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Mapping pesticide residues in soil for China: Characteristics and risks

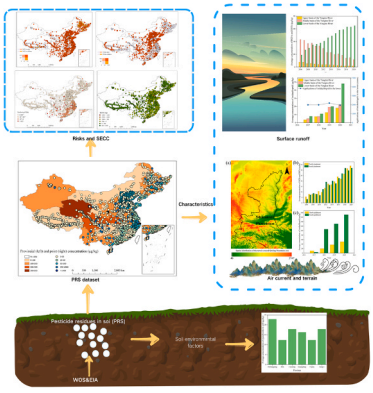
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HIGHLIGHTS

- The dataset of pesticide residues in soil (PRS) was developed.
- Organochlorine pesticide residues were significant contributors to PRS.
- Increased soil moisture, temperature and acidity lower PRS levels.
- Pesticides in watershed soil migrate downstream through surface runoff.
- Pesticides in the soil deposited in piedmont soil through airflow transport.

GRAPHICAL ABSTRACT



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ABSTRACT

The widespread application of pesticides in China has led to the accumulation of residues in soil. However, few regional studies have fully elucidated the characteristics of pesticide residues in soil (PRS) and the associated risks to the ecosystem and human health on a national level. Therefore, this study aims to compile a dataset on PRS in China from 2006 to 2020 and analyze the interactions and impacts between PRS and the environment. The average concentration of PRS in China was 243.96 μg/kg which was lower than the levels reported in Euro-Americans and other nations. This study revealed PRS in China predominantly originates from organochlorine pesticide residues, with DDTs and HCHs being significant contributors. Despite the high intensity of pesticide application in the Southeast China, PRS concentrations were comparable to those in the Northeast, due to environmental factors that favor pesticide degradation in the Southeast. Both legacy and in-use pesticides were transported by surface runoff or air current, resulting in their accumulation in soil of the lower Yangtze River basin or the piedmont soil of Qinling Mountains, respectively. The average soil environment carrying capacity of PRS in China was −69.5 kg. The ecological risk contributed by PRS in China was mainly at a negligible level. Carcinogenic risks of PRS to adults (4.6×10^{-4}) and children (6×10^{-4}) exceeded the tolerable thresholds (10^{-5}) by a small margin.

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1. Introduction

Pesticides play a vital role in agricultural production [1]. With their continuous and extensive application, much attention has been given to pesticide residues in soil (PRS). PRS not only poses a direct threat to soil organisms, birds and plants but also jeopardizes the stability of the soil environment and potentially harms human health. The complexity of pesticide residue in the soil is exacerbated by the mixed application of different pesticides. As reported by the Food and Agriculture Organization (FAO), China ranks among the top three countries globally in terms of pesticide application [2]. The large amount and variety of pesticides applied in China have raised concerns and prompted investigations into their effects on the soil environment.

High levels of dichlorodiphenyltrichloroethane (DDTs) and hexachlorocyclohexane (HCHs) were found in the soil of the Pearl River Delta (PRD), potentially endangering local ecosystems [3]. Moreover, the presence of organophosphorus (OPs) and pyrethroid pesticide residues in the soil of the PRD exceeded 1000 μg/kg, with dimethoate residues being the most prevalent, leading to an increase in the non-carcinogenic risk (NCR) for children [4]. The NCR of neonicotinoids (NEOs) in the PRD soil to humans was deemed insignificant but posed a toxic threat to earthworms [5]. Conversely, the levels of organochlorines (OCPs) in the soil of the Yangtze River Delta (YRD) were found to be below China's soil quality standards, indicating a low risk [6]. It was reported that pesticide application in Tianjin was the main cause of pesticide accumulation in soil [7]. Despite being banned in China for many years, DDTs and HCHs were discovered in soil in central China, posing potential exposure risks [8]. Additionally, researchers have reported that in Northeast China, which accounts for 20 % of the national cultivated land, the OCPs in the topsoil were greater than those in other parts of China [9]. Despite lower levels of cultivated land and pesticide applications in Tibet, pesticide residue levels in the local soil remained elevated, with a maximum recorded level of 925 μg/kg [10].

While these findings have advanced our understanding of spatiotemporal pesticide residue levels in China and their environmental implications, their limited scope has resulted in fragmented outcomes. As a result, these findings do not offer a comprehensive overview of pesticide residue levels and their effects on the soil environment in China. The absence of information and data has led to a lack of knowledge regarding soil carrying capacity and ecological and health risks in China.

This study gathered PRS samples from the Web of Science and environmental impact assessment reports to construct a PRS dataset specific to China. Utilizing this dataset, we conducted an analysis of environmental factors influencing the distribution of PRS in China, evaluated the soil pesticide carrying capacity and assessed the potential risks posed by PRS to ecosystems and human health. The PRS database and the environmental impact assessment conducted in this study offer valuable scientific insights that can help policymakers develop more evidence-based and rational management strategies for pesticides and soil.

2. Materials and methods

2.1. Data collection

PRS samples used in this study were obtained from the Web of Science (WOS) and Chinese Environmental Impact Assessment (EIA) reports. Searches for pesticide residues in Chinese soil were conducted using specific keywords ("soil", "pesticide" and "China") in the WOS up to September 22, 2023. The screening criteria included the exclusion of preprints and reviews, as well as sampling time and site parameters restricted to the years 2006–2020 in China. Data on pesticide residues from Chinese EIA reports were extracted by downloading reports from the websites of provincial departments of ecology and the environment in China covering the period from 2006 to 2020. The papers and websites used are shown in Table S (1−2).

2.2. Data cleaning and extraction

References and reports contained PRS data at various soil depths. This study took data cleaning procedures to retain only topsoil data. There were two types of reference data: one with precise latitude and longitude coordinates and the other lacking such information. Leveraging a geographic information system (GIS), this paper relocated the sample sites with latitude and longitude in the image and exported a file containing latitude, longitude and concentration information. For reference data lacking specific site location information, the study averaged it with other data as regional averages. Other types of data (e.g., tables, words and PDFs) can be used directly without conversion.

2.3. Data processing

This study gathered a total of 185 pesticides from the reference document and categorized them into insecticides, herbicides and fungicides (Table S3). The concentrations of PRS in the reference were presented as the maximum, minimum, median, quartile and average values. In cases where the average value was not provided, it can be derived from the available data. The application of averages aids in evaluating the potential ecological and health risks associated with PRS.

2.4. Analysis of other environmental factor datasets and PRS data

The soil total nitrogen density (TN), soil total phosphorus (TP), soil total potassium (TK), soil organic matter (SOC), cation exchange capacity (CEC) and pH data utilized in this study were sourced from previous research [11,12]. Soil humidity and surface net radiation data were obtained from previous studies [13–15]. The surface wind speed data were from a previous study [16]. The terrain data were from the National Geospatial-Intelligence Agency [17]. This study subsequently integrated both PRS and environmental factor datasets into the GIS to derive various environmental factors corresponding to each PRS site. Finally, analyses can be conducted between pesticide residue data and environmental factors.

2.5. Methods for assessing ecological risk

Risk quotients (RQ) were used to evaluate the ecological risk of PRS in China. The method for RQ calculation was from the Environmental Protection Agency (EPA) [18], and it can be determined by Eq. (1).

$$RQ_i = \frac{MEC_i}{PNEC_i} \tag{1}$$

where RQ_i indicates the RQ value of the ith pesticide, MEC_i (μg/kg) is the concentration of the ith pesticide in the soil, and $PNEC_i$ (μg/kg) is the predicted no-effect concentration of the ith pesticide. RQ values are divided into four categories: negligible (RQ <0.01), low ($0.01 \leq$RQ <0.1), medium ($0.1 \leq$RQ <1) and high risk (RQ ≥ 1) [19].

The PNEC can be calculated by Eqs. (2) or (3).

$$PNEC_i = \frac{NOEC_i \text{ or } NOEL_i}{AF} \tag{2}$$

$$PNEC_i = \frac{EC_{50i} \text{ or } LC_{50i}}{AF} \tag{3}$$

where $NOEC_i$ (μg/kg), $NOEL_i$ (μg/kg), $EC50_i$ and $LC50_i$ represent no observed effect concentration, no observed effect level, half-maximal effective and half-maximal lethal, respectively. AF is the assessment factor, and it can be 10, 50, 100 or 1000 under different trophic levels and toxicity conditions. In accordance with a previous study [20], this paper selects 1000 as the AF value. The toxicity data ($NOEC_i$, $NOEL_i$, $EC50_i$ and $LC50_i$) are from the EPA [18] and the Pesticide Properties DataBase (PPDB) [21]. To best reflect ecological systems, terrestrial

organisms (i.e., honeybees, earthworms and birds) are considered.

Soil often contains residues of a variety of pesticides. To evaluate the ecological risk of the PRS, the ecological risks of pesticides detected at the sampling points were calculated.

To assess the ecological risk of PRS for a site, RQ_i for each pesticide residue calculated in the soil sample is summed [22] as shown in Eq. (4).

$$RQ_{site} = \sum_{i=1}^{n} RQ_i \tag{4}$$

where RQ_{site} is the sum of the ecological risks of all pesticides at the same sampling site, and its evaluation criteria are the same as those of RQ_i, which are divided into four levels.

2.6. Methods for calculating the SECC

The soil environment carrying capacity (SECC) for PRS in China is determined by assessing the soil environment static capacity, defined as the maximum pesticide residue load that can be sustained by the soil without posing an unacceptable ecological risk [22]. It can be evaluated by Eq. (5):

$$SECC_i = d \times \rho \times A \times (S_i - MEC_i) \times F_1 \times F_2 \times F_3 \tag{5}$$

where SECC represents the soil environment carrying capacity; d and ρ represent the soil depth (m) and bulk density (kg/m^3), respectively; and A is the cultivation area at the sample site (1 km^2). S_i and MEC_i represent the soil residue standard and sample concentration (mg/kg) of the ith pesticide, respectively. Since 185 pesticides were included in this study and most of the pesticides currently have no residue standard values, the average value of the whole pesticide residue was used instead of S_i. The evaluation of SECC involved three key factors: soil attenuation capacity (F_1), cumulative rate (F_2) and risk coefficient (F_3).

$$F_1 = 10 - (\omega_1 \left|\log\frac{SOC_{obs}}{SOC_{ref}}\right| + \omega_2 \left|\log\frac{pH_{obs}}{pH_{ref}}\right| + \omega_3 \left|\log\frac{CEC_{obs}}{CEC_{ref}}\right| + \omega_4 \left|\log\frac{CLA_{obs}}{CLA_{ref}}\right|) \tag{6}$$

where F_1 depends on the natural attenuation capacity (NAC) of the soil, which has been widely calculated using the ecosystem-service performance index [23]. The subscripts obs and ref represent observations and reference values of environmental factors, respectively. These values can be obtained through national surveys conducted by authoritative authorities. The obs and ref values of this study are from Shangguan et al. [15] and previous studies [11,12], respectively. ω is the contribution of the corresponding parameters to NAC [22]. To quantify the performance of NAC, four core evaluation variables [22] were used: SOC, pH, CEC and clay (CLA).

$$F_2 = 1 - \frac{aps}{d \times \rho \times A \times (S_i - MEC_i)} \tag{7}$$

where F_2 is the ratio of the annual cumulative number of pesticides in the soil to the soil environment static capacity. aps represents the annual cumulative amount of pesticides in soil (kg/ha), which was obtained from a previous study [24].

$$F_3 = 1 - RQ \tag{8}$$

where F3 is determined by the potential environmental risk of pesticide residues.

2.7. Methods for Health Risk Assessment

For this study, the EPA human health risk assessment methodology was followed. This method has been used extensively in the field [25,26].

Health risk assessments were conducted to quantify the health risks (carcinogenic and non-carcinogenic) for children and adults exposed to soil via ingestion, dermal contact and inhalation exposure pathways. Eqs. (9)–(11) show the specific calculation process.

$$ADD_{ing} = \frac{MEC_i \times IngR \times CF \times EF \times ED}{BW \times AT} \tag{9}$$

$$ADD_{inh} = \frac{MEC_i \times InhR \times EF \times ED}{PEF \times BW \times AT} \tag{10}$$

$$ADD_{derm} = \frac{MEC_i \times AF \times CF \times SA \times ABS \times EF \times ED}{BW \times AT} \tag{11}$$

where the subscripts ing, inh and derm represent the exposure pathways of ingestion, dermal contact and inhalation, respectively; ADD is the average daily exposure dose (mg/kg-day); and MEC_i is the pollutant concentration in a particular exposure medium (mg/kg). For adults, the ingestion (IngR, mg/day) and inhalation frequencies of soil (InhR, m^3/day) were 200 and 15, respectively and the corresponding values for children were 100 and 7.5 [27]; PEF is the particle emission factor (m^3/kg) and sourced from the EPA [28]; SA is the surface area of exposed skin (cm^2); AF is the adhesiveness degree of skin (mg/(cm$_2$/d)); and BW is body weight (kg). The values of SA, AF and BW were obtained from a previous study [25]. ABS is the absorption factor of skin and can be found in PPDB. ED is the exposure duration (year). EF is the exposure frequency (day/year). CF is the conversion factor (kg/mg). AT is the average exposure time.

The NCR values of PRS can be calculated by Eqs. (12) and (13).

$$HQ_i = \frac{ADD_i}{REF_i} \tag{12}$$

$$HQ_T = \sum HQ_i \tag{13}$$

where HQ_i is the non-carcinogenic risk of the ith pesticide; ADD_i is the average daily exposure dose of the ith pesticide; REF_i is the reference dose for the non-carcinogenic ingestion of the ith pesticide, which is derived from PPDB; and HQ_T is the total non-carcinogenic risk. $HQ_T > 1$ indicates that the pollutant causes health risks to humans, and $HQ < 1$ indicates that the pollutant causes negligible health risks to humans [29].

The CR values of PRS can be calculated by Eqs. (14) and (15).

$$CR_i = ADD_i \times SF_i \tag{14}$$

$$CR_T = \sum CR_i \tag{15}$$

where CR_i is the carcinogenic risk quotient for the ith PSR. SF_i is the carcinogenic slope factor of the ith PSR, and the SF values are from the EPA [30]. Pesticides with slope factors accounted for only 8.5 % of the total pesticides. Representative organochlorine pesticides, DDTs and HCHs (accounting for 70 % of the pesticides with slope factors) were selected to assess the corresponding CR values. CR_T is the total carcinogenic risk quotient. If the CR is less than 10^{-5}, human health risks are acceptable [25].

3. Results and discussion

3.1. Characteristics of PRS in China

Using PRS data sourced from references and reports, this study constructed a PRS dataset specific to China spanning the years 2006 to 2020. The mean PRS concentration in China was calculated to be 243.96 μg/kg (Fig. 1a). The provinces of Qinghai, Sichuan, Hubei, Tianjin, Chongqing, Jiangsu, Beijing, Liaoning, Heilongjiang and Xinjiang exhibited the high average PRS concentrations, with values of 578.7, 300.0, 296.7, 279.8, 234.1, 202.5, 148.4, 139.3, 134.3 and 121.6 μg/kg, respectively. Pesticide applications [24] in Qinghai,

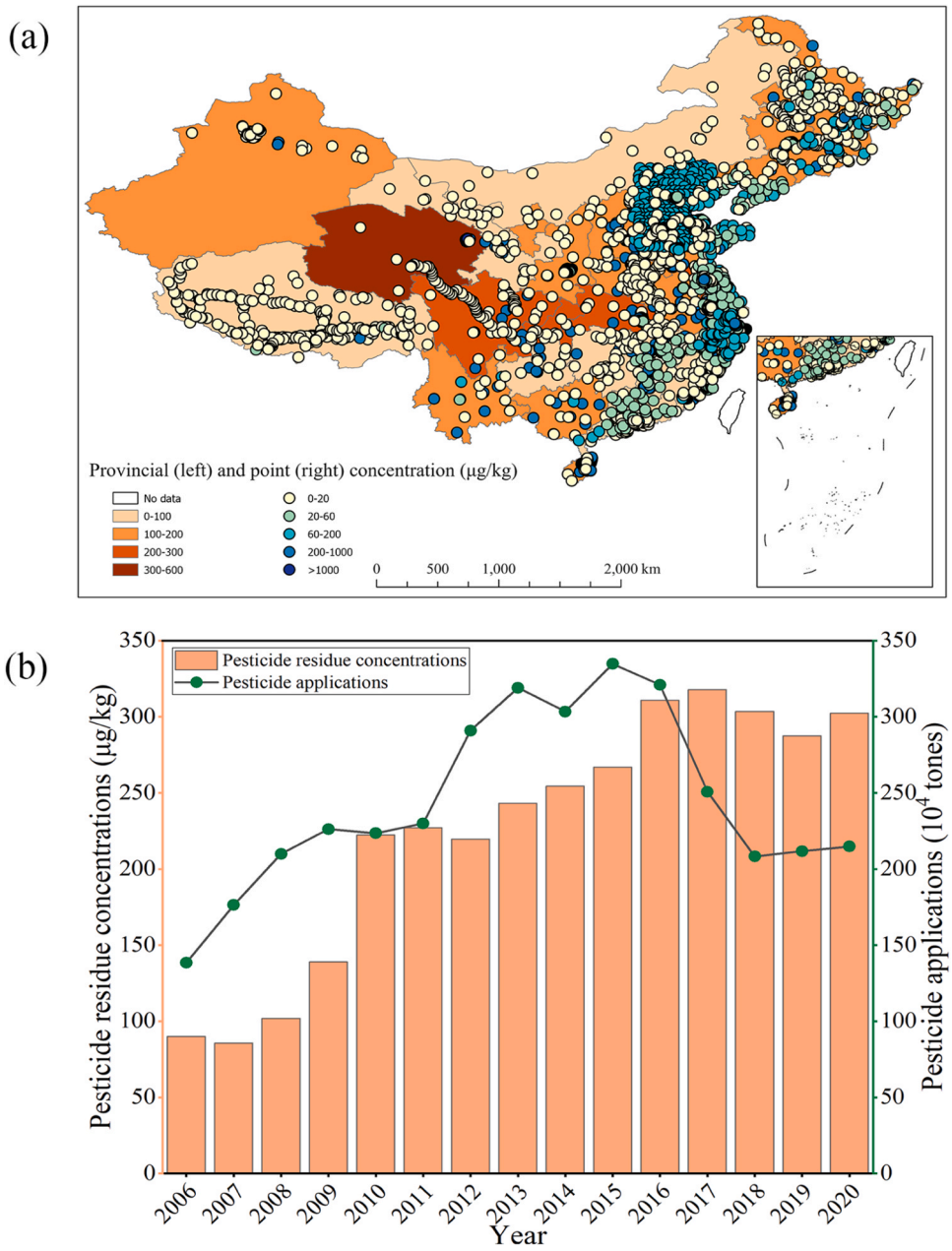


Fig. 1. Spatiotemporal distributions of PRS in China: (a) Spatial distribution of PRS in China; (b) Change curves of pesticide applications and average levels of PRS from 2006 to 2020.

Beijing and Tianjin were lower than those in other regions in China, yet the PRS levels were greater. These findings suggested that PRS accumulation in China is influenced not only by pesticide application but also by local environmental factors, soil capacity and other variables. The average PRS levels in various regions of China ranged from 35.7 to 96.9 μg/kg, with significantly higher levels observed in the eastern region compared to the western region, as indicated by the fourfold difference in numbers of sampling points and the twofold difference in average PRS levels. The sampling sites on the eastern coast comprised 53.4 % of the total sampling points, with an average concentration of 70.4 μg/kg, while the sampling points in the western region accounted for 11.7 % of the total sites, with an average concentration of 31.2 μg/kg. The sites exhibiting PRS levels exceeding 1000 μg/kg were located primarily in the industrial parks surrounding Tianjin and Qinghai provinces, with an average concentration of 1392.3 μg/kg. This phenomenon can be attributed to the presence of enterprises within these parks that are engaged in the production of chemical intermediates for pesticides, leading to elevated PRS concentrations. Sampling sites with concentrations ranging from 200 to 1000 μg/kg were uniformly distributed across all provinces of China, comprising 9.8 % of the total sample sites, with an average concentration of 279.7 μg/kg. In comparison, sites with concentrations of 20 to 60 μg/kg and 0 to 20 μg/kg accounted for 15.2 % and 48.7 % of the total sites, respectively, with average concentrations of 32.3 μg/kg and 5.8 μg/kg. The global average concentration of PRS was reported to be 620 μg/kg [31]. In 2015, the maximum and median levels of PRS in Europe were reported to be 28700 μg/kg and 1500 μg/kg, respectively [32]. During the same time frame, the levels of PRS in France [33] and Poland [34] ranged from 12.4 to 1724 μg/kg and 0.01 to 1031.6 μg/kg, respectively. Additionally, a PRS concentration of 338.7 μg/kg was detected in Alabama, USA [35]. These regions share similar latitudes with parts of China, where the concentration of PRS was lower. In the 1940s, pesticides were

applied not only in agriculture but also for disease control in Euro-American countries [36], leading to a significant accumulation of residues in the environment. This may explain why PRS in Euro-American countries were greater than those in China. Compared to Euro-American countries, China has the lowest pesticide use per unit area at 52 kg/km^2 [2], which helps understand why low levels of PRS occurred in China. The PRS levels in northern India [37] in 2017 varied from 540 to 37,420 μg/kg, surpassing those in China. Rajendran et al. proposed that pesticides in the air from the Indian tropics may be carried to colder regions and rapidly deposited into the soil, leading to elevated PRS concentrations [38].

The relationship between pesticide application [39] and the average PRS concentrations in China from 2006 to 2020 was shown in Fig. 1b. Pesticide application exhibited an increasing trend from 2006 to 2016, followed by a decreasing trend from 2016 to 2020. Pesticide application in 2016 was 2.4 and 1.5 times greater than those in 2006 and 2020, respectively. Similarly, the PRS levels mirrored the trend of pesticide application during the same period, with the PRS in 2016 being 3.4 times greater than that in 2006, before decreasing to 302.3 μg/kg in 2020. This can be attributed to the fact that after decades of pesticide applications, China finally realized the environmental impact of pesticides and proposed the "Program of Zero Growth in Pesticides Use" [40] in 2015, resulting in decreases in pesticide application and PRS levels after 2015.

This study examined the characteristics of various PRS at the provincial level (Fig. 2a). For instance, soil insecticide residues in Sichuan accounted for 91.4 % of the PRS, while the residues in Shaanxi accounted for only 26.5 % of the PRS. Similarly, Gansu had the highest percentage of soil fungicide residues at 48.0 %, while Qinghai had a percentage of 2.9 %. Chongqing had the highest share of soil herbicide residues at 48.0 %, whereas Jilin had the lowest at 0.04 %. In the eastern and southern coastal provinces of China, the PRS was predominantly composed of insecticides and fungicides, with soil insecticide residues representing more than 50 % of the total PRS. Nationally, insecticides accounted for 57.5 % of the PRS, followed by fungicides at 29.2 % and herbicides at 13.3 %. These findings suggested that insecticides were the primary contributors to PRS concentrations in China. Subsequently, insecticides were categorized [24] into various classes, including organochlorine, organophosphorus, carbamates, pyrethroids, neonicotinoids, diamides, acaricides and other pesticides, to determine their respective contributions to insecticide residues. Specifically, the levels of organochlorine, organophosphorus, carbamates, pyrethroids, neonicotinoids, diamides, acaricides and other pesticides in soil represented 42.8 %, 11.0 %, 1.2 %, 15.7 %, 9.7 %, 4.1 %, 1.4 % and 14.1 % of the total insecticide concentrations in China, respectively (Fig. 2b). DDTs and HCHs, as persistent organic compounds in organochlorine pesticides, have garnered global attention. Despite DDTs and HCHs have being prohibited for production and application since the last century, a recent study [6] has identified their presence in soil. This paper highlighted that DDTs (including o,p′-DDE, p,p′-DDE, o,p′-DDT, p,p′-DDT, o,p′-DDD and p,p′-DDD) and HCHs (comprising α-HCH, β-HCH, γ-HCH and δ-HCH) constituted 75.6 % and 23.1 % of the organochlorine residues, respectively. The regions of Hubei, Beijing, Guangdong, Shanghai, Hubei and Chongqing exhibited the highest levels of o,p′-DDE (22.9 %), p,p′-DDE (45.6 %), o,p′-DDT (26.2 %), p,p′-DDT (62.0 %), o,p′-DDD (3.0 %) and p,p′-DDD (51.9 %) residues (Fig. 2c). Conversely, Qinghai had the lowest shares of p,p′-DDE (1.1 %), o,p′-DDT (1.3 %), o,p′-DDD (0.01 %) and p,p′-DDD (0.2 %). Tianjin and Hubei had the lowest shares of o,p′-DDE and p,p′-DDT at 0.02 % and 0.13 %, respectively. The shares of α-HCH (0.1 %) and γ-HCH (0.2 %) were the lowest in Ningxia, and the shares of β-HCH (2.0 %) and δ-HCH (0.1 %) were the lowest in Henan and Yunnan, respectively (Fig. 2d). Taken together, the PRS in China

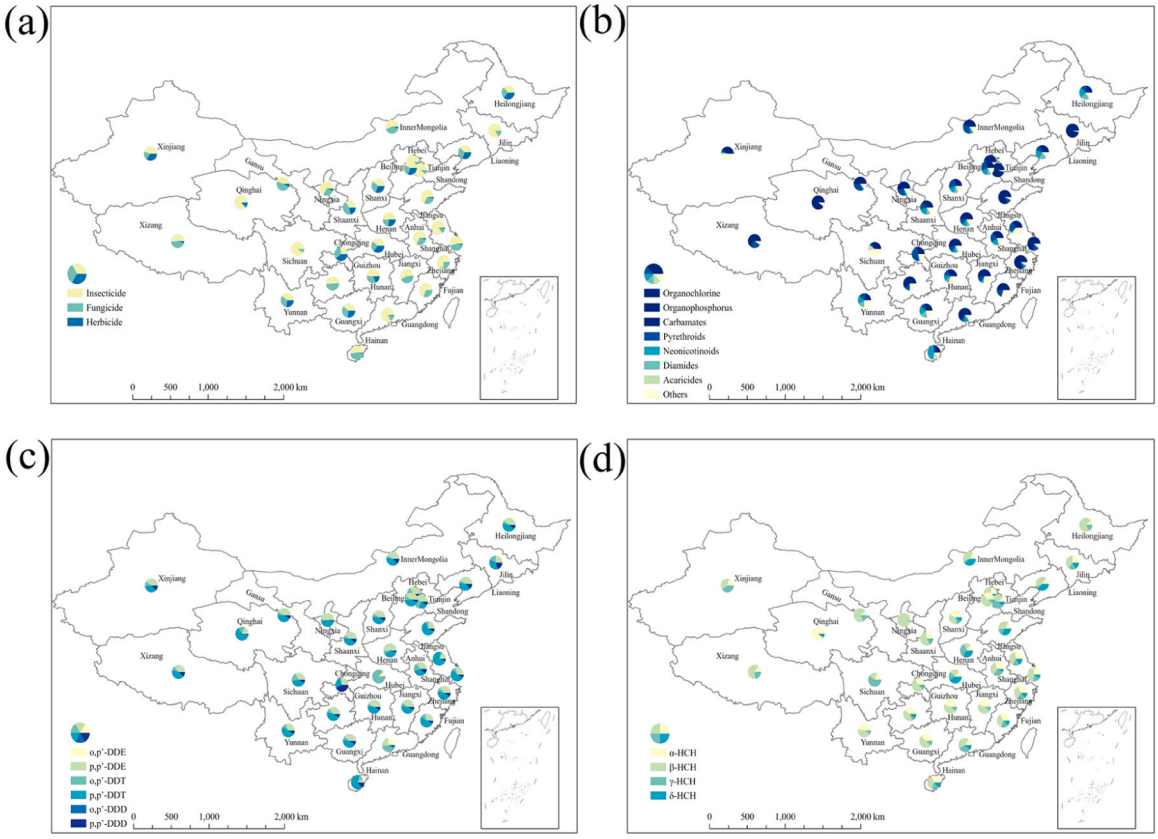


Fig. 2. Distributions of PRS in provinces: (a) Distributions of insecticides, fungicides and herbicides; (b) Distributions of organochlorine, organophosphorus, carbamates, pyrethroids, neonicotinoids, diamides, acaricides and other pesticides, respectively; (c) Distributions of o,p′-DDE, p,p′-DDE, o,p′-DDT, p,p′-DDT, o,p′-DDD and p,p′-DDD, respectively; (d) Distributions of α-HCH, β-HCH, γ-HCH and δ-HCH, respectively.

mainly originates from organochlorine pesticide residues, especially from DDTs and HCHs. In fact, the residues of these two pesticides were attributed to their historical application. The Chinese government has acknowledged this issue and implemented a set of regulations to oversee the levels of DDTs and HCHs in soil, with the aim of protecting public health and ecological stability. For example, the law [41] requires that the concentrations of DDTs and HCHs in the soil should be determined prior to the project construction. Additionally, residues of the two pesticides in the soil were identified during the implementation of the Three Lines One Permit [42]. Chinese governments should therefore pay attention to establishing more national monitoring plans for PRS, especially for DDTs and HCHs.

3.2. Comprehensive analysis on PRS and corresponding environmental factors

This study investigated the effects of soil environmental factors on PRS levels in representative regions of China. For instance, the application intensity of pyrethroids (in-use pesticides) in the Southeastern provinces (Guangdong, Fujian and Jiangxi) was 1.3 times higher than that in the Northeastern provinces (Heilongjiang, Jilin and Liaoning) [24]. Nevertheless, the average soil residual levels of pyrethroids in the Southeast (15.48 μg/kg) and Northeast (17.68 μg/kg) were found to be comparable (Fig. 3). This can be attributed to local soil environmental factors. The average SOC in Southeast China was 0.71 g/kg, lower than that in the Northeast. Lower SOC values in Southeast China may hinder soil adsorption of pesticides [43]. In the case of low soil adsorption, pyrethroids were susceptible to degradation through extensive interactions with various environmental factors present in the soil. In comparison to the water content of 24.67 % in Northeastern soil, the higher water content of 35.60 % in Southeastern soil has been found to enhance the hydrolysis of pesticides [44]. The soil pH values in the Northeast and Southeast regions are measured at 7.12 and 5.77, respectively [11,12]. It is noteworthy that the acidity of soil in Southeast regions promotes the hydrolysis of pyrethroids. Additionally, the average thermal radiation in the Southeast was found to be 1.4 times [14] higher than that in the Northeast, leading to an increase in soil temperature that facilitates pesticide vapor diffusion [45] and degradation processes [46]. This may explain why the greater intensity of pyrethroid application occurred in the Southeast, yet the observed residual levels were comparable to those in the Northeast.

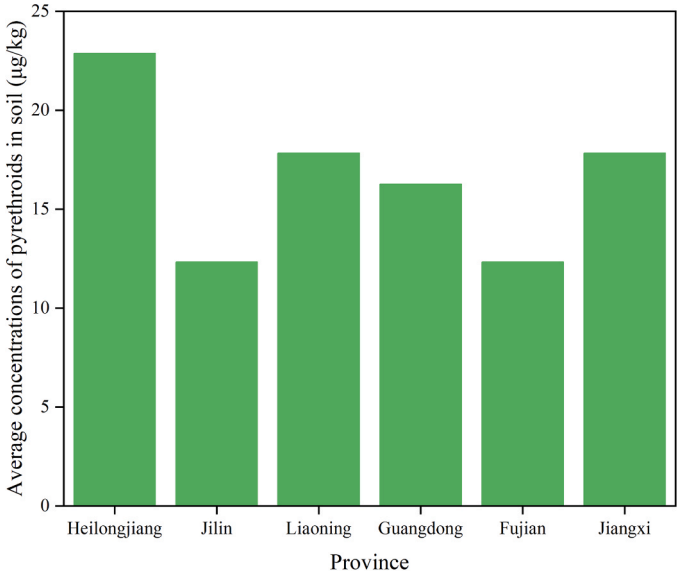


Fig. 3. Average concentrations of soil pyrethroids in the Southeastern and Northeastern provinces.

The cultivated land area of the Yangtze River basin accounts for 24 % of the national cultivated area and supports 34.2 % of the total population. This study examined the impact of surface runoff on the transport of HCHs and DDTs in Yangtze River basin. It was found that the average levels of HCHs and DDTs in the soil of the upper and middle Yangtze River basins showed a declining trend from 2006 to 2020 (Fig. 4a). The decreased levels of HCHs and DDTs in the soil of the middle basin were larger than those in the upper basin. It can be attributed to the less cultivated land and a previous lower application of HCHs and DDTs in the upper basin. Conversely, the concentrations in the lower basin soil have been increasing during this period. Some studies revealed that the surface runoff in the middle and upper river basins resulted in a higher concentration of pesticide residue in soil of the lower basin [47,48]. Therefore, it can be concluded that the enrichment of HCHs and DDTs in the soil of the lower Yangtze River basin is due to surface runoff transporting them from the upper and middle basins. The concentrations of imidacloprid (an in-use pesticide) in the soil of Yangtze River basin showed an increasing trend (Fig. 4b). The largest increase was recorded in the lower basin, reaching 40.13 μg/kg. However, the application of imidacloprid in the lower basin has been stable in recent years (Fig. 4b). It can be inferred that the in-use pesticide in soil was also transported by surface runoff and accumulated in downstream soil.

Qinling Mountains serve as a geographical barrier dividing China into northern and southern regions, leading to distinct variations in atmospheric transport patterns and terrain conditions on either side (Fig. 5a). The Qinling Mountains region is dominated by northwest winds in winter and spring, and southeast winds in summer and autumn. Observations indicate that the annual average wind speeds in the north and south piedmonts of Qinling Mountains are lower compared to those in the corresponding areas far from the mountains [16]. The residues of HCHs and DDTs in soil on both piedmonts of Qinling Mountains showed an upward trend from 2008 to 2020 (Fig. 5b). Literature reported that HCHs and DDTs are easily transported by downwind air current [49–51]. Therefore, HCHs and DDTs were transported by the wind and subsequently blocked by Qinling Mountains, causing them to accumulate in the soil of both piedmonts. On the other hand, the concentrations of soil imidacloprid residue in both piedmonts of Qinling Mountains showed an increasing trend (Fig. 5c). The imidacloprid residue in the south piedmont soil were larger than that in the north piedmont soil. To sum up, the transportation of both legacy and in-use pesticides was influenced by atmospheric and terrain conditions.

3.3. SCEE of PRS in China

This study utilized environmental factors and the PRS to assess the SECC. Generally, high SECC values indicate the high soil environment capacity [52]. The numbers of SCEE values ranged −3000 to −1500 , −1500 to −1000 , −1000 to −500 , −500 to 0 , 0 to 500 and 500 to 1500 kg accounted for 0.26 % , 0.06 % , 63.8 % , 32.6 % and 3.28 % of the total, respectively (Fig. 6a). With an average SECC of −69.5 kg, soil in China may not afford the current intensity of pesticide application. The average SECC values in regions such as Heilongjiang, Jilin and Tibet were above 0, indicating that pesticide application did not threaten the soil environmental carrying capacity. However, in regions such as the North China Plain (NCP), YRD, PRD, Yunnan, Guizhou and Sichuan, the application of pesticides may threaten the local soil environment, as the SECC values were less than 0.

3.4. Ecological and health risk assessment of the PRS

As shown in Figs. 6b, 69.4 %, 14.0 %, 11.3 % and 5.3 % of the total samples exhibited negligible, low, medium and high risk, respectively. The application of pesticides has been reported to threaten ecosystems in Czech Republic [53] and Argentina [54]. Compared to these countries, the ecological risk of PRS in China was negligible. Negligible risk in Tibet, Inner Mongolia and Ningxia can be attributed to their low

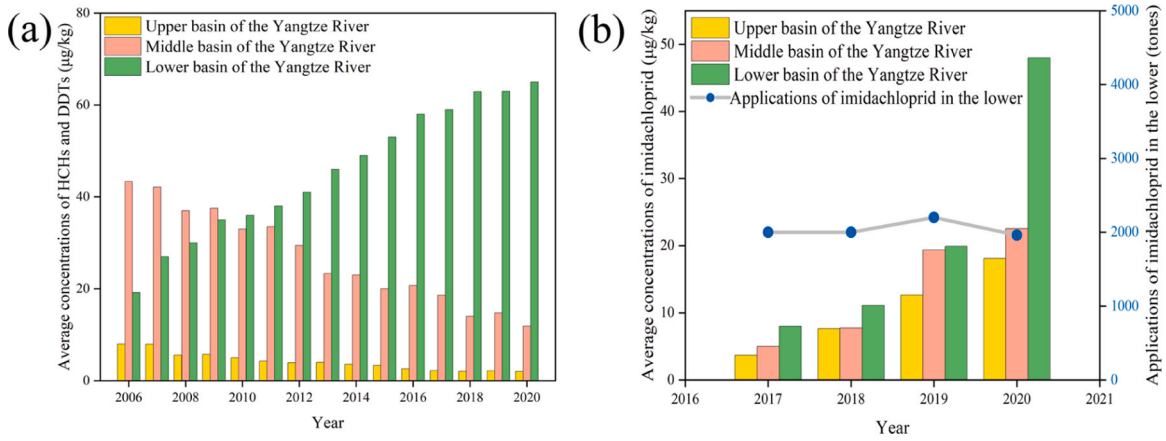


Fig. 4. (a) Average concentrations of HCHs and DDTs in the Yangtze River basins; (b)Average concentrations of imidacloprid in the Yangtze River basins and imidacloprid applications in the lower of the Yangtze River.

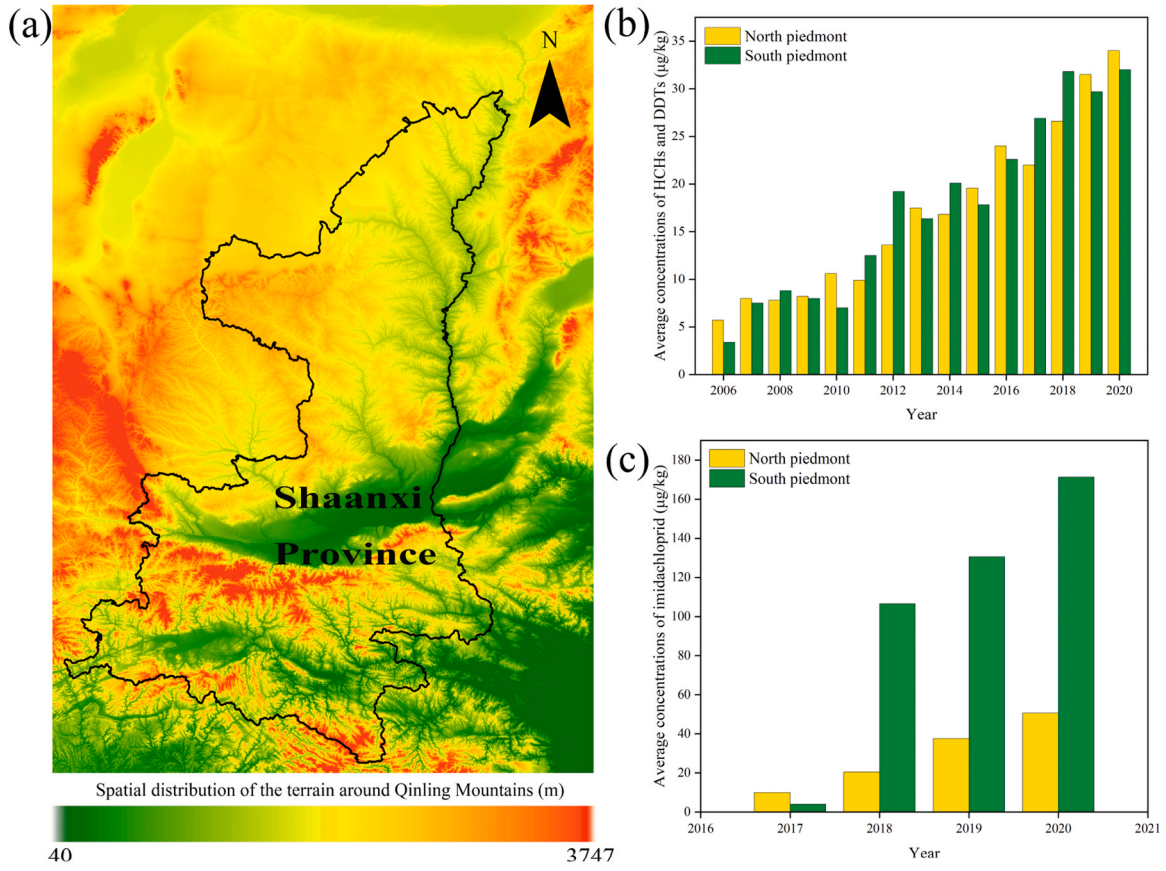


Fig. 5. (a) Spatial distribution of terrain around Qinling Mountains; (b) Distribution of soil HCHs and DDTs concentrations in north and south piedmonts; (c) Distribution of soil imidacloprid concentrations in north and south piedmonts.

pesticide application and good ecological protection. The regions at low risk encompassed the North China Plain (NCP), while the PRS posed a medium risk to the PRD and YRD. Northeast, Northwest, Central and Southwest China were classified as high ecological risk areas in China. The risk distributions demonstrated the varying effects of PRS on distinct regional ecosystems. The State Council has issued the "Opinions on Strengthening Ecological Zoning Controls" [55] to elucidate the environmental impacts of chemicals, including PRS. It may further reduce the ecological impact of PRS.

Birds, earthworms and honeybees are integral components of ecosystems. The data presented in Fig. 6c illustrated the average and maximum RQ values for each species. The RQ values for birds, earthworms and honeybees accounted for 58.8 %, 17.8 % and 23.4 %, respectively, of the total RQ. Specifically, the average RQ values for birds, earthworms and honeybees were 9.8×10^{-4}, 3.0×10^{-3} and 0.15, respectively. Accordingly, the maximum RQ values for birds, earthworms and honeybees were 0.04, 0.25 and 6.8, respectively.

This study examined the RQ of various pesticides, specifically focusing on their impacts on birds, earthworms and honeybees. The combined RQ values of carbamates, organochlorine, organophosphorus, pyrethroids and neonicotinoid pesticides accounted for more than 90 % of the total RQ values for honeybees (Fig. S1). Additionally, the RQ

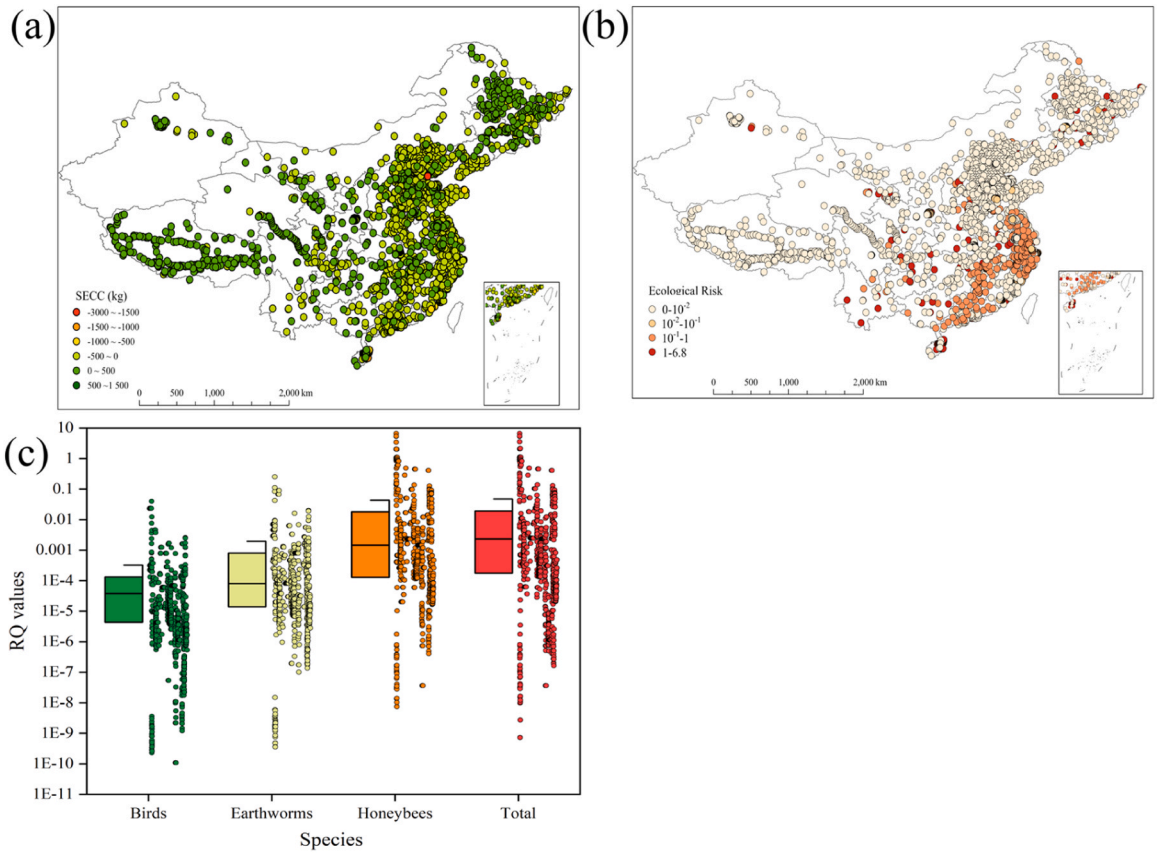


Fig. 6. (a) SECC values across China; (b) Ecological risks in China; (c) Ecological risks for species.

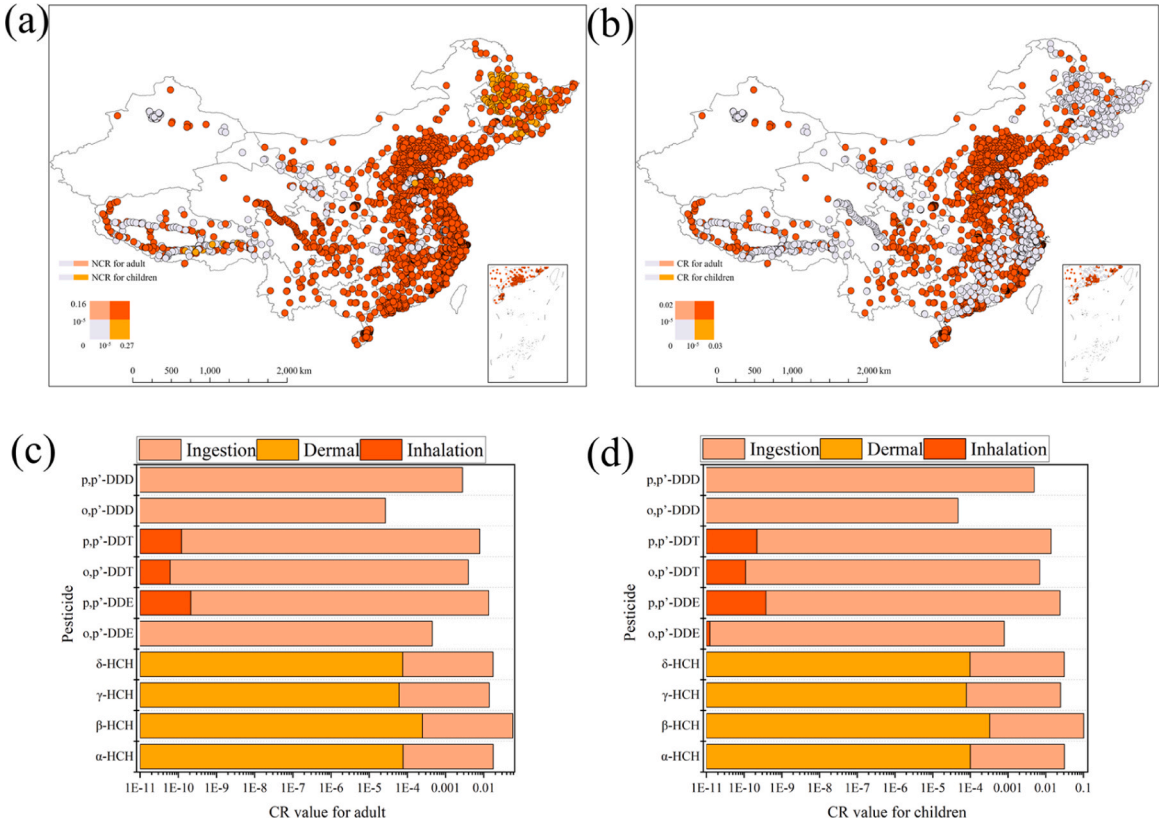


Fig. 7. (a) NCR values for adult and children; (b) CR values for adult and children; (c) CR values of different pesticides to adults; (d) CR values of different pesticides to children.

values of pesticides such as acaricides and organochlorines constituted 90 % of the total RQ values for birds. Furthermore, fungicides, organochlorine and pyrethroids collectively accounted for more than 90 % of the RQ values for earthworms.

The mean NCR for adults and children were 7×10^{-3} and 1×10^{-2}, respectively, both of which were less than 1, indicating no risk from non-carcinogenic effects (Fig. 7a). This study examined the contributions of various exposure routes and PRS to the NCR. Analysis revealed that the average NCR for adults due to ingestion, inhalation and dermal contact were 6.4×10^{-5}, 2×10^{-9} and 2×10^{-7}, respectively (Fig. S2). Ingestion was identified as the primary route of exposure for adults, accounting for 90 % of the NCR. Similarly, ingestion remained the primary non-carcinogenic risk exposure route for children (Fig. S3).

Fig. 7b showed that the average CR values for adults (4.6×10^{-4}) and children (6×10^{-4}) exceeded the tolerable thresholds (10^{-5}) by a small margin. CR values for both DDTs and HCHs were estimated through three exposure routes, revealing varying individual contributions to overall exposure risk. The ingestion route emerged as the primary contributor to carcinogenic effects, surpassing the maximum acceptable level of risk. Consequently, adults faced a significant risk of carcinogenic effects from exposure to DDTs and HCHs, with average CR values for α-HCH, β-HCH, γ-HCH and δ-HCH calculated at 8.9×10^{-3}, 2.9×10^{-2}, 7.0×10^{-3} and 8.8×10^{-3}, respectively (Fig. 7c). As β-HCH is the major congener of HCHs in soil and is the most stable isomer and is hardly degraded by microorganisms, it contributes the most to the carcinogenic risk [3]. Furthermore, the average contributions of o,p′-DDE, p,p′-DDE, o,p′-DDT, p,p′-DDT, o,p′-DDD and p,p′-DDD to the CR were determined to be 2.2×10^{-4}, 6.8×10^{-3}, 2.0×10^{-3}, 3.9×10^{-3}, 1.3×10^{-5} and 1.4×10^{-3}, respectively. This can be attributed to the degradation routes of DDTs [56,57]. For example, a previous study suggested that p,p′-DDT was transformed to p,p′-DDE in soil [58]. Therefore, as persistent organic pollutants originating from degradation, p,p′-DDT exhibited high concentrations and CR values. As a degradation product, the levels and CR values of p,p′-DDE were still high, indicating that DDTs soil is being degraded in China. For children, the characteristics of CR and associated exposure pathways were found to be comparable to those observed in adults. Nevertheless, the carcinogenic impacts increased, resulting in a maximum CR value of 0.1 (Fig. 7d).

3.5. Uncertainty and limitation

Methods and errors in the detection of PRS vary among different studies, which leads to uncertainty in their data. To mitigate this uncertainty, peer-reviewed articles were utilized in this study. The test data reported by the EIA are legally recognized in China, ensuring data authenticity and reliability. This paper presents a novel PRS dataset compiled from references and reports. The evaluation of the SECC and the analysis of the relationships between the PRS and environmental factors were dependent on additional datasets, potentially introducing uncertainties into the findings of this study. The environmental factor datasets in this study were published and peer-reviewed; thus, the associated uncertainty was deemed acceptable. The methods used for assessing ecological and health risks were sourced from the EPA, and the parameters utilized were derived from EPA guidelines and similar studies. Additionally, the risks identified in this paper closely align with previous research conducted both domestically and internationally, suggesting the reliability of the findings presented.

This study failed to evaluate the relationship among PRS, soil microorganism and leaching potential due to the absence of the corresponding authoritative soil environmental factor datasets. Therefore, future research should prioritize biodegradation analyses of PRS and further examine the relationships between PRS and soil environmental factors.

4. Conclusion

This paper established a PRS dataset from 2006 to 2020 with fusion data from WOS and EIA. The average concentration of PRS in China was 243.96 μg/kg, with higher concentrations predominantly observed in the eastern regions of the country. Furthermore, the PRSs exhibited a decreasing trend following the implementation of the "Program of Zero Growth in Pesticides Use". Increasing soil moisture, temperature and acidity can effectively reduce pesticide residues in the soil. Pesticides in the soil of the middle and upper watersheds can be transported downstream through surface runoff, leading to their accumulation in the soil of the lower watersheds Pesticides in the soil can also be transported by the local air current, resulting in their deposition in the piedmont soil. The average SECC in China was estimated at −69.5 kg. The ecological risks associated with pesticide residues in soil were predominantly categorized as negligible, resulting in an acceptable level of risk. PRS posed a carcinogenic risk in both children and adults, and the risk was greater in children than in adults.

CRediT authorship contribution statement

Zhongjun Xu: Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Xin Bo:** Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Shaobo Chen:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.135696.

References

[1] Latif, Y., Sherazi, S.T., Bhanger, M.I., 2011. Assessment of pesticide residues in commonly used vegetables in Hyderabad, Pakistan. Ecotoxicol Environ Saf 74 (8), 2299–2303. https://doi.org/10.1016/j.ecoenv.2011.07.030.

[2] Food and Agriculture Organization of the United Nations, 2024. https://www.fao.org/home/en.

[3] Yu, H., Li, F., Yu, W., Li, Y., Yang, G., Zhou, S., Zhang, T., Gao, Y., Wan, H., 2013. Assessment of organochlorine pesticide contamination in relation to soil properties in the Pearl River Delta, China. Sci Total Environ 447, 160–168. https://doi.org/10.1016/j.scitotenv.2012.12.070.

[4] Yao, R., Yao, S., Ai, T., Huang, J., Liu, Y., Sun, J., 2023. Organophosphate Pesticides and Pyrethroids in Farmland of the Pearl River Delta, China: Regional Residue, Distributions and Risks. Int J Environ Res Public Health 20 (2), 1017. https://doi.org/10.3390/ijerph20021017.

[5] Yu, Z., Li, X., Wang, S., Liu, L., Zeng, E., 2021. The human and ecological risks of neonicotinoid insecticides in soils of an agricultural zone within the Pearl River Delta, South China. Environ Pollut 284, 117358. https://doi.org/10.1016/j.envpol.2021.117358.

[6] Jiang, Y., Wang, X., Jia, Y., Wang, F., Wu, M., Sheng, G., Fu, J.-M., 2009. Occurrence, distribution and possible sources of organochlorine pesticides in agricultural soil of Shanghai, China. J Hazard Mater 170 (2), 989–997. https://doi.org/10.1016/j.jhazmat.2009.05.082.

[7] Wang, X., Piao, X., Chen, J., Hu, J., Xu, F., Tao, S., 2006. Organochlorine pesticides in soil profiles from Tianjin, China. Chemosphere 64 (9), 1514–1520. https://doi.org/10.1016/j.chemosphere.2005.12.052.

[8] Zhou, Q., Wang, J., Meng, B., Cheng, J., Lin, G., Chen, J., Zheng, D., Yu, Y., 2013. Distribution and sources of organochlorine pesticides in agricultural soils from central China. Ecotoxicol Environ Saf 93, 163–170. https://doi.org/10.1016/j.ecoenv.2013.03.029.

[9] Wang, L., Zhang, Z., Liu, L., Zhu, F., Ma, W., 2023. National-scale monitoring of historic used organochlorine pesticides (OCPs) and current used pesticides (CUPs) in Chinese surface soil: Old topic and new story. J Hazard Mater 443, 130285. https://doi.org/10.1016/j.jhazmat.2022.130285.

[10] Zhou, Y., Jing, J., Yu, R., Zhao, Y., Gou, Y., Tang, H., Zhang, H., Huang, Y., 2023. Distribution of pesticide residues in agricultural topsoil of the Huangshui catchment, Qinghai Tibet Plateau. Environ Sci Pollut Res 30 (3), 7582–7592. https://doi.org/10.1007/s11356-022-22704-7.

[11] Liu, F., Wu, H., Zhao, Y., Li, D., Yang, J., Song, X., Shi, Z., Zhu, A.X., Zhang, G., 2022. Mapping high resolution National Soil Information Grids of China. Sci Bull 67 (3), 328–340. https://doi.org/10.1016/j.scib.2021.10.013.

[12] Liu, F., Zhang, G., Song, X., Li, D., Zhao, Y., Yang, J., Wu, H., Yang, F., 2020. High-resolution and three-dimensional mapping of soil texture of China. Geoderma 361, 114061. https://doi.org/10.1016/j.geoderma.2019.114061.

[13] Sun, H., Quan, X., Zhao, Z., 2023. Monthly gap-filled CCI soil moisture over region of China (Combined Product). Sci Data Bank. https://doi.org/10.57760/sciencedb.07849.

[14] Zhang, X., Shen, B., Shen, Y., 2023. China surface net radiation dataset (1982–2017). Science Data Bank, http://doi.org/10.57760/sciencedb.03651.

[15] Shangguan, W., Dai, Y., Liu, B., Zhu, A., Duan, Q., Wu, L., Ji, D., Ye, A., Yuan, H., Zhang, Q., Chen, D., Chen, M., Chu, J., Dou, Y., Guo, J., Li, H., Li, J., Liang, L., Liang, X., Liu, H., Liu, S., Miao, C., Zhang, Y., 2013. A China data set of soil properties for land surface modeling. J Adv Model Earth Syst 5 (2), 212–224. https://doi.org/10.1002/jame.20026.

[16] Yang, Y., Xu, X., Wei, J., You, Q., Wang, J., Bo, X., 2023. A method of gas-related pollution source layout based on multi-source data: A case study of shaanxi province, China. J Environ Manag 347, 119198. https://doi.org/10.1016/j.jenvman.2023.119198.

[17] Agency, N.G.-I., 2014. Landsat—Earth observation satellites.

[18] United States Environmental Protection Agency, 2021. Ecol Risk Models Tools (https://www.epa.gov/risk/ecological-risk-models-and-tools).

[19] SánchezBayo, F., Baskaran, S., Kennedy, I.R., 2002. Ecological relative risk (EcoRR): another approach for risk assessment of pesticides in agriculture. Agric Ecosyst Environ 91 (1), 37–57. https://doi.org/10.1016/S0167-8809(01)00258-4.

[20] Li, R., Hu, W., Liu, H., Huang, B., Jia, Z., Liu, F., Zhao, Y., Khan, K.S., 2024. Occurrence, distribution and ecological risk assessment of herbicide residues in cropland soils from the Mollisols region of Northeast China. J Hazard Mater 465, 133054. https://doi.org/10.1016/j.jhazmat.2023.133054.

[21] Pesticide Properties Database, 2024. https://sitem.herts.ac.uk/aeru/ppdb/en/.

[22] Yang, Y., Chen, T., Liu, X., Wang, S., Wang, K., Xiao, R., Chen, X., Zhang, T., 2022. Ecological risk assessment and environment carrying capacity of soil pesticide residues in vegetable ecosystem in the Three Gorges Reservoir Area. J Hazard Mater 435, 128987. https://doi.org/10.1016/j.jhazmat.2022.128987.

[23] Xie, T., Wang, M., Su, C., Chen, W., 2018. Evaluation of the natural attenuation capacity of urban residential soils with ecosystem-service performance index (EPX) and entropy-weight methods. Environ Pollut 238, 222–229. https://doi.org/10.1016/j.envpol.2018.03.013.

[24] Chen, S., He, Y., Jiang, M., You, Q., Ma, X., Xu, Z., Bo, X., 2024. Unveiling the importance of VOCs from pesticides applicated in main crops for elevating ozone concentrations in China. J Hazard Mater 465, 133385. https://doi.org/10.1016/j.jhazmat.2023.133385.

[25] Zeng, S., Ma, J., Yang, Y., Zhang, S., Liu, G.-J., Chen, F., 2019. Spatial assessment of farmland soil pollution and its potential human health risks in China. Sci Total Environ 687, 642–653. https://doi.org/10.1016/j.scitotenv.2019.05.291.

[26] Wu, W., Wu, P., Yang, F., Sun, D., Zhang, D., Zhou, Y., 2018. Assessment of heavy metal pollution and human health risks in urban soils around an electronics manufacturing facility. Sci Total Environ 630, 53–61. https://doi.org/10.1016/j.scitotenv.2018.02.183.

[27] Beijing Municipal Bureau of Ecology and Environment, 2009. Environmental site assessment guideline (DB11/T 656–2009). Beijing municipal administration of quality and technology supervision Beijing.

[28] United States Environmental Protection Agency, 2011. Exposure Factors Handbook. Office of Research and Development. National Center for Environmental Assessment, U. S. Environmental Protection Agency, Washington, DC.

[29] Baghaie, A.H., Aghili, F., 2019. Health risk assessment of Pb and Cd in soil, wheat, and barley in Shazand County, central of Iran. J Environ Health Sci Eng 17 (1), 467–477. https://doi.org/10.1007/s40201-019-00365-y.

[30] United States Environmental Protection Agency, 2024. CompTox chemicals dashboard (https://comptox.epa.gov/dashboard/chemical).

[31] Tang, F.H.M., Maggi, F., 2021. Pesticide mixtures in soil: a global outlook. Environ Res Lett 16 (4), 044051. https://doi.org/10.1088/1748-9326/abe5d6.

[32] Silva, V., Mol, H.G.J., Zomer, P., Tienstra, M., Ritsema, C.J., Geissen, V., 2019. Pesticide residues in European agricultural soils – A hidden reality unfolded. Sci Total Environ 653, 1532–1545. https://doi.org/10.1016/j.scitotenv.2018.10.441.

[33] Froger, C., Jolivet, C., Budzinski, H., Pierdet, M., Caria, G., Saby, N.P.A., Arrouays, D., Bispo, A., 2023. Pesticide Residues in French Soils: Occurrence, Risks, and Persistence. Environ Sci Technol 57 (20), 7818–7827. https://doi.org/10.1021/acs.est.2c09591.

[34] Ukalskajaruga, A., Smreczak, B., Siebielec, G., 2020. Assessment of Pesticide Residue Content in Polish Agricultural Soils. Molecules 25 (3), 587. https://doi.org/10.3390/molecules25030587.

[35] Harner, T., Wideman, J.L., Jantunen, L.M.M., Bidleman, T.F., Parkhurst, W.J., 1999. Residues of organochlorine pesticides in Alabama soils. Environ Pollut 106 (3), 323–332. https://doi.org/10.1016/S0269-7491(99)00110-4.

[36] Beard, J., 2006. DDT and human health. Sci Total Environ 355 (1), 78–89. https://doi.org/10.1016/j.scitotenv.2005.02.022.

[37] Yadav, I.C., Devi, N.L., Syed, J.H., Cheng, Z., Li, J., Zhang, G., Jones, K.C., 2015. Current status of persistent organic pesticides residues in air, water, and soil, and their possible effect on neighboring countries: A comprehensive review of India. Sci Total Environ 511, 123–137. https://doi.org/10.1016/j.scitotenv.2014.12.041.

[38] Babu Rajendran, R., Venugopalan, V.K., Ramesh, R., 1999. Pesticide residues in air from coastal environment, South India. Chemosphere 39 (10), 1699–1706. https://doi.org/10.1016/S0045-6535(99)00070-3.

[39] National Bureau of Stastics of China, 2021. China Statistical Yearbook 2020. China Statistics Press, Beijing, China.

[40] Planting Industry Management Department Ministry of Agriculture, 2015. Zero growth action plan for pesticide use by 2020. (http://www.moa.gov.cn/govpublic/ZZYGLS/201503/t20150318_4444765.htm). (In Chinese).

[41] Standing Committee of the National People's Congress, 2019. Law of the People's Republic of China on Environmental Impact Assessment (https://www.mee.gov.cn/ywgz/fgbz/fl/201901/t20190111_689247.shtml) (in Chinese).

[42] Ministry of Ecological Environment of the People's Republic of China, 2022. Guidelines of the Ministry of Ecology and Environment on the Implementation of the 'Three Lines One Permit' Ecological Environment Zoning Control (Trial) (https://www.gov.cn/gongbao/content/2022/content_5683858.htm) (in Chinese).

[43] Kata, M., Srinivasa Rao, S., Rama Mohan, K., 2015. Spatial distribution, ecological risk evaluation and potential sources of organochlorine pesticides from soils in India. Environ Earth Sci 74 (5), 4031–4038. https://doi.org/10.1007/s12665-014-3189-6.

[44] Gan, Q., Jans, U., 2007. Nucleophilic Reactions of Phorate and Terbufos with Reduced Sulfur Species under Anoxic Conditions. J Agric Food Chem 55 (9), 3546–3554. https://doi.org/10.1021/jf063296m.

[45] Sugeng, A.J., Beamer, P.I., Lutz, E.A., Rosales, C.B., 2013. Hazard-ranking of agricultural pesticides for chronic health effects in Yuma County, Arizona. Sci Total Environ 463-464, 35–41. https://doi.org/10.1016/j.scitotenv.2013.05.051.

[46] Liu, P., Xu, Y., Li, J., Liu, J., Cao, Y., Liu, X., 2012. Photodegradation of the isoxazolidine fungicide SYP-Z048 in aqueous solution: kinetics and photoproducts. J Agric Food Chem 60 (47), 11657–11663. https://doi.org/10.1021/jf3034607.

[47] Wang, R., Chen, H., Luo, Y., Yen, H., Arnold, J.G., Bubenheim, D., Moran, P., Zhang, M., 2019. Modeling Pesticide Fate and Transport at Watershed Scale Using the Soil & Water Assessment Tool: General Applications and Mitigation Strategies. Pesticides in Surface Water: Monitoring, Modeling, Risk Assessment, and Management. American Chemical Society,, pp. 391–419. https://doi.org/10.1021/bk-2019-1308.ch020.

[48] Zhang, B., Zhang, Q., Zhang, S., Xing, C., Ying, G., 2020. Emission estimation and fate modelling of three typical pesticides in Dongjiang River basin, China. Environ Pollut 258, 113660. https://doi.org/10.1016/j.envpol.2019.113660.

[49] Ji, T., Lin, T., Wang, F., Li, Y., Guo, Z., 2015. Seasonal variation of organochlorine pesticides in the gaseous phase and aerosols over the East China Sea. Atmos Environ 109, 31–41. https://doi.org/10.1016/j.atmosenv.2015.03.004.

[50] Jakobi, G., Kirchner, M., Henkelmann, B., Körner, W., Offenthaler, I., Moche, W., Weiss, P., Schaub, M., Schramm, K., 2015. Atmospheric bulk deposition measurements of organochlorine pesticides at three alpine summits. Atmos Environ 101, 158–165. https://doi.org/10.1016/j.atmosenv.2014.10.060.

[51] Ding, Y., Huang, H., Chen, W., Zhang, Y., Chen, W., Xing, X., Qi, S., 2022. Background levels of OCPs, PCBs, and PAHs in soils from the eastern Pamirs, China, an alpine region influenced by westerly atmospheric transport. J Environ Sci 115, 453–464. https://doi.org/10.1016/j.jes.2020.11.022.

[52] Li, X., Cundy, A., Chen, W., Liu, R., Lv, S., 2020. Dynamic capacity modelling of soil environment carrying capacity, and developing a soil quality early warning framework for development land in China. J Clean Prod 257, 120450. https://doi.org/10.1016/j.jclepro.2020.120450.

[53] Vašíčková, J., Hvězdová, M., Kosubová, P., Hofman, J., 2019. Ecological risk assessment of pesticide residues in arable soils of the Czech Republic. Chemosphere 216, 479–487. https://doi.org/10.1016/j.chemosphere.2018.10.158.

[54] Pérez, D.J., Iturburu, F.G., Calderon, G., Oyesqui, L.A.E., De Gerónimo, E., Aparicio, V.C., 2021. Ecological risk assessment of current-use pesticides and biocides in soils, sediments and surface water of a mixed land-use basin of the Pampas region, Argentina. Chemosphere 263, 128061. https://doi.org/10.1016/j.chemosphere.2020.128061.

[55] State Council, 2024. Opinions on strengthening ecological zoning control (https://www.mee.gov.cn/zcwj/zyygwj/202403/t20240318_1068588.shtml) (in Chinese).

[56] Sudharshan, S., Naidu, R., Mallavarapu, M., Bolan, N., 2012. DDT remediation in contaminated soils: a review of recent studies. Biodegradation 23 (6), 851–863. https://doi.org/10.1007/s10532-012-9575-4.

[57] Wedemeyer, G., 1967. Dechlorination of 1,1,1-Trichloro-2,2-bis(p-chlorophenyl) ethane by Aerobacter aerogenes. Appl Microbiol 15 (3), 569–574. https://doi.org/10.1128/am.15.3.569-574.1967.

[58] Huang, H., Zhang, Y., Chen, W., Chen, W., Yuen, D.A., Ding, Y., Chen, Y., Mao, Y., Qi, S., 2018. Sources and transformation pathways for dichlorodiphenyltrichloroethane (DDT) and metabolites in soils from Northwest Fujian, China. Environ Pollut 235, 560–570. https://doi.org/10.1016/j.envpol.2017.12.071.