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Pollution Status of Agricultural Land in China: Impact of Land Use and Geographical Position

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Abstract

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According to the Soil Pollution Prevention and Control Action Plan released in May 2016, the soil quality of 666 666.7 ha of agricultural soil requires remediation before 2020. Despite the survey on the environmental quality of soil in China released in 2014, detailed data on current pollutant concentrations remain unavailable. To date, reports on soil environmental quality on the national scale are few. The current research aimed to gain a detailed understanding of soil pollution in China through literature study and data analysis. Data for eight potentially toxic elements (i.e., arsenic (As), cadmium (Cd), zinc (Zn), chromium (Cr), mercury (Hg), copper (Cu), nickel (Ni), and lead (Pb)) and two organic pollutants (i.e., hexachlorocyclohexane (HCH) and dichlorodiphenyltrichloroethanes (DDTs)) were collected from 367 areas involving 163 prefecture-level cities. Principal component analysis and clustering analysis were conducted to understand the relationships among pollutants. Results indicated that organic pollution was less severe than inorganic pollution. In terms of land-use types, garden soil showed the heaviest pollution, followed by arable land and woodland. Regarding geographic distribution, the south central and southwest areas displayed heavy pollution. Principal component and clustering analyses revealed that As, Hg, HCH, and DDTs were mainly contributed by anthropogenic sources; Cr, Cu, Ni, and Zn were primarily caused by natural background; and Cd and Pb were contributed by both sources. The soil pollution status varied among land-use types and geographic areas. The implementation of proper remediation strategies requires detailed investigations on soil environmental quality.

Keywords: environmental hazard; land use; organic contaminants; soil contamination; toxic elements

Soil pollution is much less visible than water and air pollution (Azizullah *et al.* 2011; Henschel *et al.* 2012). Therefore, soil pollution has only recently attracted considerable attention. Soil quality is directly related to food safety, human health, and sustainable economic and social development (Teng *et al.* 2014). Given the high health risks associated with soil pollution, soil pollution control is gradually receiving research attention (Beesley *et al.* 2011; Ali *et al.* 2013).

On April 17, 2014, the first results of a nationwide soil pollution survey revealed the pollution of one-fifth of agricultural land with inorganic chemicals, such as cadmium, nickel, and arsenic. Accordingly, the Soil Pollution Prevention and Control Action Plan was released in May 2016, which stipulated that 666 666.7 ha of contaminated agricultural land should be remediated before 2020 (CSC 2016).

Despite the released brief report on the national soil environmental quality, detailed data on soil pollution were not published. Therefore, the pollution distribution and related land-use types are unknown. A clear understanding of the soil pollution status is urgently needed.

The main contaminants in soil include inorganic and organic pollutants. Inorganic pollutants are mainly

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toxic elements (TEs), including heavy metals and As (which is often studied with heavy metals given its similarity to their toxicity). Such pollutants are difficult to remove because they cannot be degraded in soil. The entry of TEs into the food chain may pose risks to human health and environmental safety. A national survey revealed that TE contamination in China is severe, indicating a higher risk than observed organic pollutants. Investigations were conducted on soil TE pollution and its control measures (Hu *et al.* 2014; Fatoba *et al.* 2016). Conversely, organic pollutants mainly include organic chlorine, such as hexachlorocyclohexane (HCH) and dichlorodiphenyltrichloroethane (DDT), which are difficult to decompose and pose potential risks to the environment (Guzzella *et al.* 2005; Shi *et al.* 2005).

Soil is an extremely heterogeneous medium. The distribution of pollutants can vary from hundreds to thousands of times in the national scale (Chen *et al.* 1997; Shi *et al.* 2016). According to the national survey, soil pollution in different geographic areas varies significantly due to the natural background and anthropogenic input. Land use is another essential influential factor of soil environmental quality (Zhao *et al.* 2013). Varied extents of pollution materialize under dissimilar land-use types because of the diversity in agricultural input and cultivation habits.

To acquire a better understanding of the pollution status of agricultural land, the current study summarized the published data on soil environmental quality from 2000 to 2016, analyzed the distribution of pollutants in various areas of China, and examined the pollution levels of different land-use types.

MATERIAL AND METHODS

Soil environmental quality data were collected from literature published from 2000 to 2016. Two databases were searched: the China Knowledge Resource Integrated Database (the main database for Chinese research) and Web of Science. The literature selection criteria were as follows: (1) data must be from the municipal administrative level, taking into account the provincial and county administrative units, and avoiding research on mine, sewage irrigation, and other special small-scale soil pollution cases; (2) the data collection site must be agricultural land, including arable land, gardens, woodlands, and grasslands; (3) data must be published after 2000 to enable a comparison of soils located in different regions; (4) the original pollutant concentrations should be provided; (5) the sampling and analysis methods of pollutants should be of international or domestic standards to facilitate the comparison of results (details described in the next paragraph). The studied pollutants include arsenic (As), cadmium (Cd), zinc (Zn), chromium (Cr), mercury (Hg), copper (Cu), nickel (Ni), and lead (Pb), and organic pollutants HCH and DDTs. Through sample collection and pollutant analysis methods, 167 papers were chosen from more than 2000 papers.

In the current study, agricultural land refers to land directly used for agricultural production, including arable land (ploughed land), gardens (for planting perennial woody and herbaceous plants intended for intensive fruit and leaf management), woodlands (natural, secondary, or man-made forests), and grasslands (for growing herbs or shrubs aimed to develop livestock production) (NPC 1986).

Data of soil environmental quality were collected from 367 areas involving 163 prefecture-level cities. Those areas were categorized into arable land (159 areas), gardens (77 areas), woodlands (30 areas), grasslands (20 areas), and agricultural land (without discrimination of land-use types, 81 areas).

China was divided into six areas: north, northeast, northwest, south central, southwest, and east. This division was in accordance with the genetic soil classification of China (Figure 1). The typical agricultural soil type was Semi-Luvisols in the north, Pedocal and Luvisols in the northeast, Entisols and Inceptisols in the northwest, Ferralsols in the south central area, Ferralsols and Anthrosol in the southwest, and Ferralsols and Luvisols in the east (Shi *et al.* 2006).

In the incorporated literature, grid method was used to collect samples from the top soil (0–20 cm layer). Diverse chemical analysis methods for the

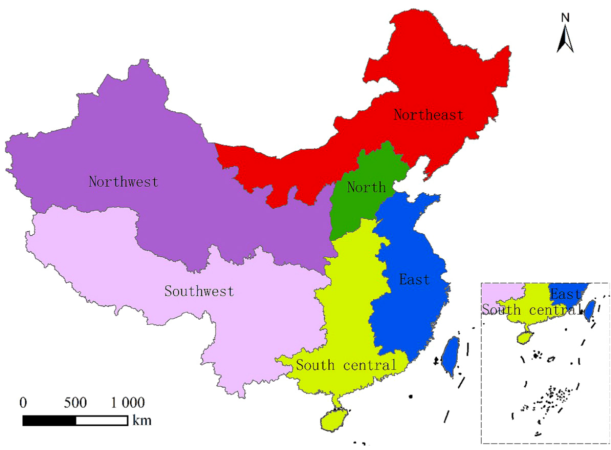

Figure 1. Geographic division of China

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concentrations of inorganic and organic pollutants in the soil samples were performed in the reviewed studies, but all the research indicated acceptable quality control scenarios and recovery rates.

To analyze TE concentrations, we used the two most popular digestion and analysis methods: (1) Method 3050B of the United States Environmental Protection Agency and (2) GB 15618-1995, the method regulated by the Environmental Quality Standard for Soils in China. Certified standard reference materials for soils from the China National Standard Materials Center were examined together with the experimental samples. The measurement results were within the permissible error ranges of the known values of the reference materials.

In the analysis of DDTs and HCHs, the most common method was gas chromatography for the determination of BHC and DDT in soil (GB/T 14550-2003). For quality control, the usual standard addition recovery method was employed. All the incorporated literature recorded a recovery rate higher than 80%.

An over-standard rate indicates the percentage of samples with TE concentrations higher than those recommended by China's Environmental Quality Standard for Soils (GB15618–1995, Grade II) in all the collected samples. The said Standard for Soils regulates the maximum permitted concentrations of pollutants in soil, and the Grade II rating applies to agricultural soil.

Data were analyzed using SPSS software (PASW Statistics for Windows, Ver. 18.0., Chicago, SPSS Inc.). Principal component analysis and clustering analysis were conducted to understand the relationships among pollutants. The former calculates new variables (principal components) that are linear combinations of the original variables, thereby accounting for the variability in the data and enabling the study of covariances or correlations between variables (Marques *et al.* 2008). The latter facilitates the grouping of a set of objects such that objects in the same cluster are more similar to one another than to objects from other clusters.

RESULTS AND DISCUSSION

Concentrations and over-standard rates of pollutants under different land-use types

Average concentration. The average concentrations of pollutants (in mg/kg) were determined: Cd (0.69), As (13.99), Zn (149), Cr (61.66), Hg (0.27), Cu (32.03), Ni (31.60), Pb (44.20), HCH (0.004), and DDT (0.04; Table 1). Except for Cd, all the average concentrations were lower than the standard.

Pollutant concentrations also varied under different land-use types. Garden soil pollution was the most serious, and it had the highest concentrations of As, Cu, Hg, Ni, and Pb. The average As concentration of garden soil was 19.1 mg/kg, which was 102% higher than that of grassland soil. The average Hg concentration of garden soil was 0.68 mg/kg or 580% higher than that of grassland soil. Woodland showed the highest Cd and Cr concentrations, whereas arable land had the highest Zn concentration.

Over-standard rate. The over-standard rates of pollutants were as follows: Cd (29.6%), As (3.88%), Zn (4.52%), Cr (0.86%), Hg (8.47%), Cu (2.72%), Ni (7.14%), Pb (1.4%), HCH (0%), and DDT (1.54%; Table 2). These levels were in accordance with the national soil pollution survey data. Inorganic pollutants showed heavier contamination than organic pollutants. The over-standard rates calculated from the literature were higher than the national soil pollution data. This disparity was due to the fact that the national survey data provided the over-standard rate for all the surveyed data without distinguishing among land-use types. Considering that the over-standard rates of arable land and garden soil were higher than those of grassland, woodland, and unused land, the high over-standard rate obtained in this literature review was reasonable.

Cd was the most significant pollutant with an over-standard rate of 29.63%. Hg, Ni, Zn, and As also showed high over-standard rates. Similar to the concentration results, the over-standard rates suggested that garden soil had the highest pollution. The over-standard rate of Cd in garden soil was 42.86%, which was much higher than the rest of the land-use types. The over-standard rates of Zn, Cr, Hg, and Ni were also highest in garden soil. Unexpectedly, woodland, instead of arable land, showed the second highest over-standard rate. The over-standard rates of As, Cu, and Pb were highest in woodlands. Grasslands showed the least pollution, with the lowest over-standard rates of Cd, As, Zn, Cr, Cu, Ni, Pb, HCH, and DDT. Organic pollution was more serious in arable land than in the rest of the land-use types.

From the current literature study, the pollution level followed the order of garden > arable land > woodland > grassland. Garden and arable land pollution were comparatively higher than pollution in

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other land-use types because of the large amount of agricultural inputs. Grasslands are less prone to human disturbance, and they are often located in areas with limited populations.

A similar trend was found in several previous small-scale studies. In a sewage irrigation region in northwest China, metal concentrations in different land uses were in the order of orchard > paddy field/maize field > village > barren hill (Liu *et al.* 2016). An investigation of the Huabei Plain in north China reported that pollution is more severe in bare and greenhouse vegetable fields than in grain crop fields (Huang & Jin 2008). Excessive agrichemical application may contribute to pollutant accumulation in garden soil. In addition, different land-use types may influence soil properties (such as carbon, nitrogen, potassium, and phosphorus contents) and soil mechanical compositions (Azizullah *et al.* 2011), which may further aggravate soil pollution. In some areas, the high background value also contributed to the high concentrations of TEs (Cutillas-Barreiro *et al.* 2016).

Concentrations and over-standard rates of pollutants in different geographic areas

Average concentration. In general, TE pollution is more serious in south central and southwest China than

Table 1. Concentrations of pollutants in soil under different land-use types (in mg/kg)

		Cd	As	Zn	Cr	Hg	Cu	Ni	Pb	HCH	DDT
Agricultural land (*n* = 367)	average	0.69	13.9	149.0	61.7	0.27	32.0	31.6	44.2	0.004	0.004
	min	0.01	0.05	1.13	0.53	N.D.	1.17	0.93	0.77	N.D.	N.D.
	max	40.6	328.0	3598	370.0	12.7	175.0	129.9	976.9	0.16	0.51
	SD	4.9	27.1	404.5	40.2	0.95	27.2	20.2	74.8	0.03	0.07
Arable land (*n* = 159)	average	0.75	13.8	178.3	65.7	0.20	33.3	32.2	45.3	0.03	0.07
	min	0.04	1.70	43.6	0.92	N.D.	2.50	12.7	11.2	N.D.	N.D.
	max	40.6	93.4	3598	370.0	1.79	111.0	111.5	976.9	0.11	0.51
	SD	4.01	14.3	503.6	38.7	0.29	19.1	15.8	93.0	0.04	0.15
Garden (*n* = 77)	average	0.58	19.1	117.6	58.6	0.68	35.3	35.7	56.2	0.01	0.03
	min	0.03	0.05	26.1	5.50	0.03	1.80	8.10	10.4	N.D.	N.D.
	max	9.95	328.0	480.2	150.8	12.7	175.0	101.3	360.0	0.05	0.17
	SD	1.46	52.1	95.6	29.7	2.25	26.4	20.4	69.2	0.02	0.05
Woodland (*n* = 30)	average	0.94	18.6	76.5	76.6	0.26	28.6	37.6	35.8	0.01	0.00
	min	0.07	1.37	22.6	9.24	0.02	2.30	5.99	9.05	0.00	0.00
	max	6.06	92.5	214.7	315.0	1.5	168.6	129.9	132.3	0.02	0.01
	SD	1.63	30.7	41.2	82.5	0.47	32.5	39.6	32.6	0.01	0.00
Grassland (*n* = 20)	average	0.75	9.47	89.2	32.3	0.10	28.9	16.9	20.2	0.003	0.001
	min	0.01	3.21	27.10	5.33	0.02	13.8	5.19	3.16	N.D.	N.D.
	max	3.28	19.4	218.3	59.3	0.40	73.7	25.2	28.5	0.022	0.0074
	SD	1.42	6.0	62.0	19.0	0.15	22.5	10.5	8.08	0.007	0.002
Recommended value*	pH < 6.5	0.30	40.0 (30.0$)	200.0	150.0 (250.0£)	0.30	50.0 (150.0#)	40.0	250.0	0.50	0.50
	6.5 < pH < 7.5	0.30	30.0 (25.0$)	250.0	200.0 (300.0£)	0.50	100.0 (200.0#)	50.0	300.0	0.50	0.50
	pH > 7.5	0.60	25.0 (20.0$)	300.0	250.0 (350.0£)	1.0	100.0 (200.0#)	60.0	350.0	0.50	0.50

SD – standard deviation; N.D. – below the detection limit; *recommended value indicates the value regulated by China's Environmental Quality Standard for Soils (GB15618–1995, Grade II), which applies to agricultural soil, including farmland, garden, woodland, and grassland; $recommended value for As concentration specifically in paddy soil, regulated by China's Environmental Quality Standard for Soils (GB15618–1995, Grade II); £recommended value for Cr concentration specifically in paddy soil, regulated by China's Environmental Quality Standard for Soils (GB15618–1995, Grade II); #recommended value for Cu concentration specifically in orchard soil, regulated by China's Environmental Quality Standard for Soils (GB15618–1995, Grade II)

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Table 2. Over-standard rates of pollutants in soil under different land-use types (in %)

	Agricultural land (*n* = 367)	Arable land (*n* = 159)	Garden (*n* = 77)	Woodland (*n* = 30)	Grassland (*n*=20)
Cd	29.60	30.50	42.90	21.10	11.10
As	3.88	4.72	9.09	10.00	0.00
Zn	4.52	4.08	9.62	3.70	0.00
Cr	0.86	0.97	1.92	0.00	0.00
Hg	8.47	3.19	32.40	18.20	11.10
Cu	2.72	1.96	1.72	3.70	0.00
Ni	7.14	2.00	19.40	12.50	0.00
Pb	1.40	0.72	3.33	3.57	0.00
HCH	0.00	0.00	0.00	0.00	0.00
DDT	1.54	9.09	0.00	0.00	0.00

in other parts of China. Average concentrations of TEs that exceeded the recommended values included Cd in the northeast and south central areas, Zn in northeast areas, and Hg in south central and northwest areas (Table 3). Except for Cd and Zn, the concentrations of other TEs were highest in south central or southwest areas. The concentrations of Cd and Zn were highest in the northeast areas. Several elevated values led to the unexpected high Cd and Zn concentrations in the northeast because of the limited number of large-scale investigations in this region. Organic pollution was less serious than TE pollution. HCH and DDT concentrations were below the recommended values of the China Environmental Quality Standard for Soils. Unlike the TE distribution, organic pollutants showed high concentrations in the north and northeast.

Over-standard rate. Regarding pollution distribution, the over-standard rates of TEs were apparently higher in the south central and southwest areas than in the other areas (Table 4), and this finding may be related to high initial values and active mining activities.

Similar to the concentration results, the over-standard rates were highest for Cd. High over-standard rates were also observed at the national level. The highest over-standard rate of Cd reached 55.56% in the northwest. Hg, Ni, and As also exhibited high over-standard rates. High over-standard rates of As, Zn, and Cr were observed in the east and south central areas. For organic pollutants, the over-standard rates were highest in the east. Coastal areas in the east were the main production base of small electric

Table 3. Concentrations of pollutants in soil at different areas of China (in mg/kg)

	North	Northeast	East	South central	Southwest	Northwest	Recommended value
Cd	0.25	3.46	0.39	1.96	0.43	0.41	0.30
As	9.59	8.46	14.4	11.2	18.3	13.6	30.0
Zn	78.8	303.1	98.6	218.5	119.5	94.5	250.0
Cr	62.9	54.8	56.9	74.5	66.4	64.5	200.0
Hg	0.30	0.15	0.25	0.75	0.16	0.72	0.50
Cu	26.4	35.8	35.9	33.7	47.7	32.1	100.0
Ni	33.2	23.5	29.4	43.3	25.2	40.6	50.0
Pb	24.9	50.2	43.9	75.4	38.6	39.5	300.0
HCH	0.05	0.01	0.01	0.02	0.00	0.00	0.50
DDT	0.02	0.02	0.05	0.01	0.00	0.00	0.50

North: Beijing, Tianjin, Hebei, Shanxi, and Inner Mongolia; Northeast: Liaoning, Jilin, and Heilongjiang; East: Shanghai, Jiangsu, Zhejiang, Anhui, Fujian, Jiangxi, and Shandong; South Central: Henan, Hubei, Hunan, Guangdong, Hainan, and Guangxi; Southwest: Chongqing, Sichuang, Guizhou, Yunnan, and Tibet; and Northwest: Shaanxi, Gansu, Qinghai, and Xinjiang

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Table 4. Over-standard rate of pollutants in soil at different areas of China (in %)

	North	Northeast	East	South central	Southwest	Northwest
Cd	12.90	36.80	21.90	42.10	41.40	55.6
As	0.00	0.00	6.15	7.14	4.00	0.00
Zn	2.22	5.88	4.49	8.82	6.25	0.00
Cr	0.00	0.00	1.14	5.26	0.00	0.00
Hg	18.90	9.52	7.69	17.40	0.00	28.60
Cu	0.00	6.25	3.33	5.71	10.00	0.00
Ni	12.50	0.00	2.13	22.70	0.00	20.0
Pb	0.00	4.00	1.01	2.44	1.41	0.00
HCH	0.00	0.00	3.23	0.00	0.00	0.00
DDT	0.00	0.00	3.45	0.00	0.00	0.00

North: Beijing, Tianjin, Hebei, Shanxi, and Inner Mongolia; Northeast: Liaoning, Jilin, and Heilongjiang; East: Shanghai, Jiangsu, Zhejiang, Anhui, Fujian, Jiangxi, and Shandong; South Central: Henan, Hubei, Hunan, Guangdong, Hainan, and Guangxi; Southwest: Chongqing, Sichuang, Guizhou, Yunnan, and Tibet; and Northwest: Shaanxi, Gansu, Qinghai, and Xinjiang

products. The unregulated disposal of waste residue and waste water irrigation in the past may have caused the high concentrations of organic pollutants in the east (Sun *et al.* 2018).

Present large-scale investigations on soil pollution are limited. Most studies focused only on soil environmental quality at small-scale levels (Wang & Stuanes 2003; Yang *et al.* 2011). A national survey revealed that soil pollution in China is more severe in the south than in the north. An apparent phenomenon for the over-standard rates of TEs was found in the south central and southwest areas. Heavy TE pollution was observed in the said regions. These results were in accordance with findings of the current study and in line with the distribution of soil genetic classification in China. The typical agricultural soil types in south central and south west were Ferrasols and Anthrosol. Ferrasols are characterized by low pH and low availability of phosphorus (P). They are formed in tropical and sub-tropical climate and adequate precipitation, which can increase the bio-availability of TEs in the form of cations; by contrast, Anthrosols are characterized by large amounts of agricultural inputs, which inevitably introduce pollutants (Lehmann *et al.* 2003). Climate and parent material are two key factors that determine soil classification, and they both significantly influence the transfer of pollutants in soil (Žigová & Šťastný 2014). Therefore, soil genetic classification is suggested as a possible reason for heavy soil contamination in the south central and southwest areas. Further studies are needed on the distribution of pollutants in the soil profile, and the bio-availability of metals is required for additional information.

Relationships among pollutants

Correlation analysis of pollutant concentrations indicated that Cd was positively related to Zn, Cu,

Table 5. Correlation analysis of pollutants

	Cd	As	Zn	Cr	Hg	Cu	Ni	Pb	BHC
As	0.072								
Zn	0.765**	0.189*							
Cr	–0.066	–0.005	0.121						
Hg	–0.007	–0.014	–0.002	0.034					
Cu	0.514**	0.489**	0.429**	0.141	0.118				
Ni	0.269**	0.094	0.084	0.752**	0.105	0.315**			
Pb	0.343**	0.368**	0.812**	0.037	0.083	0.385**	0.099		
HCH	0.641*	0.498	0.223	0.379	–0.223	0.135	0.504	0.153	
DDT	0.784**	0.676*	0.584	0.518	–0.194	0.453	0.612	0.319	0.202

*, **significant at the level of 0.05 and 0.01

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Table 6. Total variance from principal component analysis

Component No.	Eigen value	Cumulative variance contribution (%)
1	7.133	71.326
2	2.119	92.516
3	0.506	97.576
4	0.228	99.857
5	0.014	100

Ni , Pb, HCH, and DDT concentrations ($P < 0.05$; Table 5). The remaining pollutants were all positively related to one or several other pollutants.

Two principal components were extracted, reflecting 92.5% of all studied pollutants (Table 6). The two extracted components can be described as follows:

$$Y1 = 0.128\ (Cd) + 0.121\ (As) + 0.109\ (Zn) + \\ + 0.112\ (Cr) + 0.127\ (Hg) + 0.099\ (Cu) + \\ + 0.135\ (Ni) + 0.138\ (Pb) + 0.077\ (HCH) + \\ + 0.125\ (DDTs) \quad (1)$$

$$Y2 = 0.161\ (Cd) + 0.18\ (As) - 0.262\ (Zn) - \\ - 0.275\ (Cr) + 0.184\ (Hg) - 0.324\ (Cu) - \\ - 0.066\ (Ni) - 0.047\ (Pb) + 0.33\ (HCH) + \\ + 0.124\ (DDTs) \quad (2)$$

Y1 was mainly contributed by inorganic pollutants, whereas Y2 was chiefly generated by organic pollutants.

Further clustering analysis indicated that inorganic and organic pollutants followed different patterns that belonged to two distinct clusters (Figure 2). Among inorganic pollutants, Cd, As, and Hg were in the same cluster that was affected by human activities, whereas Cr, Cu, Ni, Zn, and Pb were in another cluster that was affected by both human activities and natural background.

Numerous studies have been conducted to differentiate the anthropogenic sources of TEs from the natural background. As and Hg are commonly regarded as being contributed by anthropogenic sources; Cr, Cu, Ni, and Zn are mainly contributed by the natural background value; and Cd and Pb are contributed by both sources (Meng *et al*. 2014; Zhou *et al*. 2015;Zou *et al*. 2015). The involvement of natural background may require the establishment of geochemical background concentrations to distinguish the natural background from the anthropogenic concentrations of TEs in soils (Dung *et al*. 2013).

CONCLUSION

Through literature collection and analysis, the current study obtained a fundamental understanding of agricultural soil pollution in China. In terms of the pollutants included in this work, the observed organic pollution was less severe than inorganic pollution. For different land-use types, garden soil showed the heaviest pollution, followed by arable land and woodland. In terms of geographic distribution, south central and southwest areas displayed heavier pollution than the other areas. Principal component and clustering analyses showed a clear

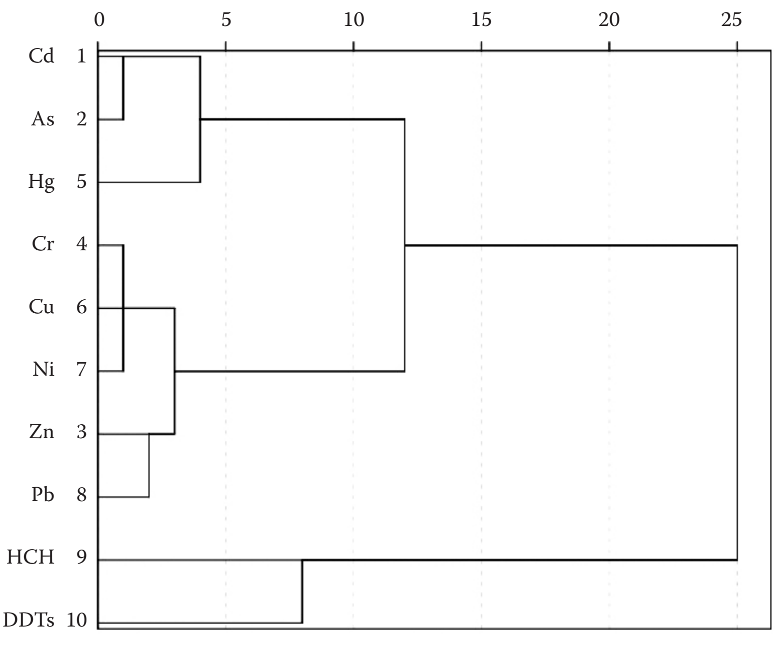

Figure 2. Clustering analysis of pollutants

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difference between organic and inorganic pollutants, suggesting their varied pollution sources. TEs had two sources, namely, anthropogenic and natural sources. As and Hg were commonly regarded as contributed by anthropogenic sources; Cr, Cu, Ni, and Zn were mainly caused by natural background value; and Cd and Pb resulted from both sources.

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