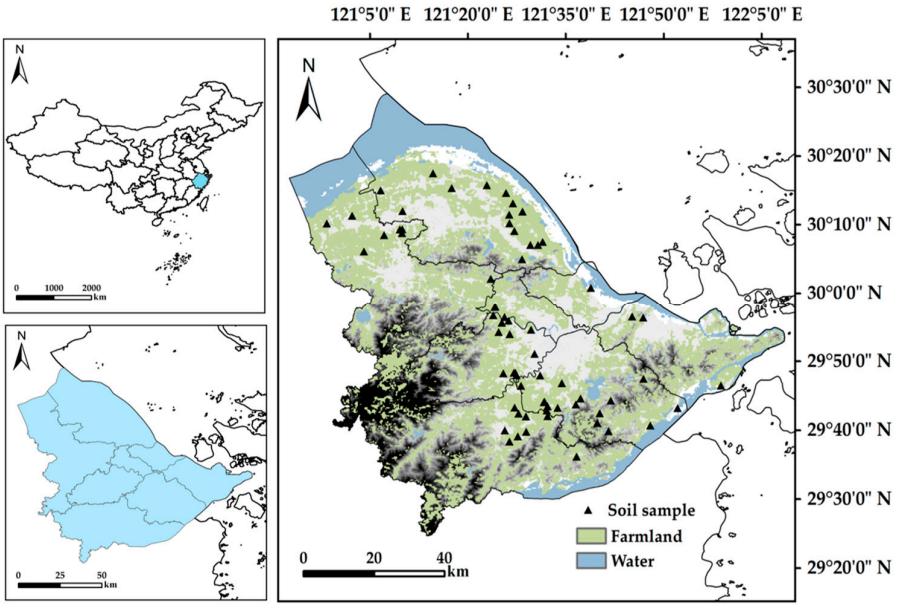

Figure 1. Sample sites.

2.2. Sample Testing

2.2.1. Sample Preparation and Instrumental Analysis

The soil samples, stored at −20 °C until analysis, were first sifted to remove extraneous materials such as roots, leaves, and stones. They were then subjected to freeze-drying using a vacuum freeze-dryer (Biosafer-10A, Safer, Nangjing, China) to achieve dehydration. After drying, the samples were ground and passed through a 100-mesh sieve to produce a homogeneous powder. All subsequent analyses were performed on these prepared samples, with results reported on a dry-weight basis. The samples were extracted using an n-hexane–acetone mixture (1:1, v/v) under the following conditions: pressure of 0.8 MPa, extraction temperature of 100 °C, preheating equilibration time of 5 min, static extraction for 5 min, solvent rinse of 60% extraction cell volume, and two extraction cycles. The extract was concentrated to 1 mL via nitrogen blow-down evaporation, purified by gel permeation

chromatography (GPC), further concentrated, and reconstituted to a final volume of 1 mL for subsequent analysis.

The samples were analyzed using Gas Chromatography/Mass Spectrometry (GC/MS, AMD9Pro, PANNATEK, Changzhou, China). GC employed a splitless injection (250 °C, 1.0 μL) with a constant column flow of 1.0 mL/min. The oven temperature protocol included: 120 °C (2 min) → 12 °C/min to 180 °C (5 min) → 7 °C/min to 240 °C (1 min) → 1 °C/min to 250 °C (2 min). MS operated in electron ionization (EI) mode at 230 °C (ion source) and 150 °C (quadrupole), with a 5 min solvent delay and full-scan acquisition across 45–450 amu.

The pollutants to be tested included α-HCH (α-Hexachlorocyclohexane), β-HCH (β-Hexachlorocyclohexane), γ-HCH (γ-Hexachlorocyclohexane), δ-HCH (δ-Hexachlorocyclohexane), p, p′-DDE (p, p′-Dichlorodiphenyldichloroethylene), o, p′-DDT (o, p′-Dichlorodiphenyltrichloroethane), p, p′-DDD (p, p′-Dichlorodiphenyldichloroethane), and p, p′-DDT (p, p′-Dichlorodiphenyltrichloroethane).

2.2.2. Quality Assurance and Quality Control

All samples were comprehensively analyzed at the China Customs Science and Technology Research Center (STRC), which holds accreditation under China Metrology Accreditation (CMA) [27] and is recognized as a proficiency testing provider by the China National Accreditation Service for Conformity Assessment (CNAS PT0012) [28]. These accreditations confirm compliance with nationally and internationally recognized standards for technical competence and quality management in testing activities.

All procedures strictly followed national standards (HJ 835-2017) [29] and were conducted under quality assurance (QA) and quality control (QC). The results conformed to the standard requirements, with a relative standard deviation (RSD) of less than 5% and a recovery rate of 85.6% to 96.0%. The limits of detection (LOD) were determined three times using a signal-to-noise ratio (S/N) and summarized as follows: α-HCH (0.07 $mg \cdot kg^{-1}$), β-HCH (0.06 $mg \cdot kg^{-1}$), γ-HCH (0.06 $mg \cdot kg^{-1}$), δ-HCH (0.10 $mg \cdot kg^{-1}$), p, p′-DDE (0.04 $mg \cdot kg^{-1}$), o, p′-DDT (0.08 $mg \cdot kg^{-1}$), p, p′-DDD (0.08 $mg \cdot kg^{-1}$), and p, p′-DDT (0.09 $mg \cdot kg^{-1}$). For quantified OCPs, concentrations below the LOD were considered undetectable (n.d.).

2.3. Health Risk Assessment

The health risk assessment (HRA) model developed by the United States Environmental Protection Agency (US EPA) was utilized in this study, which has been widely practiced [30–33]. The average daily dose (ADD, $mg \cdot kg^{-1} \cdot day^{-1}$) of OCPs via three pathways (soil ingestion, dermal contact and inhalation) were calculated using the following equations.

$$ADD_{ingest} = (C \times IngR \times EF \times ED)/(BW \times AT) \times 10^{-6} \quad (1)$$

$$ADD_{inhale} = (C \times InhR \times EF \times ED)/(PEF \times BW \times AT) \quad (2)$$

$$ADD_{dermal} = (C \times AF \times SA \times ABS \times EF \times ED)/(BW \times AT) \times 10^{-6} \quad (3)$$

where C represents the concentration of chemical substances in soil ($mg \cdot kg^{-1}$); IngR represents the soil ingestion rate ($mg \cdot day^{-1}$); EF is used to indicate the frequency of exposure ($days \cdot year^{-1}$); ED is the exposure duration (year); BW is the body weight (kg); AT refers to the average lifetime (days); InhR is the inhalation rate ($m^3 \cdot day^{-1}$); PEF is the particulate emission factor ($m^3 \cdot kg^{-1}$); AF is the skin adherence factor ($mg \cdot cm^{-2} \cdot day^{-1}$); SA is the exposed skin area (cm^2); ABS is the fraction of applied dose absorbed across skin. Given that the sampling area is not environmentally sensitive, the health risks of soil OCPs

exposure are assessed only for adults. All the parameters used in the non-cancer risk and cancer risk assessments are listed in Table S1.

HRA involves two main steps: The initial step focuses on evaluating the non-cancer risk that pollutants have the potential to cause non-cancer effects. In contrast, the second step involves assessing the carcinogenic potential posed by the pollutants. In general, the non-cancer risk is quantified using the Hazard Index (HI) (Equation (4)), while the carcinogenic risk is expressed as the Carcinogenic Risk (CR) (Equation (6)).

$$HI = ADD/RfD \quad (4)$$

$$THI = \sum_{i=1}^{n} ADD_i/RfD_i \quad (5)$$

Here, RfD means reference dose ($mg \cdot kg^{-1} \cdot day^{-1}$) (Table S2), which from the US EPA website. The cumulative non-cancer risk is expressed as the Total Hazard Index (THI) (Equation (5)). When HI or THI ≤ 1, non-carcinogenic risks can be ignored. If HI or THI exceeds 1, it indicates the presence of noncarcinogenic health risks, which increase with the increase of HI value.

The calculation of CR is seen in Equation (6):

$$CR = ADD \times SF \quad (6)$$

$$TCR = \sum_{i=1}^{n} ADD_i \times SF_i \quad (7)$$

where SF is the slope factor ($kg \cdot day \cdot mg^{-1}$) (Table S2), which from the US EPA website. When considering multiple types of pollutants (n), the Total Carcinogenic Risk (TCR) can be calculated in Equation (7). If the CR or TCR values between 10^{-6} and 10^{-4} are considered to be acceptable, while those exceeding 10^{-4} are considered to constitute a lifetime carcinogenic risk to humans. A risk factor < 10^{-6} is regarded as negligible or no risk.

2.4. Monte Carlo Simulation

The Monte Carlo simulation was performed using Oracle Crystal Ball 11.1.2.4, a user-friendly tool that facilitates the simulation process and provides cumulative risk frequency distributions [34]. The probabilistic distribution of health risks associated with soil OCPs was derived using the Monte Carlo method with the following steps: (1) define the distribution ranges for the input variables; (2) specify the output variables; (3) randomly sample values for the input variables to simulate the probability distribution of the output variables. Some exposure parameters were optimized and presented in Table S3 after citing references [17–19,35].

2.5. Ecological Risk (Eco-Risk) Assessment

2.5.1. Effects Range Low/Median

Assessment of OCPs for their ecotoxicological significance was evaluated using effects range low (ERL) and effects range median (ERM). ERL refers to the concentration below which toxic effects are scarcely observed, while ERM represents the concentration above which adverse effects are likely to occur [21]. The US National Oceanic and Administration (NOAA) guidelines provide two values for each chemical. When the pollutant content is below the ERL value, it represents a minimal-effects range, a range intended to estimate conditions in which effects would be rarely observed. Concentrations equal to and above the ERL, but below the ERM, represent a possible-effects range within which effects would occasionally occur. Finally, the concentrations equivalent to and above the ERM value represent a probable-effects range within which effects would frequently occur.

2.5.2. The Mean ERM Quotients

In order to obtain a more realistic measure of predicted toxicity than simply summing up the numbers of ERM exceeded, the mean ERM quotients (m-ERM-q) were calculated as Equation (8) [22]:

$$m - ERM - q = \frac{\sum(C_i / ERM_i)}{n} \quad (8)$$

where C_i is the measured concentration of soil pollutant i; ERM_i is the effects range median of pollutant i; n represents the number of pollutants. The classification criteria for m-ERM-q risk levels are shown in Table 1.

Table 1. The classification criteria for m-ERM-q risk levels.

The Value of m-ERM-q	Risk Level
≤0.1	Low
0.1 < m-ERM-q ≤ 0.5	Medium-Low
0.5 < m-ERM-q ≤ 1.5	Medium-High
>1.5	High

2.6. Data Analysis

Statistical analysis was conducted using Microsoft Excel 2016. The characteristics of organochlorine pesticide residues and associated risk data were visualized using Origin 2025. Additionally, the spatial distribution of OCPs was analyzed and mapped using the inverse distance weighted (IDW) geostatistical method in ArcMap 10.8.

3. Results and Discussion

3.1. Residual Characteristics of OCPs

In the analysis of 69 soil samples, 5 OCPs were identified, exhibiting concentrations that varied from below detection limits to 43.08 μg·kg^{-1}, with an overall average concentration of 15.58 μg·kg^{-1}. The detection rates for these compounds were as follows: δ-HCH at 75.36%, p, p′-DDE at 82.61%, o, p′-DDT at 84.06%, p, p′-DDD at 79.71%, and p, p′-DDT at 78.26%. These high-detection frequencies underscore the pervasive presence of OCPs in the study area's soil. The residual levels of OCPs in the soil are described in Figure 2a. The residual levels of δ-HCH ranged from n.d. to 21.52 μg·kg^{-1}, with a mean concentration of 4.87 μg·kg^{-1}. The content range of DDTs is n.d.~39.16 μg·kg^{-1}, with an average content of 10.71 μg·kg^{-1}. Notably, the residual concentrations of DDTs substantially exceeded those of HCHs, represented by δ-HCH, which is consistent with the monitoring results of OCPs in a large number of domestic data research areas [36–38]. Walker et al. [39] demonstrated that HCHs, characterized by lower octanol-water partition coefficients and higher vapor pressures than DDTs, are more likely to volatilize from soil and engage in long-range atmospheric transport.

Among the DDTs, p, p′-DDE exhibited concentrations ranging from n.d. to 9.00 μg·kg^{-1} (mean: 1.76 μg·kg^{-1}), while o, p′-DDT and p, p′-DDD showed ranges of n.d. to 16.41 μg·kg^{-1} (mean: 2.49 μg·kg^{-1}) and n.d. to 9.85 μg·kg^{-1} (mean: 2.15 μg·kg^{-1}), respectively. Strikingly, p, p′-DDT demonstrated the highest contamination level, with residues spanning n.d. to 31.01 μg·kg^{-1} and a mean concentration of 4.31 μg·kg^{-1}, underscoring its dominance within the DDTs group. As depicted in Figure 2b, δ-HCH contributes the most to the total residue levels of OCPs, accounting for 31%, followed by p, p′-DDT, which contributes 28%. These findings highlight the pervasive occurrence of OCPs in the studied soils, with δ-HCH and p, p′-DDT identified as the primary contributors to the residual contamination levels.

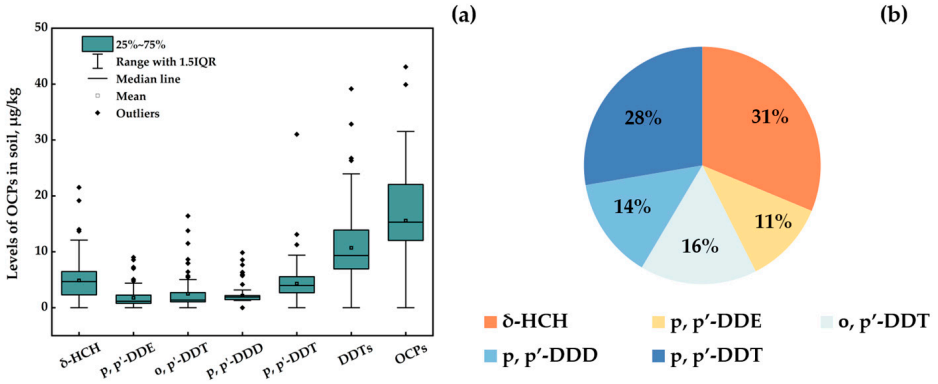

Figure 2. Residual characteristics of OCPs in soil. (**a**) The residual levels of OCPs in the soil; (**b**) the composition characteristics of OCPs in soil.

The spatial distribution of OCPs is illustrated in Figure 3. As indicated, δ-HCH is predominantly found in areas such as Beilun, certain regions of Yuyao, and Yinzhou, where residue levels are notably higher. Meanwhile, DDTs have higher residual levels in areas such as Cixi and Fenghua. Previous research has indicated that, except in coastal saline soils, DDTs residues in the soils of Cixi are generally higher than those of HCHs [40]. This is likely attributed to the higher historical application rates of DDTs compared to HCHs, as well as the more stable chemical structure of DDTs.

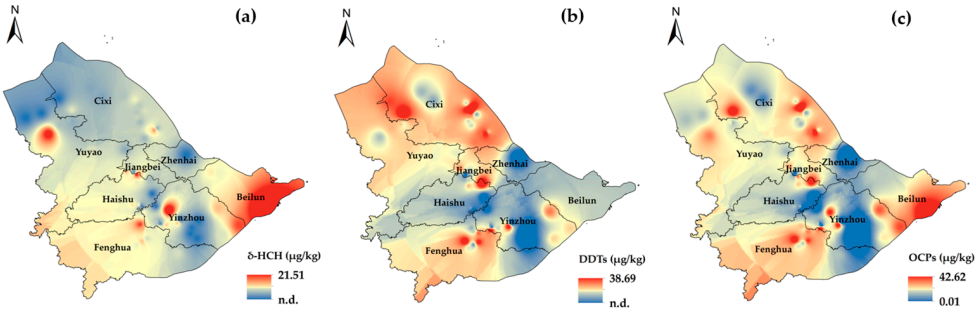

Figure 3. Spatial distribution of soil OCPs in the study area. (**a**) Spatial distribution characteristics of δ-HCH; (**b**) spatial distribution characteristics of DDTs; (**c**) spatial distribution characteristics of OCPs (δ-HCH and DDTs). Note: n.d. indicates a concentration below the detection limit.

To further analyze the residual levels of soil OCPs in the study area, the content of HCHs and DDTs in soils from some regions at home and abroad were compared with the results of this study, as shown in Table 2. The HCHs concentration in farmland soils substantially exceeds those reported in the Three Gorges Dam region (China), Mai Mahiu–Narok (Kenya), and the Campanian Plain (Italy), aligns with levels in the Pearl River Delta, Qinghai Province, and the Yellow River Basin (China), yet remains lower than those in Kaifeng, the Yangtze River Basin (China), and the Indus Basin (Pakistan). Similarly, soil DDTs levels surpass those in the Three Gorges Dam region and Mai Mahiu–Narok, are approximately the same values as those in Qinghai Province, the Yangtze River Basin, and the Yellow River Basin, but are markedly lower than concentrations in the Pearl River Delta, Kaifeng, the Indus Basin, and the Campanian Plain. These spatial disparities likely reflect the interplay of region-specific factors, including land use patterns and soil physicochemical properties associated with agricultural practices, which significantly influence the residual levels of pesticides in the soil environment [41,42].

Table 2. Concentration of HCHs and DDTs in other regions around the world.

Sampling Locations	HCHs (μg·kg^{-1})	DDTs (μg·kg^{-1})	References
Kaifeng City, China	45.35	119.76	[38]
The Three Gorges Dam region, China	1.27	1.80	[36]
Qinghai Province, China	4.78	7.76	[37]
The Pearl River Delta, China	4.52	34.2	[16]
The Yangtze River Basin, China	13.04	8.90	[43]
Yellow River basin, China	6.23	16.78	[44]
Mai Mahiu, Kenya	1.50	3.51	[45]
Narok, Kenya	1.62	4.71	[45]
The Indus Basin, Pakistan	15.41	142.26	[46]
The Campanian Plain, Italy	1.38	107	[47]

3.2. Health Risk Assessment (HRA)

3.2.1. Non-Carcinogenic Health Risk Evaluation

The non-carcinogenic health risks posed by δ-HCH, o, p′-DDT, and p, p′-DDT were evaluated (Table 3). The results indicate that, even at the maximum concentrations of OCPs, the non-carcinogenic risk indices for adults (THI_{max} = 1.71 × 10^{-4}) remain significantly below the risk thresholds (1.00). This suggests that the non-carcinogenic health impacts of OCPs on exposed populations within the study area are within a negligible range. Notably, among the three exposure pathways, the non-carcinogenic risks for adults are ranked as follows: ingestion (68.32%) > dermal (0.01%) > inhalation (31.67%). This finding is consistent with the research of Liu et al. [36], which also confirms that ingestion is the primary pathway of exposure. Additionally, further analysis revealed that, regardless of the exposure pathway, the risk values associated with δ-HCH are significantly higher than those of p, p′-DDT and o, p′-DDT.

Table 3. Results of non-carcinogenic risk assessment of OCPs in soil.

	The Mean of the HI				The Maximum of the HI			
	Ingest	Dermal	Inhale	Total	Ingest	Dermal	Inhale	Total
δ-HCH	1.13 × 10^{-5}	4.93 × 10^{-6}	1.82 × 10^{-9}	1.63 × 10^{-5}	5.02 × 10^{-5}	2.18 × 10^{-5}	8.03 × 10^{-9}	7.19 × 10^{-5}
o, p′-DDT	3.48 × 10^{-6}	1.74 × 10^{-6}	6.40 × 10^{-10}	5.22 × 10^{-6}	2.29 × 10^{-5}	1.14 × 10^{-5}	4.22 × 10^{-9}	3.44 × 10^{-5}
p, p′-DDT	6.02 × 10^{-6}	3.00 × 10^{-6}	1.11 × 10^{-9}	9.02 × 10^{-6}	4.34 × 10^{-5}	2.16 × 10^{-5}	7.97 × 10^{-9}	6.50 × 10^{-5}
DDTs	9.50 × 10^{-6}	4.74 × 10^{-6}	1.75 × 10^{-9}	1.42 × 10^{-5}	6.63 × 10^{-5}	3.31 × 10^{-5}	1.22 × 10^{-8}	9.94 × 10^{-5}
OCPs	2.08 × 10^{-5}	9.66 × 10^{-6}	3.56 × 10^{-9}	3.05 × 10^{-5}	1.16 × 10^{-4}	5.48 × 10^{-5}	2.02 × 10^{-8}	1.71 × 10^{-4}

3.2.2. Carcinogenic Health Risk Evaluation

The carcinogenic health risks posed by δ-HCH and DDTs (p, p′-DDE, o, p′-DDT, p, p′-DDD, and p, p′-DDT) were calculated (Table 4). The maximum value of TCR (5.97 × 10^{-8}) is significantly lower than the risk threshold (10^{-6}), indicating that the overall cancer risk associated with all exposure pathways through soil is negligible, even at the highest risk level. Among the exposure pathways for adults, the order of carcinogenic risk is ingestion (71.19%), dermal contact (0.01%), and inhalation (28.80%), consistent with the pattern observed for non-carcinogenic risks. In terms of the five OCPs, δ-HCH poses the highest carcinogenic risk, nearly an order of magnitude higher than DDTs.

Table 4. Results of carcinogenic risk assessment of OCPs in soil.

	The Mean of the CR				The Maximum of the CR			
	Ingest	**Dermal**	**Inhale**	**Total**	**Ingest**	**Dermal**	**Inhale**	**Total**
δ-HCH	6.13×10^{-9}	2.24×10^{-9}	9.01×10^{-13}	8.37×10^{-9}	2.71×10^{-8}	9.91×10^{-9}	3.98×10^{-12}	3.70×10^{-8}
p, p′-DDE	4.19×10^{-10}	2.11×10^{-10}	7.79×10^{-14}	6.30×10^{-10}	2.14×10^{-9}	1.08×10^{-9}	3.98×10^{-13}	3.22×10^{-9}
o, p′-DDT	5.92×10^{-10}	2.99×10^{-10}	8.70×10^{-14}	8.90×10^{-10}	3.90×10^{-9}	1.97×10^{-9}	5.74×10^{-13}	5.87×10^{-9}
p, p′-DDD	3.61×10^{-10}	1.80×10^{-10}	6.64×10^{-14}	5.42×10^{-10}	1.65×10^{-9}	8.24×10^{-10}	3.04×10^{-13}	2.48×10^{-9}
p, p′-DDT	1.02×10^{-9}	5.16×10^{-10}	1.50×10^{-13}	1.54×10^{-9}	7.37×10^{-9}	3.72×10^{-9}	1.08×10^{-12}	1.11×10^{-8}
DDTs	2.39×10^{-9}	1.21×10^{-9}	3.82×10^{-13}	3.60×10^{-9}	1.51×10^{-8}	7.59×10^{-9}	2.36×10^{-12}	2.27×10^{-8}
OCPs	8.52×10^{-9}	3.45×10^{-9}	1.28×10^{-12}	1.20×10^{-8}	4.21×10^{-8}	1.75×10^{-8}	6.34×10^{-12}	5.97×10^{-8}

3.2.3. Health Risk Assessment Based on Monte Carlo

The health risks associated with adult exposure to OCPs in the study area were assessed using Monte Carlo simulation, and the results are presented in Figure 4. As shown in Figure 4a, the 95th percentile values of the HI for δ-HCH, o, p′-DDT, and p, p′-DDT are 1.78×10^{-4}, 5.86×10^{-5}, and 1.02×10^{-4}, respectively, all of which are significantly below the critical threshold of 1. This indicates that the non-carcinogenic health risks posed by these three OCPs to adults are negligible. Figure 4b reveals that the contribution order of the five OCPs to the CR is δ-HCH > p, p′-DDT > o, p′-DDT > p, p′-DDE > p, p′-DDD. The 95th percentile values of the carcinogenic risks for these pollutants are all below the critical threshold of 10^{-6}, suggesting that the carcinogenic risks posed by these OCPs to adult health are also negligible.

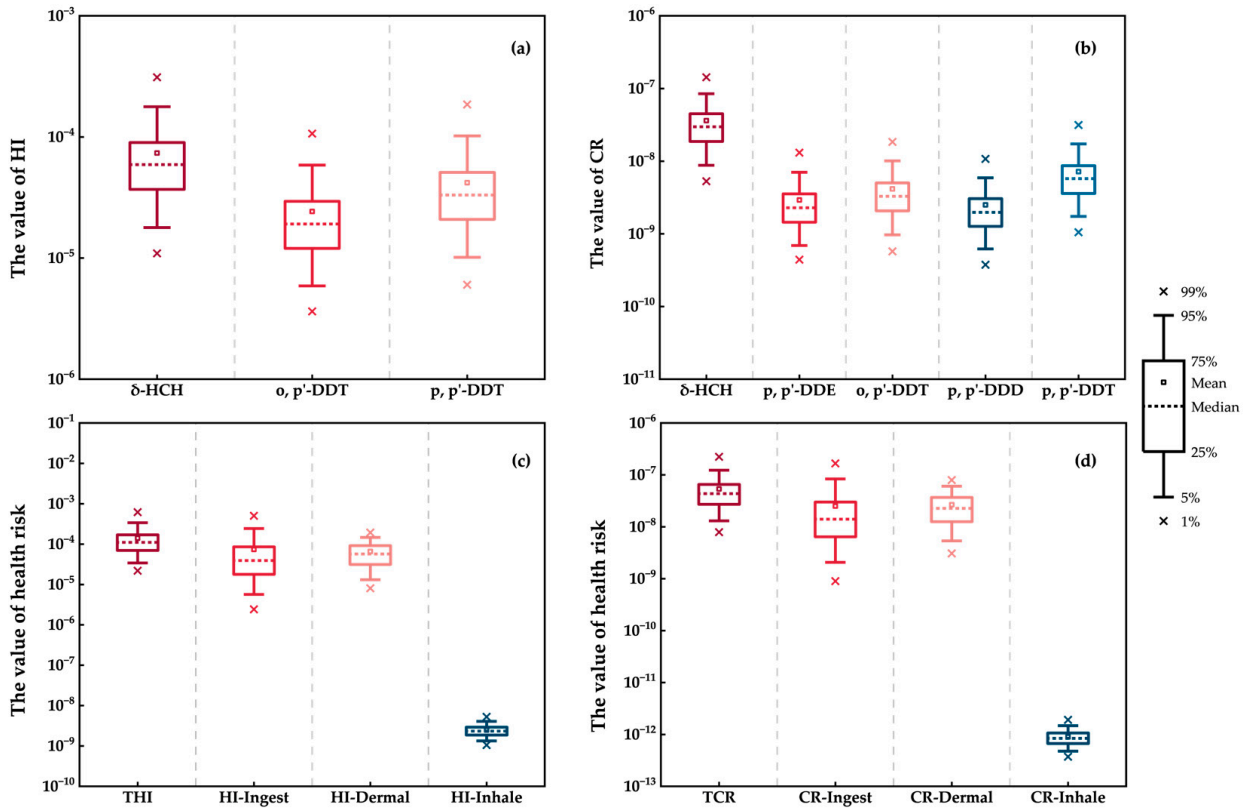

Figure 4. Health risk assessment of soil OCPs by the Monte Carlo simulation. (**a**) Non-carcinogenic risk values for δ-HCH, o, p′-DDT, and p, p′-DDT; (**b**) carcinogenic risk values for δ-HCH, p, p′-DDE, o, p′-DDT, p, p′-DDD, and p, p′-DDT; (**c**) non-carcinogenic risks via three exposure pathways and total risk; (**d**) carcinogenic risks via three exposure pathways and total risk. Note: THI stands for the Total Hazard Index; THQ stands for the Total Carcinogenic Risk.

Figure 4c,d reveals significant differences in health risks for adults across different exposure pathways. For non-carcinogenic risks, ingestion poses a higher risk than dermal contact, while inhalation generates a much lower risk, with the 95th percentile values being 2.43×10^{-4} (ingestion), 1.46×10^{-4} (dermal contact), and 4.09×10^{-9} (inhalation), respectively. For carcinogenic risks, ingestion again dominates with a 95th percentile value of 8.37×10^{-8}, while dermal contact has a 95th percentile value of 6.05×10^{-8}, which is significantly lower than that of ingestion. Inhalation poses the lowest risk, with a 95th percentile value of 1.48×10^{-12}, far below the other two pathways. These results indicate that ingestion is the primary exposure pathway for adults to soil OCPs in the study area.

Further analysis revealed that THI ranged from 8.44×10^{-6} to 2.83×10^{-3}, with the 95th percentile value at 3.39×10^{-4}. These values are markedly beneath the critical threshold of 1, indicating that the non-carcinogenic risks posed by OCPs in the study area soils are extremely low and well within an acceptable range for adults. The range of TCR spans from 2.85×10^{-9} to 1.07×10^{-6}, with the 95th percentile value at 1.23×10^{-7}, which is well below the risk limit of 10^{-6}. Therefore, the carcinogenic risk faced by adults in the study area is negligible. Additionally, the simulation results indicate that there is only a 0.02% probability of TCR falling within the range of 10^{-6} to 10^{-4}, a likelihood so low that it can be disregarded. The application of the Monte Carlo method has significantly enhanced the precision and reliability of risk assessment, while also greatly enriching the dimensionality of risk information. This enables decision-makers to develop more scientific and rational risk management strategies and response measures based on a more comprehensive and detailed risk profile.

3.3. Eco-Risk Assessment

3.3.1. Environmental Quality Assessment

α-HCH, β-HCH, and γ-HCH were not detected in this study, and the risk limits proposed by Urzelai et al. [20] are not suitable for evaluating the ecological risk of δ-HCH in the soil of the study area. Therefore, based on the soil environment quality-risk control standard for soil contamination of agricultural land (GB 15618-2018) [10], the evaluation of HCHs and DDTs in soil was carried out, and the results are shown in Table 5. The content of HCHs and DDTs in the soil is lower than the risk screening value (specified in GB 15618-2018), indicating low pollution risk, and can generally be ignored.

Table 5. Environmental quality assessment of OCPs in soil.

	The Range of Residual Content (μg·kg^{-1})	**Risk Screening Value (μg·kg^{-1})**	**Pass Rate (%)**
HCHs	n.d.–21.52	100	100
DDTs	n.d.–39.16	100	100

3.3.2. Eco-Risk Assessment of DDTs

Table 6 shows that the incidence of adverse effects consistently and markedly increased with increasing concentrations of all compounds. The incidence of effects was 17.39–73.91% in the minimal-effects range and 26.09–82.61% in the possible-effects range. All DDTs that have no probability fall into the probable-effects range except for p, p'-DDT, which has 13.04%. In terms of the total DDTs, the incidence of effects in the possible-effects range exceeds 80%, indicating that occasional adverse biological effects are predicted.

Table 6. Ecological risk assessment of DDTs in soil.

	ERL (μg·kg^{-1})	ERM (μg·kg^{-1})	Range (μg·kg^{-1})	<ERL (%)	ERL~ERM (%)	>ERM (%)
p, p′-DDE	2.2	27	n.d.–9.00	73.91	26.09	0
p, p′-DDD	2	20	n.d.–9.85	56.52	43.48	0
p, p′-DDT	1	7	n.d.–31.01	21.74	65.22	13.04
DDTs	1.58	46.1	n.d.–39.16	17.39	82.61	0

The m-EMR-q values were employed to assess the ecological risk posed by residual DDTs in soil, with results depicted in Figure 5. Among the soil samples, 73.91% were categorized as medium-low risk, exhibiting m-ERM-q values between 0.15 and 0.49. Samples with low ecological risk accounted for 21.74%, including those with undetectable levels of OCPs. Medium-high-risk samples constituted 2.90%, while high-risk samples were minimal at 1.45%, with the highest m-ERM-q value recorded at 1.55.

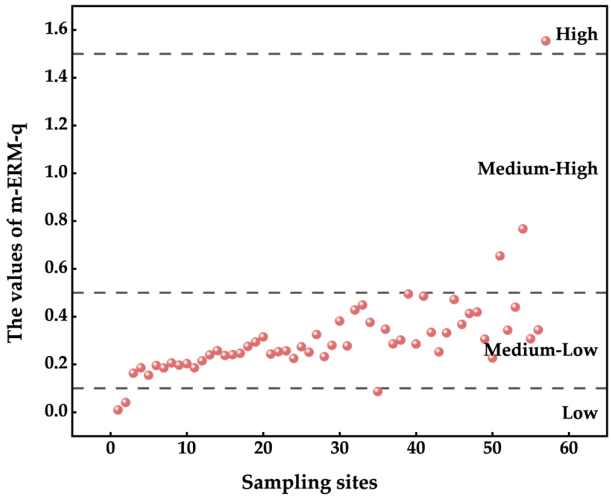

Figure 5. The m-ERM-q values of residual DDTs in the soil of the study area.

Overall, the residual OCPs in the soil of the study area pose a medium-low ecological risk, with DDTs being the primary contributor. To mitigate this risk, precise identification of contamination sources is recommended, such as isotopic fingerprinting analysis and principal component analysis. Secondly, a tiered zoning management system should be implemented based on ERL/ERM thresholds: priority control zones (>ERM) require microbial-nanomaterial co-remediation; key monitoring zones (ERL–ERM) should adopt low-accumulation crop rotation combined with humic acid application to inhibit pollutant uptake; safe zones (<ERL) need enhanced organic fertilizer substitution. Additionally, agricultural management strategies should be optimized by screening low-accumulation crop varieties.

4. Conclusions

In the study area, five OCPs were detected in agricultural soils, with detection rates exceeding 75%. Among these, the residual levels of δ-HCH ranged from n.d. to 21.52 μg·kg^{-1}, with an average concentration of 4.87 μg·kg^{-1}. The content of DDTs (including p, p′-DDT, o, p′-DDT, p, p′-DDE, and p, p′-DDD) varied from n.d. to 39.16 μg·kg^{-1}, with an average content of 10.71 μg·kg^{-1}. Notably, δ-HCH and p, p′-DDT were the primary contributors to the residual contamination levels.

The health risk assessment results demonstrate that even at the maximum exposure levels of OCPs, both the non-carcinogenic risk (1.71×10^{-4}) and the carcinogenic risk (5.97×10^{-8}) are significantly lower than their respective risk thresholds of 1 and 10^{-6}.

Further analysis using Monte Carlo simulation revealed that the 95th percentile values for non-carcinogenic and carcinogenic risks were 3.39×10^{-4} and 1.23×10^{-7}, respectively. These values are well below the risk limits of 1 and 10^{-6}. Therefore, the risks posed by OCPs in the soils of the study area to adults, whether non-carcinogenic or carcinogenic, are considered negligible.

In comparison with the soil environmental quality standard (specified in GB 15618-2018) [10], the concentrations of HCHs and DDTs in the study area's soil were found to be below the risk screening values. Subsequent ecological risk assessment revealed that the vast majority of soil samples exhibited medium-low ecological risk, with DDTs being the primary contributor to ecological risk.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/land14030612/s1, Table S1: Parameters used in the non-cancer risk and cancer risk assessments; Table S2: Slope Factor (SF) and Reference Dose (RfD) for OCPs [48]; Table S3: Details of the parameters of the Monte Carlo distribution [49].

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