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Status, Distribution and Ecological Risk Assessment of Persistent Toxic Substances in Suburban Agricultural Soils in Hohhot City, North China

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Abstract

A total of 52 soil samples of Hohhot City were collected and four heavy metals and eight organochlorine pesticides were analyzed. The results showed that Cr, Cu, Zn, and Pb in soil were in range of 20.54 - 48.15 mg/kg, 40.10 - 94.60 mg/kg, 35.14 - 110.48 mg/kg, 38.86g - 245.36 mg/kg, with a mean value of 37.24 mg/kg, 60.76 mg/kg, 80.49 mg/kg, and 145.99 mg/kg, respectively. The high degree of variation of Pb, Zn, and Cu, reflected that the heavy metals were mainly interfered by human factors. The content values of ∑DDTs ranged from 5.01 ng/g to 105.08 ng/g with a mean of 36.94 ng/g, while the ∑HCHs ranged from 6.52 ng/g to 48.65 ng/g with an average of 23.29 ng/g, indicated that DDTs were highly used than HCHs in the study area. The relatively low α-HCH/γ-HCH ratio and relatively high o,p'-DDT/p,p'-DDT ratio indicated the application of lindane and dicofol on regional agricultural soil. The mean Igeo values was Pb (2.9) > Cu (1.5) > Cr (−0.31) > Zn (−0.35), revealing that the soil in Hohhot City was not polluted by Cr and Zn (Igeo < 0), moderate polluted by Cu (1 < Igeo < 2), moderate to heavy pollution by Pb (2 < Igeo < 3). The range of potential ecological risk index of metals was from 35.02 to 132.96, indicating low to moderate potential ecological risk. HCHs in all soil samples were less than 50 ng/g, which could be considered as unpolluted, while DDTs in several samples were between 50 and 500 ng/g, which could be regarded as having low pollution.

Keywords

Heavy Metals, Organochlorine Pesticides, Source Identification, Pollution

Assessment, Index of Geoaccumulation

1. Introduction

The land resources, especially agricultural soil, are the material foundation for the survival and development of human society and the important carrier for a sustainable development of human beings (Lu, 2017). As the land resources provide essential nutrients for the growth and reproduction of land life, they are also suffering from all kinds of pollutions caused by human activities. The suburb agricultural soil is an important part of urban ecological environment, and it has great significance for the sustainable development of the city. As a frequent area of interaction between rural and urban economic activities, suburban is not only the main "source" of urban life and production raw materials, but also the "sink" of the discharge of three wastes such as urban domestic sewage and industrial wastewater (Lu et al., 2012; Huang et al., 2007). With the development of urban-rural integration and agricultural modernization, the soil resources in suburb are shrinking rapidly. While the intensive agriculture enhanced the output of agricultural products, it has also caused serious environment pollution in China. The use of excessive agrochemical products (including organochlorine pesticides and fertilizers containing heavy metals) has led to the accumulation of toxic and harmful substances in suburban soils. In addition, a large number of pollutants produced from industrial production, transportation, livestock farming and living also enter the soil through different ways and forms, resulting in suburban soil degradation, and these pollutants can accumulate through the food chain, causing a serious threat to human health.

Persistent toxic substances (PTSs) are a large group of environmental pollutants that can be bioaccumulated by living organisms, that are resistant to degradation and that possess acute or chronic toxic properties (Barra et al., 2005; Jin et al., 2016; Wang et al., 2017). Many of the PTSs can transport over long distances through the atmosphere or oceans, resulting in global distribution and even detectability in remote areas where they have never been used before. PTSs and their resulting effects give rise to specific concerns at local, national, regional and global scales due to their properties (Jiao et al., 2009; Curtosi et al., 2010; Li & Yuan, 2017). With the rapid development of urbanization and intensive agriculture in China, more and more agricultural soils have been contaminated with PTSs, of which, heavy metals and organochloride pesticides (OCPs) are two important classes of compounds that have caused serious environmental problems. Particularly most of OCPs are classified as persistent organic pollutants by Stockholm Convention UNEP in 2004. Compared with other developed cities in China, the industries in Hohhot City are of the characteristics of large pollutant discharge and high energy consumption. It has been reported the presence of several activities around the soil in Hohhot including industrial activities, urban domestic sewage and industrial waste water, urban traffic pollution, power station, etc. These activities have their negative impacts on the soil since many hazardous materials are transported into soil. Off these pollutants, heavy metals loads are moved into the soil either in the dissolved or particulate form, then adsorbed on the surface soil as a final reservoir for metals. Therefore the pollution of pesticides and heavy metals in the soil of Hohhot City has attracted the attention of many scientists, and the soil contamination of Hohhot City was monitored in some previous studies (Guo et al., 2013; Shi & Huang, 2010; Zhang et al., 2013). The heavy metal contamination characteristics of soil in different functional areas of Hohhot City were analyzed and evaluated by Guo (Guo et al., 2013), it was found that the heavy metals in the investigated soil exceeded the soil background values of Inner Mongolia autonomous regions, in which Cu and Zn were 2.33 and 1.85 times of the background values respectively. A total of 928 samples of four soil subtypes in Hohhot were analyzed to determine the soil background values of Cu and Pb, it was found that the highest content of Cu in tidal soil was 18.83 mg/kg, and the highest content of Pb in proluvial soil was 17.50 mg/kg (Shi & Huang, 2010). A detailed analysis and evaluation on PAHs in agricultural soils of Hohhot was carried out by Zhang et al., and it was found that the total of 15 PAHs in agricultural soil were in range of 114 - 948 μg/kg, and more than 70% of soil in Hohhot City was lightly polluted by PAHs with high molecular weights (Zhang & Zhang, 2017).

The current work has fostered a focused research on evaluating the ecological hazards of the soil of Hohhot City, and the concentration and distribution of PTS in the suburban agricultural soil of Hohhot City were systematically investigated, and several approaches are currently used to assess contamination levels in soils and thus predict the quality of the soil environment. Such investigation will provide the pollution information in the area, and help understand the suburban pollution situations.

2. Materials and Methods

2.1. Study Area and Sample Collection

The Hohhot City, located in the central part of North China's Inner Mongolia autonomous region, is an important hub city connecting Northeast China, North China and Northwest China, as well as an important node city for the construction of Belt and Road Initiative. Its coordinates are 110˚46' to 112˚10' East longitude and 40˚51' to 41˚8' North latitude. The Hohhot City covers an area of 17,200 square kilometers with a 2065-sq-km urban area, and it is home to nearly 3 million people. The territory is mainly divided into two major landform units. Daqing Mountain in the north and Manhan Mountain in the southeast are mountainous terrain, while the south and southwest are the Tumochuan Plains. Its highest point is 2280 meters above sea level, at Jinluandian, Daqing Mountain. The peak in the downtown area is 1040 meters high. Hohhot belongs to the mid-temperate continental monsoon climate and the climate change in the four seasons is obvious, with great differences. The winter is long and severely cold, the summer is short and hot and the climate changes sharply in the spring and autumn. Its coldest temperature can be −12.7˚C, and the hottest, 22.9˚C. Annual rainfall is between 335.2 and 534.6 mm with an average value of 430 mm. Most rainfall occurs between June and September, and this accounts for 80% of the total annual rainfall. Dahei River and Xiaohei River are the main rivers in Hohhot, with a drainage area of 1380.9 square kilometers. Crop in this area is primarily maize, which require considerable amounts of water for irrigation.

The samples were collected from thirteen agricultural fields of Hohhot City in September 2013. At each sampling point, four samples were collected at depths of 0 - 20 cm and the total number of samples was 52. All soil samples were collected using a hand auger, and then stored separately in polyethylene bags at −20˚C to avert any adverse changes. The soil samples were then dried using a freeze-drier for about 48 h, ground to fine powder, and sifted through a 60-mesh nylon sieve to remove the coarse debris. The location of all composite samples was recorded by a handheld global positioning system (**Figure 1**). The main

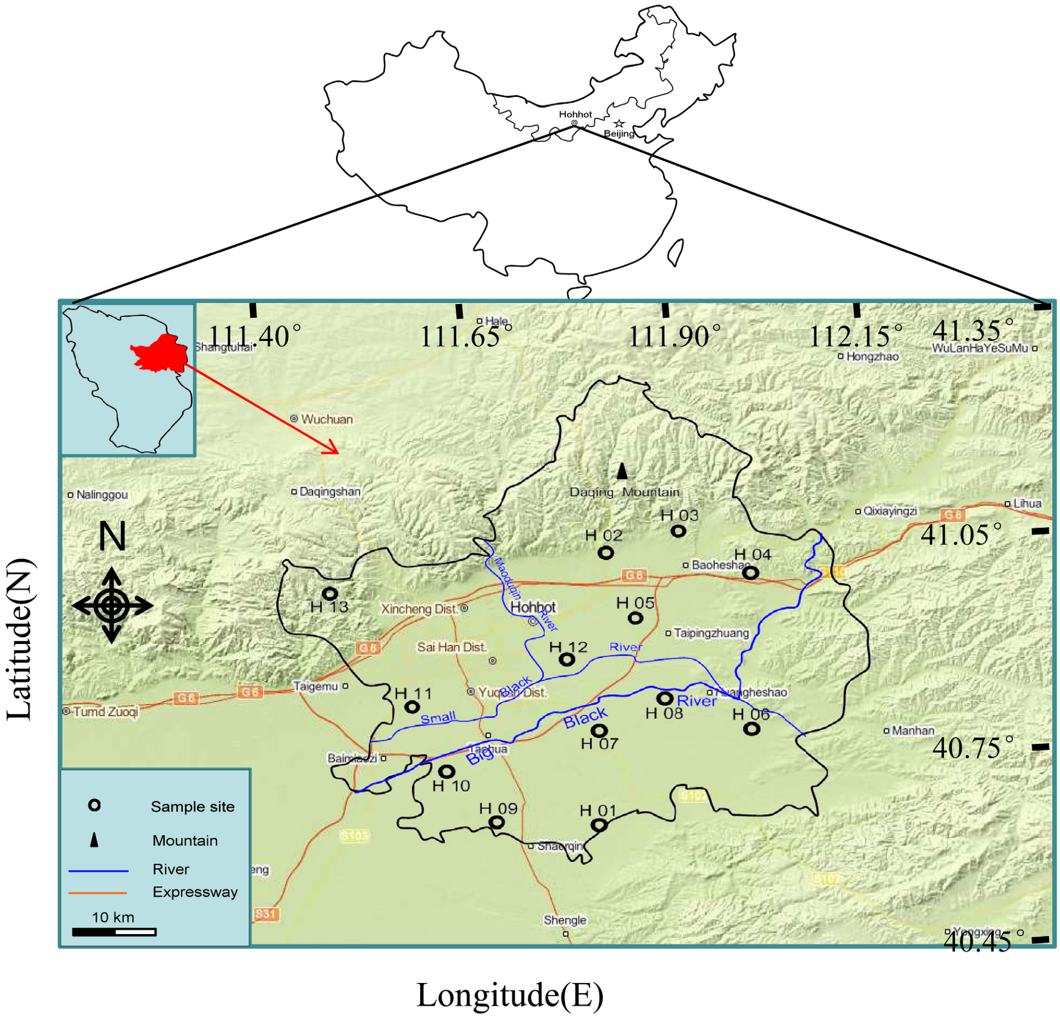


Figure 1. Distribution of sampling sites in soils of Hohhot.

types of soil were Chao soil, Chestnut-brown soil, Chestnut soil, and Aeolian Soil, and the concentrations of organic matter were about in range of 0.25% - 1.5%, and the pH value was from 7.4 to 8.9 (**Table 1**). To investigate the pollution impact of various human activities, land use was classified into three types: farmland soils, vegetable soils and fruits tree soils.

2.2. Materials and Reagents

The OCPs standard solution including HCHs (α-HCH, β-HCH, δ-HCH and γ-HCH) and DDTs (p,p'-DDT, p,p'-DDE, p,p'-DDD, and o,p'-DDT) were purchased from National Research Center for Certified Reference Materials of China. The original storage solution was 100 mg/L in n-hexane and further diluted to obtain the desired concentrations. Sodium sulfate (analytical-grade, Beijing Chemical Factory, China) was activated in a muffle furnace at 600˚C for 8 h. Florisil (100 - 200 mesh, Supelco Inc., USA) was activated in the oven at 170˚C for 13 h. The activated Florisil and sodium sulfate were stored in a sealed desiccator. The other reagents including n-hexane, dichloromethane and anhydrous sodium sulfate, were of analytical-grade (Beijing Chemical Reagent Factory, China). Dichloromethane and n-hexane were redistilled before use and a solvent check showed that there was no interference with the target compounds for acceptance. The laboratory glassware was washed with detergents and dichromate acid cleaning solution, rinsed with tap water and distilled water, and then rinsed with methanol (analytical-grade; Tianjin Chemical Reagent Company, China), acetone (analytical-grade; Tianjin Chemical Reagent Company, China) and dichloromethane (analytical-grade; Tianjin Chemical Reagent Company, China), respectively.

Table 1. The basic conditions of the sample sites.

Sample site	Land use type	Soil type	TOC (%)	pH
H1	farmland soils	chestnut soil	0.634	7.8
H2	farmland soils	chestnut soil	0.956	8.1
H3	farmland soils	chestnut soil	0.853	7.9
H4	farmland soils	aeolian soil	0.189	8.9
H5	farmland soils	chao soil	1.168	7.6
H6	farmland soils	aeolian soil	0.253	8.4
H7	farmland soils	chao soil	0.978	7.7
H8	farmland soils	chao soil	1.022	7.4
H9	farmland soils	chestnut-brown soil	0.836	8.2
H10	farmland soils	chao soil	1.011	7.6
H11	farmland soils	chestnut-brown soil	0.968	8.0
H12	vegetable soils	chao soil	1.502	7.5
H13	fruits tree soils	chestnut-brown soil	0.815	8.4

For metal analysis, nitric acid and hydrochloric acid were guaranteed reagents (GR) and purchased from Beijing Chemical Reagents Company. All glass vessels were soaked in 1:1 nitric acid for at least 12 h then rinsed with de-ionized water for several times. The PTFE containers were boiled with 50% v/v nitric acid and the PET bottles were immersed in 5% v/v nitric acid for 24 h followed by rinsing with deionized water.

2.3. Sample Pretreatment and Instrumental Analysis

The analytical procedure of OCPs was done according to a similar procedure (Zhang et al., 2013; Sun et al., 2016) where, the soil sample (10 g) was thoroughly mixed with anhydrous sodium sulfate and ultrasonic treatments with 100 mL of acetone-dichloromethane (1:1, v/v) about 0.5 h using ultrasonic bath. The extracts were concentrated to 5 mL using a rotary evaporator, and the blended matters were then conveyed into a solid-phase extraction cartridge column, which was packed with anhydrous sodium sulfate (0.5 g), Florisil (1.0 g), acidic silica gel (1.0 g), and copper powder (0.5 g) from the bottom to the top. The extracts were then eluted into glass bottles using 20 mL 1:9 (v/v) acetone and *n*-hexane. The eluents collected were further dried under a gentle stream of nitrogen gas and the residues re-solubilized with 1.0 mL of *n*-hexane. Lastly, the constituents were analyzed using gas chromatograph (GC) coupled with mass spectrometer (MS). In electron impact ionization mode, the MS source temperature was 230˚C, quadrupole rods temperature was 150˚C, and the electron energy was 70 eV, 1.0 μL sample extract was injected with a split-splitless injector held constant at 270˚C. The carrier gas was high purity nitrogen (1.0 mL/min). Gas chromatographic separation was performed on a DB-5MS capillary column (30 m × 0.25 mm × 0.25 μm film thickness). The GC column was maintained at 80˚C for 2 min and then ramped at 15˚C min^{-1} up to 270˚C, and held at this temperature for 15 min.

Heavy metal concentration in soil samples was determined by atomic absorption spectrometer after acid digestion (Demirel et al., 2008). Firstly, approximately 1.0 g of each dried soil sample was weighed and digested in a combination of concentrated nitric acid and hydrochloric acid in the ratio of 3:1. After that, the tubes were put into the digestion block at the low temperature (40˚C) for 1 h and then the temperature was increased to 140˚C for at least 3 h. The digested samples were diluted to 40 mL by double distilled water and filtered through Whatman No. 1 (filter speed: medium) filter paper in a funnel into acid washed pillboxes. The blank sample was run by following a similar procedure. Then, the extract was passed in different wavelengths (Cu: 324.8 nm, Pb: 283.3 nm, Zn: 283.3 nm, Cr: 357.9 nm) in atomic absorption spectrophotometer for detecting different metal.

For the measurement of the total organic carbon (TOC), the soil sample (50 mg) was tightly bound by a tin foil and then placed in the TOC analyzer (SSM-5000A, Shimadzu, Japan). Soil pH was determined in distilled water at soil solution ratio of 1:5 with a potentiometric glass electrode using pH meter (Al-Taisan & Gabr, 2017).

2.4. Quality Assurance and Quality Control (QA/QC)

Before analysis, the average recoveries and relative standard deviation (RSD) of OCPs were first obtained to evaluate the method performance by multiple analyses of 10 replicate spiked soil samples. A solvent blank and matrix blank were processed through the entire procedure and analyzed prior to and after every 10 samples. Each sample was analyzed in triplicate unless otherwise stated. The LODs were determined as signals three times the background signal. The detection limit of the method ranged from 0.001 to 0.030 ng/g, and the limit of quantification was 0.115 - 0.187 ng/g. The average retrievals of OCPs varied from 83% - 102% with relative standard deviation (RSDs) from 5.2% to 10.7%. The results of the correlation coefficients derived for the standard curves showed values greater than 0.997.

For metal analysis, a procedural blank and a matrix sample spiked with standards were used to determine the accuracy for each set of ten field samples. Each sample was analysed in duplicate unless otherwise stated. The recoveries for these metals in the standard reference material were around 90% - 110%. Moreover, reagent blank and replicate samples were also analyzed throughout the analysis and were used to correct the analytical results. The results showed that there was no contamination during analysis, and the relative standard deviation (RSD) of all replicate samples was less than 10%.

2.5. Measurement of Soil Pollution Index (SPI)

Pollution index was used to assess the quality of soil and to estimate the impact of anthropogenic activities (Loska et al., 2004). The geoaccumulation index was introduced by Muller (Muller, 1969) and has been applied to assess soil contamination by heavy metals (Hou et al., 2013a). For Igeo computation, the following equation was used:

$$\text{Igeo} = \log_2 \frac{C_i}{1.5Bn} \tag{1}$$

where, Igeo is the geoaccumulation index, C_i is the measured metal concentrations in the tested soil, 1.5 is a factor that is used to calculate possible changes in background value, Bn is the respective element concentration in the earth's crust. The Bn values in soil of Hohhot City for Cu, Pb, Zn and Cr were 13.4 mg/kg, 11.3 mg/kg, 65 mg/kg and 28.1 mg/kg, respectively. The seven classes of the geoaccumulation index as proposed by Müller were shown in **Table 2**.

As can be seen from the definitions of Igeo, the Igeo can only assess the ecological risks of individual metals, but not to estimate the comprehensive risks of the investigated heavy metals. So the comprehensive risk was assessed using the potential ecological risk index (RI) originally proposed by Håkanson (Håkanson, 1980) in 1980.

Table 2. Geoaccumulation index (Igeo) and Ecological risk index (RI) classification.

Igeo		E_r^i			RI	
Igeo Classes	soil quality	E_r^i	value of E_r^i	risk	value of RI	Risk
Igeo ≤ 0	No Pollution	0	$E_r^i < 40$	Low	RI < 110	Low
Igeo = 0 - 1	No to moderate pollution	1	$E_r^i = 40 - 80$	Moderate	RI = 110 - 200	Moderate
Igeo = 1 - 2	Moderate pollution	2	$E_r^i = 80 - 160$	Considerable	RI = 200 - 400	Considerable
Igeo = 2 - 3	Moderate to heavy pollution	3	$E_r^i = 160 - 320$	High	RI ≥ 400	Very high
Igeo = 3 - 4	Heavy pollution	4	$E_r^i \geq 320$	Very high		
Igeo = 4 - 5	Heavy to extreme pollution					
Igeo ≥ 5	Extreme pollution					

The value of RI can be calculated using the following formulas:

$$E_r^i = T_r^i \times \frac{C_i}{C_0} \tag{2}$$

$$\text{RI} = \sum_{i=1}^{n} T_r^i \times \frac{C_i}{C_0} \tag{3}$$

where, C_i is the present concentration of a metal in sample, and C_0 is the background concentration of metals in soil of Hohhot City, T_r^i is the toxic-response factor for a heavy metal; E_r^i is the potential risk of an individual metal, RI is the sum of the potential risk of each heavy metal. According to the Håkanson's method, the toxic-response factors for Cr, Cu, Pb and Zn are 2, 5, 5 and 1, respectively (Hou et al., 2013a; Håkanson, 1980; Chai et al., 2017). The consult values as background concentrations were selected of the concentrations in the layer of 150 - 200 cm on the soil profiles of the three land-use types, and the background values of the soil in Hohhot (Shi & Huang, 2010; Gu et al., 1995) were selected as referee background values. Different levels of risk index are presented in **Table 2**.

3. Results and Discussion

3.1. The Soil Properties of the Study Area

The descriptive statistics of soil properties in suburban agricultural soils of Hohhot City were presented in **Table 1**. The soils showed light to medium texture, they were weakly to strongly basic, and the soil pH ranged from 7.4 to 8.9, and had increase tendency as compared with natural soils. The concentrations of TOC in soil were greatly varied in range of 2.53 g/kg to 15.02 g/kg with a mean of 8.60 g/kg. The soils of the studied area were mainly consisted of four types: chao soil, chestnut-brown soil, chestnut soil, and aeolian soil, and these characteristics point to soils of varying cation exchange ability and of adequate drainage that lead to sufficient soil aeration and to oxidizing conditions. Considering the typical semi-arid climate rainfall distribution, and the mean annual rainfall ranges from about 400 mm, the soil environment was not favored the proliferation of an active soil biota community.

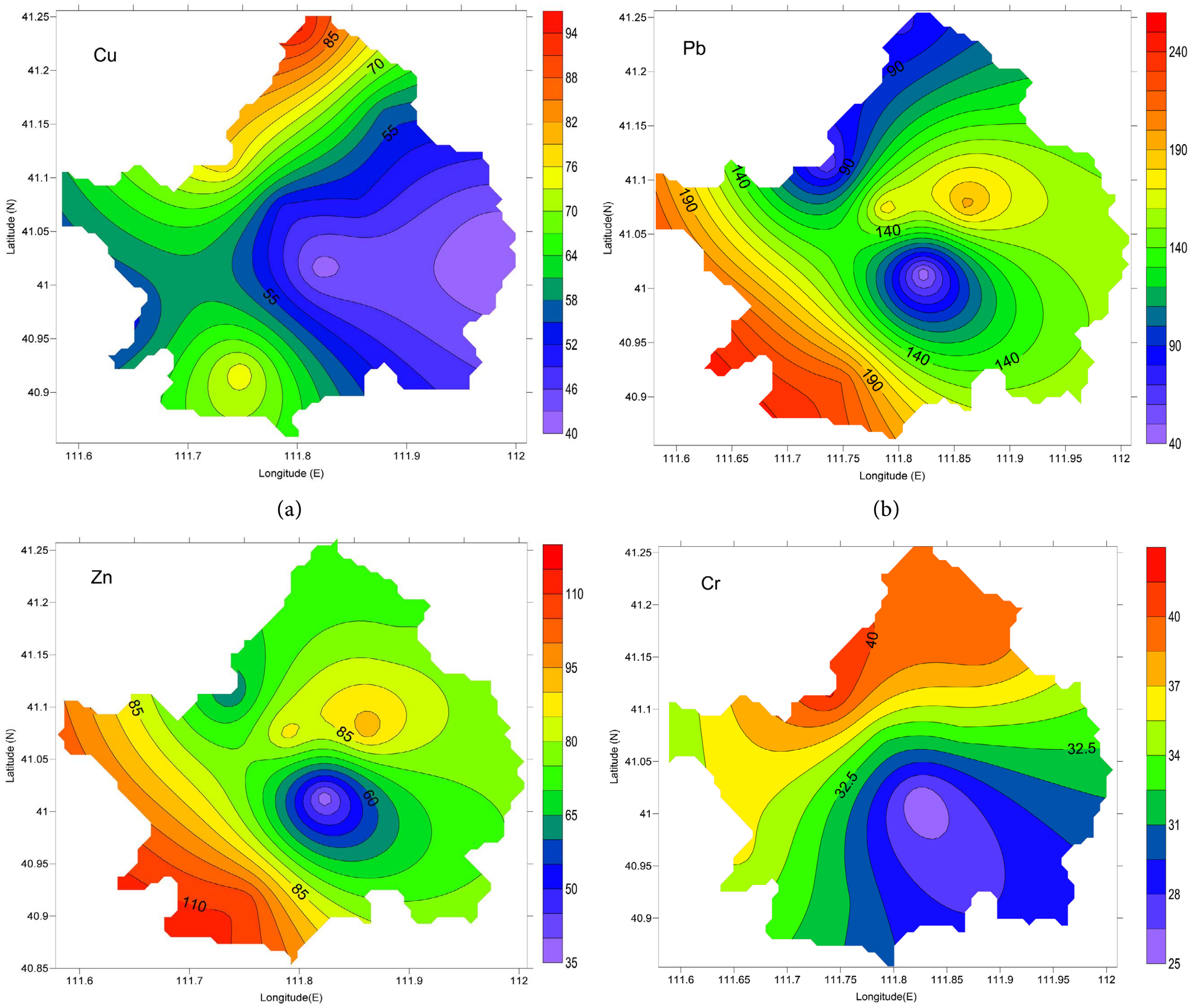
3.2. The PTSs Level in Soils and Their Comparison with Previous Study

Concentrations and patterns of heavy metals in suburban agricultural soils of Hohhot City were shown in **Table 3**. The concentrations of Cr, Cu, Zn, and Pb in soils of Hohhot City varied between 20.54 mg/kg and 48.15 mg/kg, 40.10 mg/kg and 94.60 mg/kg, 35.14 mg/kg and 110.48 mg/kg, 38.86 mg/kg and 245.36 mg/kg, respectively. The mean concentrations of Cr, Cu, Zn, and Pb were 37.24 mg/kg, 60.76 mg/kg, 80.49 mg/kg, and 145.99 mg/kg, respectively. The CV of the studied metals in suburban agricultural soils of Hohhot City decreased in the order Pb (47%) > Zn (28%) > Cu (26%) > Cr (17%). The CV of Pb, Zn, and Cu indicated a high degree of variation, reflecting that the heavy metals were mainly interfered by human factors (Nezhad et al., 2015; Huang et al., 2009). The spatial patterns of metals in suburban agricultural soil in Hohhot City were shown in **Figure 2**. The content of Pb in soil had a high value alone the city river in the western area of Hohhot, and presented a descending trend from western to eastern of Hohhot City. Compared with China's Soil Environmental Quality Standard (GB15618-1995), about 25% of the sampling sites exceeded the grade II limit for Pb in soil. The highest level of Pb in all sampling points appeared in H10 with a value of 245.36 ng/g, which was 2.5 times the grade II limit for Pb in GB15618-1995, which may be closely related to the rapid industrialisation-driven urbanisation process. In addition, power plants and other high energy consuming enterprises in southwest of Hohhot is likely to have contributed to rising Pb levels in this area. For Zn, the spatial distribution of Zn was much like that of Pb, and there were three sites (H9, H10, H11) that exceeded the background value of soil Zn in Hohhot but lower than the grade II limit for Zn in GB15618-1995, and the soil Zn in which was 108.5 mg/kg, 106.7 mg/kg and 110.5 mg/kg, respectively.

Table 3. The statistical features of concentrations of heavy metals on suburban farmland in Hohhot (mg/kg).

Metal	Range	Means	CV	The second level criterion of Standard of Soil Environment (mg/kg)[a]				The third level criterion of Standard of Soil Environment (mg/kg)[b]
				pH < 5.5	5.5 ≤ pH < 6.5	6.5 ≤ pH < 7.5	>7.5	pH > 6.5
Cr	25.04 - 48.15	37.24	17%	120	150	200	250	300
Cu	40.10 - 94.60	60.76	26%	50	50	100	100	400
Zn	35.14 - 110.48	80.49	28%	150	200	250	300	500
Pb	38.86 - 245.36	145.99	47%	80	80	80	80	500

[a]GB15618-2008; [b]GB15618-1995.



(a) (b)

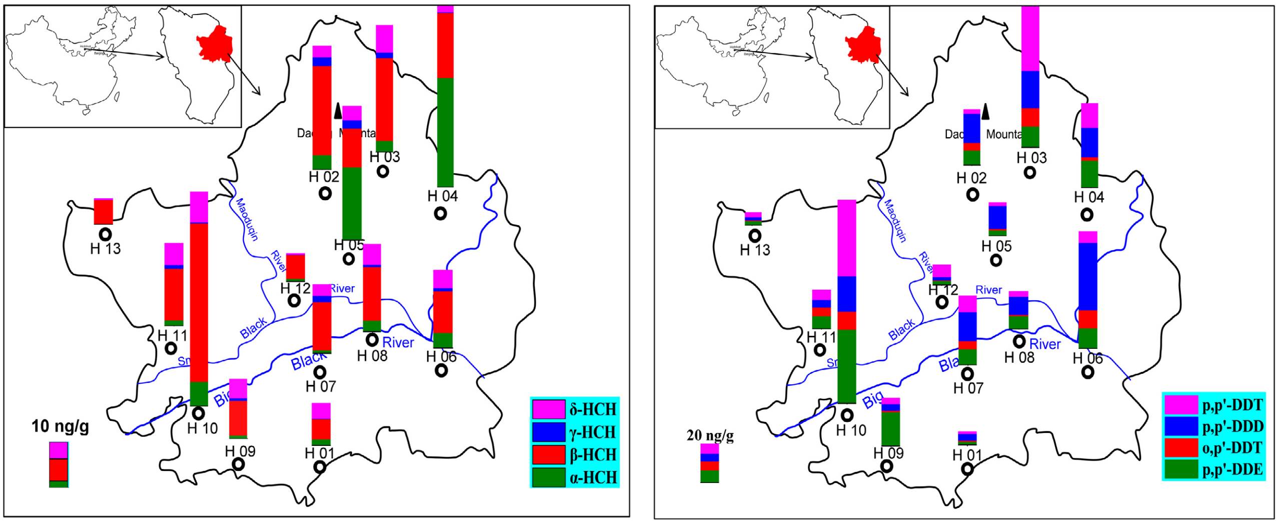
(c) (d)

Figure 2. Spatial patterns of heavy metals in agricultural soil in Hohhot.

The high intense human activities and urbanization in these points were the key factors for making the increase of soil Zn. The content of Cu in soil were in range of 40.10 mg/kg to 94.60 mg/kg, which was 1.2 - 2.7 times the background value of soil Cu in Hohhot, indicating that the farmland soil around Hohhot City had been polluted by copper to different degree. Soil contamination with copper consist mainly of natural source and human activities source, the discharge of wastewater containing copper or the usage of fertilizer containing copper can increase the soil Cu contents. For this area, the soil Cu in farmland was higher than that of background value might be caused by the excessive use of copper containing chemicals and organic fertilizer. The soil Cr in farmland soil of Hohhot ranged from 25.04 mg/kg to 48.15 mg/kg, the highest soil Cr was found in H3 and the lowest was in H1, which was lower than the soil background value of Hohhot City.

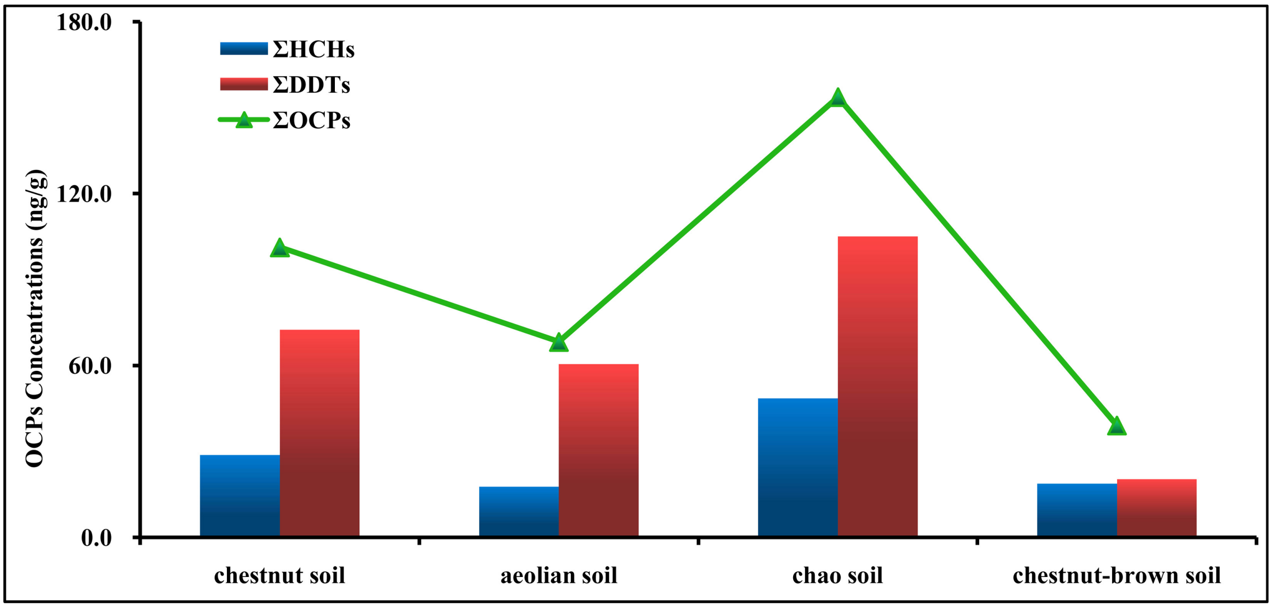
Concentrations and patterns of OCPs in soil samples were summarized in Figure 3. The obtained concentration values of ∑DDTs ranged from 5.01 ng/g to 105.08 ng/g with the mean of 36.94 ng/g, while, the ∑HCHs ranged from 6.52 ng/g to 48.65 ng/g with an average of 23.29 ng/g. These results indicated that DDTs were highly used than HCHs in the study area. The total OCPs (the sum of DDTs and HCHs) in the soil of the study area ranged from 14.71 ng/g to 153.73 ng/g, with a mean value of 60.24 ng/g. According to China's Environmental Quality Standard for Soils (GB 15618-1995), the limits for both HCHs and DDTs in soils were 50, 500 and 1000 ng/g, corresponding to Class I, II and III, respectively. In this study, the mean concentration of ∑DDTs and ∑HCHs were all below the class II of Environmental Quality Standard for Soils of China in 2009. The results suggested that the input of DDTs into farmland soil of Hohhot was higher than that of HCHs, however, the production and consumption of DDTs in China was only 10% of that of HCHs, the most likely explanation is the difference in physicochemical and biochemical properties, wherein HCHs have higher water solubility, vapor pressure, and biodegradability, but lower lipophilicity and particle affinity, thus, DDTs tend to remain in the particulate phase longer than HCHs (Yang et al., 2015).

There are some differences in OCPs concentrations between different soils and use types (shown in Figure 4). Generally, the differences of OCPs residues in soils is mainly related to historical usage and banned time (Singh et al., 2007). Soil types has a great influence on the residue of OCPs in soils, the sandy soil has relatively low adsorption ability compared with clayey soil, and OCPs in this soil easily to be lost, inducing the lower OCPs concentration in sandy soil. In this study, the concentration of OCPs in different soil types in Hohhot is sorted as chao soil > chestnut soil > aeolian soil > chestnut brown soil, and the results are in agreement with the theoretic results. It is generally agreed that the organic

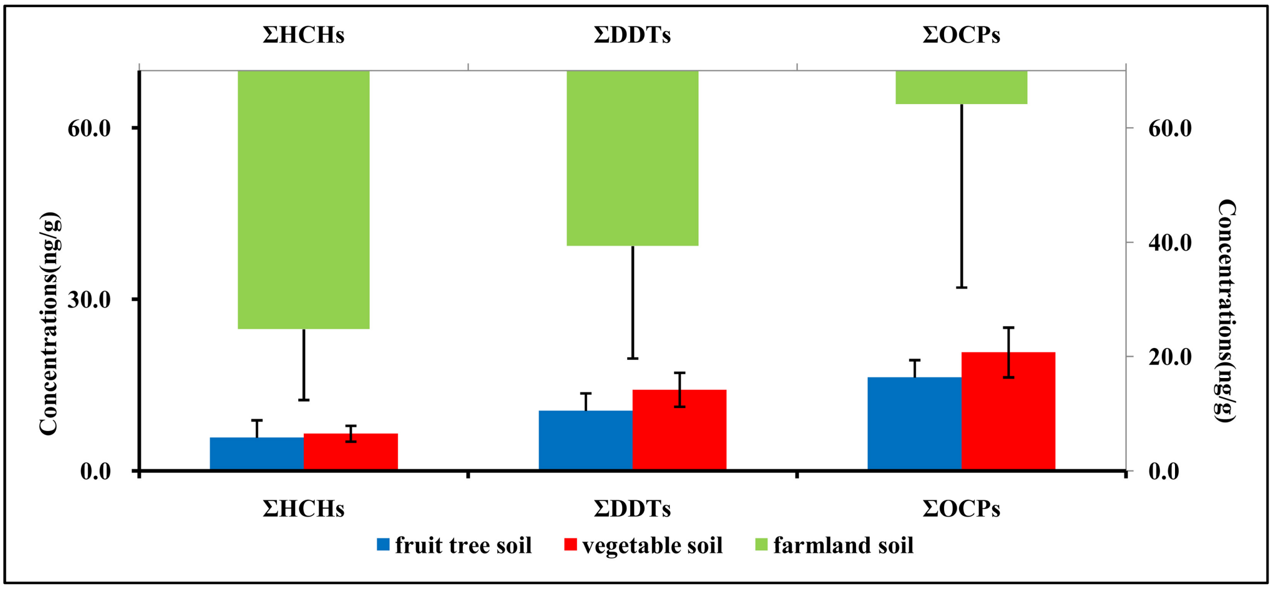


(a) (b)

Figure 3. Concentrations and patterns of OCPs in soil samples of Hohhot City [(a): HCHs; (b): DDTs].



(a)



(b)

Figure 4. Difference of OCPs contents between different soils and soil use types ((a): OCPs patterns in different soils; (b): HCHs and DDTs patterns with different soil use types).

matter is the key factors affecting the OCPs concentrations in soil and is positively correlated with OCPs in soil (Gong et al., 2004; Shao et al., 2008). The concentrations of OCPs in soil varies with different soil use types, the sequence of OCPs residues in soils is farmland soil (64.2 ng/g) > vegetable soil (17.1 mg/g) > fruit tree soil (16.4 mg/g), and these are consistent with prior studies, and these are consistent with prior studies in this region (Zhang et al., 2013). The variation of residual concentrations of OCPs in farmland soils is the largest and lowest in fruit tree soils, which may be caused by different tillage and pesticides usage.

The levels of PTSs in other regions in China, as published in the literature, were compared with those of the present study. For heavy metals, the contamination levels of Pb and Cu in the present study were higher than those reported from the surface soils in other study areas of China, such as Shangdan Valley in Northwest China (Zhuang & Lu, 2020), the farmland soils adjoining steel plants in Tangshan city of Hebei province (Yang et al., 2018), as well as in suburb cropland in Suzhou City of Anhui province (Li, 2013), however, the levels of Zn and Cr in this study were lower than the levels in the surface soil samples collected in Shangdan Valley, Tangshan city and Suzhou City. For OCPs, the levels of ∑HCHs detected in the present study were much greater than those reported from the vegetable soils in Yuanmou County of Yunnan province (Chen et al., 2014), and were compatible with those collected from urban soils in Wuhan (Tadesse, 2021). The contamination levels of HCHs in this study were significantly less than those in soils surrounding the Tanggu Chemical Industrial District of Tianjin (Hou et al., 2013b). The contamination levels of ∑DDTs in this study were higher than those reported from the soils in Shenzhen (Ni et al., 2011), as well as the vegetable soils from Changchun (Zhang et al., 2017). The concentration of ∑DDTs were lower than the levels in soils collected in Tianjin and Wuhan (Tadesse, 2021; Hou et al., 2013b).

3.3. Pollution Assessment of PTS in Soil of Hohhot

As shown in **Figure 5**, the results of the calculation of Geo-accumulation Index (Igeo) for the investigated metals in soil of Hohhot City fluctuated from uncontaminated to moderately contaminate. The investigated metals in this study demonstrated the following mean Igeo values: Pb (2.9) > Cu (1.5) > Cr (−0.31) > Zn (−0.35), revealing that the soil in Hohhot City was not polluted by Cr and Zn (Igeo < 0), moderate polluted by Cu (1 < Igeo < 2), moderate to heavy pollution by Pb (2 < Igeo < 3), and Pb contaminated was the most serious among the tested heavy metals. Classification distribution based on Igeo values for heavy metals is presented in **Figure 5(c)**. For Zn and Cr, the Igeo values range from Class 0 to Class 1, both of which, 25% and 8.3% of the soil samples fell into Class 1, respectively, indicating that there was regional slight pollution of Zn and Cr in soil of this area. For Cu, 83.4% of all the soil samples were in Class 2 with moderate pollution, and 8.3% of the soil samples were greater than Class 3. In case of Pb, 50% of the soil samples categorized into Class 3 with moderate to heavy pollution, and the soil samples belonged to Class 2 with moderate pollution and Class 4 with heavy pollution accounted for 33.3% and 16.7%, respectively.

The potential ecological risk factor of an individual metal (E_r^i) suggested that the severity of pollution of the investigated heavy metals decreased in the following sequence: Pb (64.59) > Cu (22.67) > Cr (2.14) > Zn (1.21) (**Table 4**), which was consistent with the results of Igeo. The ecological risk index of Pb was high (E_r^i > 80) for 30% of all sampling sites, which highlighted the potential risk that soil Pb poses to crops growth and human health. Additionally, only

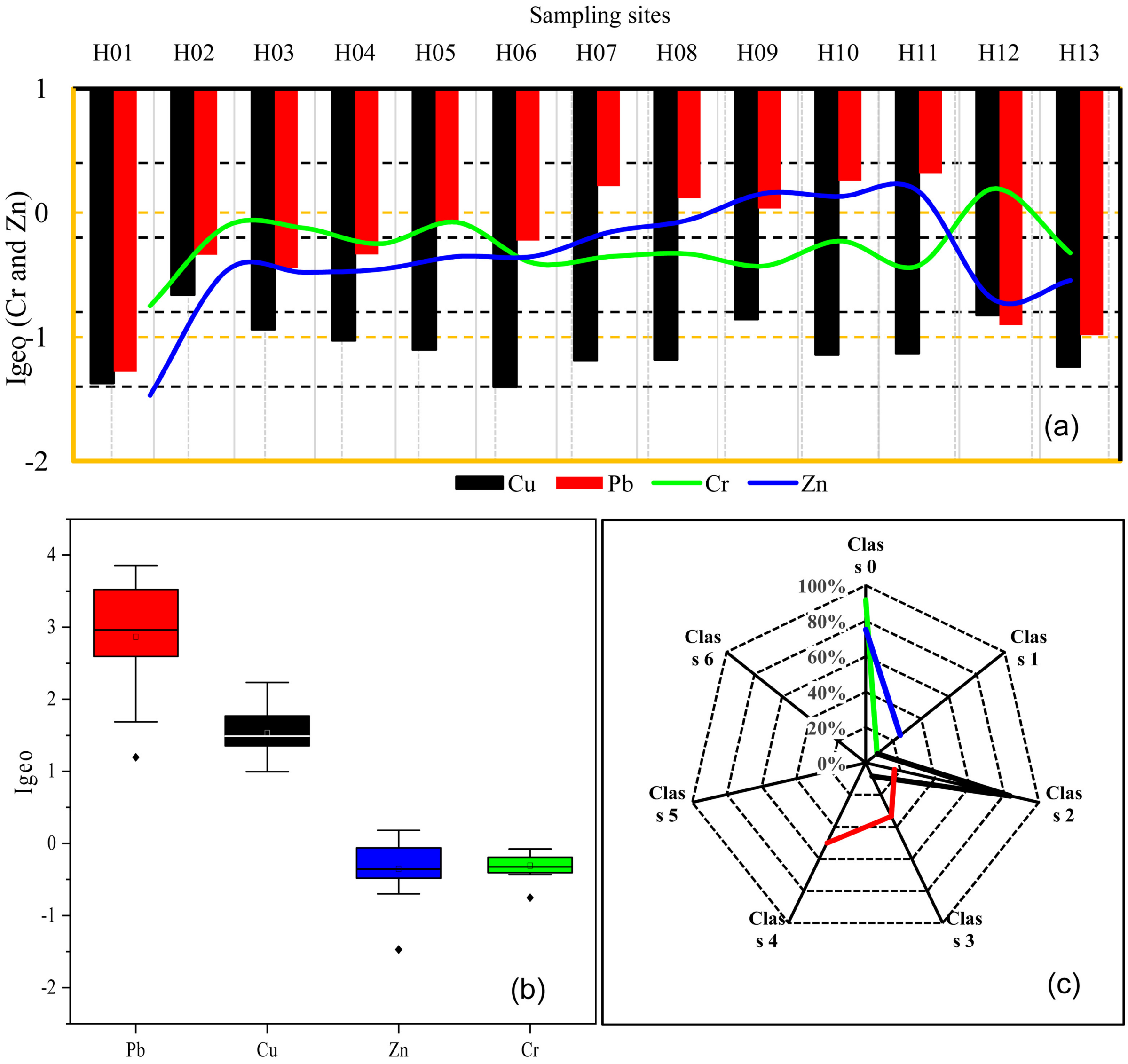


Figure 5. The results of the calculation of Igeo for metals in soil of Hohhot City [(a): Igeo value of Cu, Pb, Zn and Cr in different sampling sites; (b): Variation of Igeo for four metals; (c): Igeo levels of four metals].

Table 4. Potential ecological risk indices (RI) of heavy metals in soil.

	E_r^i				PERI
	Cr	Cu	Pb	Zn	
Min	1.78	14.96	17.19	0.54	35.02
Max	3.43	45.31	108.57	1.73	132.96
Mean	2.14	22.67	64.59	1.21	91.02
Median	2.38	20.48	69.62	1.17	83.69

certain sampling site of Cu in the study area was of moderate potential ecological risk. The range of RI for all sampling sites was from 35.02 to 132.96, indicating low to moderate potential ecological risk, those were mainly due to the fact that Pb and Cu contributes to RI, together indicating that Pb and Cu were identified as the top-priority control heavy metals in the study area.

In the past few decades, HCHs and DDTs were the most popular pesticides used in many countries and had attracted much more attention, and they could be used as representative compounds to evaluate the pollution status of OCPs from different countries and areas. According to GB 15618-1995, the limits for HCHs and DDTs in soils both were 50 ng/g, 500 ng/g and 1000 ng/g, corresponding to Class I, II and III, respectively. In this study, the soil quality of a sampling site was accordingly classed as unpolluted with HCH and DDT concentrations below 50 ng/g; low polluted with HCH and DDT concentrations between 50 ng/g and 500 ng/g; moderate polluted with HCH and DDT concentrations between 500 ng/g and 1000 ng/g; and high polluted with HCH and DDT concentrations above 1000 ng/g. For HCHs, we found that HCHs in all soil samples were less than 50 ng/g. Therefore, the levels of HCHs in soil samples from Hohhot City could be considered as unpolluted. In addition, it was found that the concentrations of DDTs were less than 50 ng/g in 15.3% of the samples; those between 50 ng/g and 500 ng/g were in 84.7% of the samples, and only one sample had DDT levels that exceeded 100 ng/g. So, the levels of HCHs in most samples from the study area of Hohhot could be considered as unpolluted. However, the DDTs in several samples were between 50 and 500 ng/g, which can be regarded as having low polluted.

3.4. Source Identification of PTSs

Pearson's correlation analysis was widely used in the analysis of soil pollutant sources (Dong et al., 2019), and it could be inferred whether there were common behaviors and source of heavy metals in the soils of the research area by studying the correlations between heavy metals and environment parameters. Pearson's correlation coefficients for heavy metals in soils of Hohhot City were summarized in **Table 5**. The results showed that significantly strong positive correlations (r = 0.96, $p < 0.01$) were observed between Zn and Pb, and Cu had moderate positive correlations with Cr with the correlation coefficients of 0.629 ($p < 0.05$), revealing the similar sources for these two group metals. It could be observed that TOC was significantly correlated with Cr and Cu, but weakly correlated with Zn and Pb, which might be related with their chemical properties in soils. Zn and Pb in soil existed mostly in forms of hydroxide or carbonate and enriched in the soil surface, making it difficult to move to deep soils and form complex with TOC. But, the element Cu can be hydrolyzed in soils and converted to stable compounds by complexation.

By studying the principal component analysis results, it can effectively eliminate the spatial diversity of sample variables and facilitate the extraction information among sample variables. The results of principal component analysis of heavy metals were shown in **Table 6**. Two principal components with eigenvalues larger than unity were extracted. The cumulative contribution rate of the

Table 5. Pearson's correlation coefficients of heavy metals, soil pH and TOC in suburban agricultural soil from Hohhot City.

	Cr	Cu	Pb	Zn	TOC	PH
Cr	1.000					
Cu	0.629*	1.000				
Pb	−0.217	−0.259	1.000			
Zn	−0.004	0.015	0.960**	1.000		
TOC	0.503*	0.588*	0.019	0.149	1.000	
pH	−0.184	0.078	−0.041	−0.037	0.832**	1.000

*Correlation is significant at the 0.05 level (2-tailed). **Correlation is significant at the 0.01 level (20 tailed).

Table 6. Eigen values of factors.

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	2.086	52.154	52.154	2.086	52.154	52.154	1.957	48.934	48.934
2	1.540	38.507	90.661	1.540	38.507	90.661	1.669	41.727	90.661
3	0.374	9.339	100.000						
4	9.88E−17	2.47E−15							

Extraction method: Principal component analysis.

two principal component factors was 90.7%, which can explain the majority of information in the raw data and reflect the pollution of heavy metals. The first principal component (PC1) accounted for 52.154% of the total variance, had strong positive loading on Pb and Zn. Contents of the element Pb is much greater than that of the background value, however, the content of Zn is lower than that of the national average soil background value of China but higher than the background value of zinc in tidal soil in Hohhot. All of these show that Pb and Zn in Hohhot farmland are seriously polluted by foreign sources. Principal component 2 (PC2) was dominated by Cr and Cu, and explained 38.507% of the total variance. It was generally believed that Cr was mainly affected by geochemical genesis (Adrie & Bert, 2002), and the element Cr in factor 2 mainly originated from the soil parent material. However, the copper in Hohhot soil might mainly source from intensive agricultural production.

The sources and the fate of HCHs in soil could be identified based on isomer composition. β-HCH would be predominant in most of soil if no fresh inputs of technical HCH while the predominance of γ-HCH in soil samples reflected the recent use of lindane. In present study, the main components of HCHs in soil were β-HCH, ranged from 4.61 ng/g to 35.91 ng/g with a mean of 13.43 ng/g, and accounting for 28.9% to 84.8% of ΣHCHs in soil, with over half of the samples above 60%. The high percentage of β-HCH implied that HCH residues derived mainly from historical usage of technical HCH in this region. Other HCHs concentrations were in a declined order of α-HCH > δ-HCH > γ-HCH, with a mean concentration of 2.82 ng/g, 1.73 ng/g and 0.81 ng/g, respectively. Previous studies have shown that the ratio of α-HCH/γ-HCH ranged from 4 to 7 for technical HCH, the lower α-HCH/γ-HCH ratios suggested the usage of lindane. In this study, the mean ratio of α-HCH/γ-HCH was 3.48, indicating usage of lindane also occurred in this area. Although HCHs had been banned for use in China over 30 years, the situation of the HCHs pollution in agricultural soil in Hohhot City is still stern. Compared with other cities in China, the concentration of HCHs in farmland soil in Hohhot was higher than that in Xi'an (3.56 ng/g) and Changchun (10.95 ng/g), and lower than that in irrigation polluted soil in Tianjin (Lu & Liu, 2013; Liu et al., 2013; Gong et al., 2004).

The relative concentrations of the parent DDT compound and its biological metabolites, DDD and DDE, can be used to assess possible pollution sources. High ratios of p,p'-DDT/∑DDTs suggest a recent input of technical DDT while low ratios indicate accumulated DDTs have undergone long-term degradation. In this study, the concentrations of DDTs in all soil samples followed the order of p,p'-DDD (12.59 ng/g), p,p'-DDE (10.56 ng/g), p,p'-DDT (9.99 ng/g) and o,p'-DDT (3.81 ng/g), and accounting for 34.1%, 28.6%, 27.0% and 10.3% of ∑DDTs, and the highest level of DDTs was found at H10 (105.8 ng/g), and the lowest level of DDTs was found at H1 (5.01 ng/g). The relatively high ratios of p,p'-DDT/∑DDTs, ranging from 0.09 to 0.62 (mean 0.47), indicate that there has been a recent input of technical DDT. Furthermore, the ratios of o,p'-DDT/p,p'-DDT can be used to assess the source of DDTs, the ratios were 0.2 to 0.3 in technical DDT (Lee et al., 2001). In this study, the ratios of o,p'-DDT/p,p'-DDT ranged from 0.05 to 1.62, and ratio in 0.2 - 0.3 were measured in about 25% of sampling sites. The ratios of o,p'-DDT/p,p'-DDT were over 1.3 in about 17% of sampling sites, suggesting an important contribution from technical dicofol (Qiu et al., 2005).

4. Conclusion

In this study, the concentrations and spatial distribution of persistent toxic substances (PTSs) including Cu, Pb, Zn, Cr and OCPs in suburban agricultural soils of Hohhot City were investigated. The higher concentration of Pb, Zn and Cu were presented in the western area and decreased from western to eastern Hohhot, indicating that the intensive agricultural production resulted in soil heavy metals accumulation. According to the local background levels, Pb and Cu appeared at different levels of accumulation in surface soils, especially for Pb with 25% of the sampled soils exceeding the Chinese threshold limit. The ecological risk assessment suggested that Pb contaminated was the most serious among the tested heavy metals. Based on investigation and analysis of ∑DDTs and ∑HCHs, The relatively low α-HCH/γ-HCH ratio and relatively high o,p'-DDT/p,p'-DDT ratio indicated the application of lindane and dicofol on regional agricultural soil. According to GB 15618-1995, HCHs in all soil samples were considered as unpolluted, while DDTs were low polluted. In view of the significant role of intensive agricultural production in PTSs accumulation in suburban agricultural soils, regular monitoring, source control and integrated environmental management should be implemented to control and reduce heavy metals and OCPs inputs and guarantee the safety of agricultural production.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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