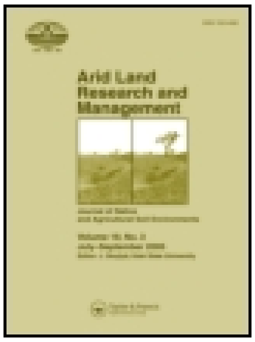

Arid Land Research and Management

ISSN: 1532-4982 (Print) 1532-4990 (Online) Journal homepage: http://www.tandfonline.com/loi/uasr20

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ARID LAND RESEARCH AND MANAGEMENT
2016, VOL. 00, NO. 00, 1–11
http://dx.doi.org/10.1080/15324982.2015.1128013

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Evaluation of semi-arid arable soil heavy metal pollution by magnetic susceptibility in the Linfen basin of China

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ABSTRACT

The values of magnetic susceptibility and Cu, Zn, Ni, Pb, Cr, Cd concentrations of 70 topsoil samples were analyzed and assessed for soil contamination in the Linfen basin of China. The contamination factor (CF) and pollution load index (PLI) were used to assess the degree of heavy metal pollution. All heavy metal concentrations in the soil were greater than the background values of Shanxi agricultural soils. PLI values ranged from 1.27 to 2.18 indicating significant soil contamination. Principal component analyses and correlation analyses were adopted for data treatment to identify heavy metal sources. Cu, Zn, Ni, and Pb were responsible for 31.2% of the total variance suggesting that these elements mainly originated from agrochemistry and atmospheric deposition. A positive correlation of magnetic susceptibility with Cu and PLI were observed in cultivated soils. This study shows that magnetic susceptibility measurements can be used as a technique in which different types of soil pollution can be distinguished and can also be used to rapidly monitor anomalies in areas that might require expensive and time consuming detailed chemical analyses.

ARTICLE HISTORY
Received 7 February 2015
Accepted 1 December 2015

KEYWORDS
Heavy metal contamination; magnetic susceptibility; semi-arid Linfen basin; topsoil

Introduction

The rapid social and economic development of China over the past several decades has led to increasingly serious and widespread levels of soil pollution by heavy metals within the country (Luo et al. 2009; Shan et al. 2013). The heavy metal content in cultivated soils is due to a variety of different sources including the local bedrock/parent materials and agricultural practices, industrial emissions, waste treatments and vehicular traffic. Another major source of heavy metals in soils is atmospheric deposition which is mainly caused by industrial processes such as coal burning, metal refining, steel-iron manufacturing and transportation. In topsoil 85% Pb, 67.5% Ni, and 43% Cr associated with industrial processes are thought to come from atmospheric deposition (Luo et al. 2009).

Evidence suggests that this accumulation of heavy metals in arable land may lead to absorbtion by crops that could lead to adverse effects in human health (Bai et al. 2010).

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The evaluation of soil heavy metal risk is often defined in terms of its contamination factor (CF) and its Tomlinson pollution load index (PLI) (Tomlinson et al. 1980). CF is a quantitative expression of the degree of each metal in soil whereas PLI index is used to provide a simple integrated parameter of heavy metal pollution.

Magnetic susceptibility (χ) measurement technique offers a relatively simple, rapid, sensitive, and inexpensive method for monitoring heavy metal pollution (Thompson, Bioemendal, and Dearing 1980; Duan et al. 2010; Jordanova et al. 2013; Yang et al. 2015). Taking a measurement of χ in the field or in the lab takes just a few seconds in an approach that is less direct but faster than the use of portable XRF-scanners. It can provide a rapid, simple, nondestructive approach that is best used in a pre-screening site assessment to most effectively target subsequent chemical sampling (Appel et al. 2003). Recent studies have shown significant relationships between χ and Cr, Cu, Pb, and Zn in urban and industrial soils, river sediments, and dust. This includes studies in Turkey, Mexico, Estonia and south France etc. (Hay et al. 1997; Bityukova, Scholger, and Birke 1999; Lecoanet, Leveque, and Ambrosl 2003; Lourenco et al. 2014) which showed that magnetic susceptibility can be used as a proxy for chemical methods due to the fact that pollutants are often closely related to magnetic phases (Canbay, Aydin, and Kurtulus 2010). This is due to the fact that magnetic carriers can often absorb and/or incorporate heavy metals in their crystalline structure (Morton-Bermea et al. 2009). It is thought that magnetic signals originating from soils or sediment might prove useful for reconstructing the development of the pollution history of the surrounding areas. Previous studies have largely focused on urban environments and industrial regions however to date few studies have used magnetic susceptibility to investigate the accumulation of heavy metals in cultivated soils.

The aim of the study were (1) to evaluate the magnetic susceptibility and heavy metal concentration using the pollution load index from a selected agricultural area, (2) to examine the relationship between magnetic susceptibility and heavy metal content by correlation analysis, and (3) to distinguish the types of pollution sources in cultivated soil.

Materials and methods

Study area

The study area was located at latitude 35° 42′-36 °02′ N, longitude 111°04′-111° 39′E in the southern region of the Linfen basin in Shanxi province. Linfen city is well known for intense coal mining, coke, iron-steel heavy industry (Yin et al. 2013; Cao et al. 2015). The Lüliang and Zhongtiao Mountain is situated in the west and east of Linfen, respectively. The region has a semi-arid and semi-humid continental climate, with the most predominant wind direction during winter coming from the northwest, leading to soil pollution and accumulation in the south. The area covers almost 1000 km^2, of which approximately 60% corresponds to farmlands. The mean annual temperature is 12.6°C and has an average annual rainfall of 475 mm with more than 60% of the precipitation falling between July and September, during which the average evaporation is 1780 mm. The soil types are Calciustepts and Haplustepts according to USDA soil Taxonomy 2014, consisting of alluvial deposits, developed on Quaternary loess. The soil textures are silt loam and sandy loam, with pH values ranging from 7.5 to 8.5, organic matter content ranging from 0.59% to 3.75%, and $CaCO_3$ content changing from 10% to 25%

(Shanxi Soil). It is an important agricultural region for cereals, vegetables, fruit trees, among others, and also contains an industrial center which is only 20 km away from Linfen city. All of these factors make the study area a suitable location for a detailed magnetic survey and geochemical analysis to assess heavy metal pollution in soil.

Soil sampling and analysis

Seventy topsoil samples (0–20 cm) were collected from arable soils during summer 2013 at random locations with an average density of approximately one sample per 9 km^2. The geographic coordinates of soil sampling were recorded using a Trimble GPS (Figure 1). Each sample was extracted from five locations within a 2 m wide area, and stored in polyethylene bags weighting approximately 1.0 kg. Each of the samples were then air-dried, homogenized and passed through a 2 mm mesh. About 0.5 g was digested with a 5:5:3 mixture of HNO_3/HF/$HClO_4$. The target pollutants Cu, Cr, Cd, Pb, and Zn were measured using an S2 Flame Atomic Absorption Spectrometry (U.S.A.) in the laboratory. A reference soil from the Chinese National Standard Soil Bank was used and compared against samples for checking accuracy of metal analysis. Quality assurance and control were ensured by using duplicates and blanks method (Cicchella et al. 2008; Bai et al. 2011).

The magnetic susceptibility was measured using a Bartington MS 2B dual frequency system at low frequency (χ_{lf}) and high frequency kHz (χ_{hf}) (Bartington instruments, UK). Frequency dependent susceptibility ($\chi_{fd\%}$) was then calculated from the expression, $\chi_{fd}\% = (\chi_{lf}-\chi_{hf})/\chi_{lf} \times 100$.

The CF is the ratio of each heavy metal concentration to the background value of the corresponding metal in the soil, $CF = C_{metal}/C_{background}$. The geochemical baseline for

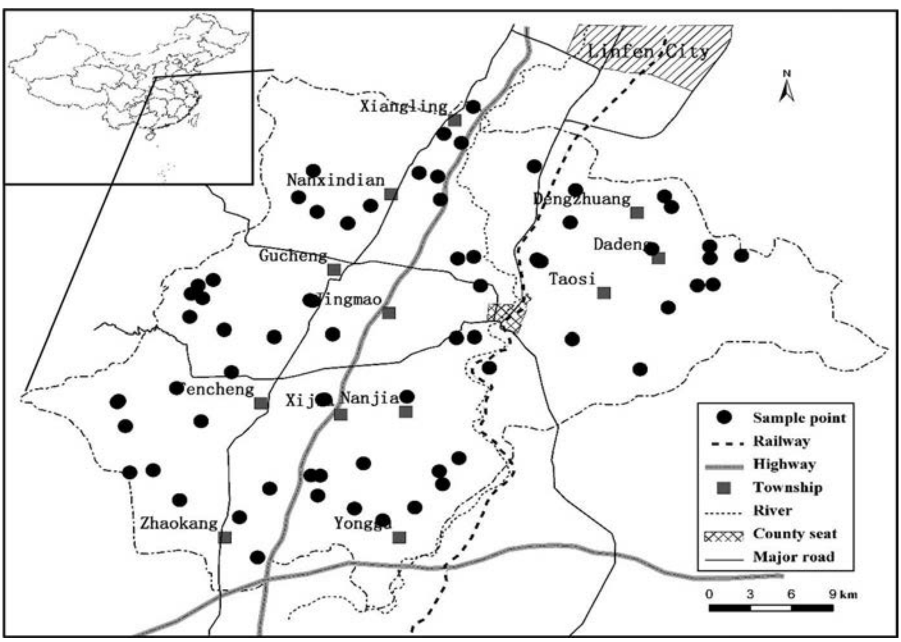


Figure 1. Schematic map of the study area and soil sampling.

Shanxi province agricultural soils of China National Environmental Monitoring Center (CNEMC) (1990) was used as the background value for each heavy metal respectively. PLI is determined from the n^{th} root of the different metal concentration factor, i.e., PLI = $(CF_1 \times CF_2 \times CF_3 \times \ldots CF_n)^{1/n}$(Tomlinson et al. 1980; Angulo 1996).

Multivariate statistical approaches were applied for data analysis using SPSS 17.0 software with significance based on probability 0.05. Spatial variation of each heavy metal in the topsoil was presented using Arc GIS 10.0 software.

Results and discussion

Soil magnetic susceptibility

Low frequency magnetic susceptibility (χ_{lf}) is a parameter that is sensitive to the concentration of ferrimagnetic minerals (Thompson, Bioemendal, and Dearing 1980). The χ_{lf} values of the soils ranged from $47.05 \times 10^{-8}\, m^3kg^{-1}$ to $199.26 \times 10^{-8}\, m^3kg^{-1}$, with median value $88.43 \times 10^{-8}\, m^3kg^{-1}$(Table 1). Similar values of χ_{lf} between $47 \times 10^{-8}\, m^3kg^{-1}$ to $141 \times 10^{-8}\, m^3kg^{-1}$, with median value $93 \times 10^{-8}\, m^3kg^{-1}$ were previously reported by Cao et al. (2015) in the Linfen agricultural area. It is possible that atmospheric fall-out released from automobile exhaust and coal burning might have contributed to the minor increase of median value compared to our samples. Higher values of χ ($29–146 \times 10^{-8}\, m^3kg^{-1}$) have been reported for the mixed agricultural and industrial area in south France (Lecoanet, Leveque, and Ambrosl 2003), which is likely due to differences in the soil derived from different parent materials.

The χ_{fd} can be used to describe ultrafine magnetic grains at the super paramagnetic (SP) and stable single domain (SD) boundary particles (Dearing et al. 1996). The average value of χ_{fd} was larger than 6% in all samples (Table 1), which is within a similar range to values determined in the agricultural soil of Linfen by Cao et al. (2015). The value reported here indicates the presence of a significant proportion of fine super paramagnetic (SP) grains rather than coarser grained particles, which tend to lead to lower values of χ_{fd}.

Heavy metal content and distribution in soil

The heavy metal content of the soils was determined for Cu (24.07- 42.22 $mgkg^{-1}$), Zn (66.29–194 $mgkg^{-1}$), Ni (35.75–53.76 $mgkg^{-1}$), Pb (19.84–79.51 $mgkg^{-1}$), Cr (67.01–101.1 $mgkg^{-1}$),

Table 1. Descriptive statistics of magnetic parameters and metal concentrations of topsoil samples.

	χ_{lf} $10^{-8}\,m^3kg^{-1}$	χ_{fd} %	Cu $mgkg^{-1}$	Zn $mgkg^{-1}$	Ni $mgkg^{-1}$	Pb $mgkg^{-1}$	Cr $mgkg^{-1}$	Cd $mgkg^{-1}$	PLI
Mean	93.01	6.04	29.67	98.08	42.86	29.20	77.19	0.17	1.52
Median	88.43	6.14	29.35	95.59	42.31	28.56	76.88	0.18	1.49
Min	47.05	1.04	24.07	66.29	35.75	19.84	67.01	0.09	1.27
Max	199.26	10.38	42.22	194.00	53.76	79.51	101.10	0.47	2.18
CV	0.32	0.35	0.11	0.20	0.09	0.28	0.08	0.35	0.11
Std. dev.	29.61	2.11	3.35	19.97	3.77	8.26	6.05	0.06	0.16
Skewness	1.70	−0.25	1.15	1.95	0.65	4.29	0.89	2.19	1.67
Kurtosis	3.89	−0.15	2.42	6.98	0.54	23.12	2.30	9.67	4.34
BG	–	–	20.70	68.00	24.90	23.50	57.30	0.079	–
CF	–	–	1.16–2.04	0.97–2.85	1.44–2.16	0.84–3.38	1.17–1.76	1.13–5.95	–

CV: coefficient variation; BG: Background values China National Environmental Monitoring Center (CNEMC) (1990); CF: Concentration factor; PLI: Pollution Load Index

and Cd (0.09–0.47 $mgkg^{-1}$) (Table 1). The enrichment of heavy metals in the topsoil suggests possible anthropogenic sources. These anthropogenic sources are likely to include the agricultural use of pesticides (12.4 $kghm^{-2}$) and fertilizers (527.8 $kghm^{-2}$) as well as other industrial sources (Yang et al. 2015). The median contents of Cu, Zn, Ni, Pb, Cr and Cd were determined to be 29.35, 95.59, 42.31, 28.56, 76.88, and 0.18 $mgkg^{-1}$ respectively, which were higher than their corresponding background values. This clearly indicates that anthropogenic activities were the main factor contributing to soil pollution in this region. The coefficients of variation (CV) ranged from 0.09 to 0.35 indicating minor variability. The concentrations of Zn, Ni, and Cd were all slightly higher than those that have been previously reported in the other agricultural soils excluding Linfen (Liao et al. 2007; Lu et al. 2012; Liu et al. 2015; Cao et al. 2015), while Cu was lower than in the other agricultural soils (Table 2), indicating soil contamination of heavy metal as a result of excessive fertilization and pesticide application.

The ordinary kriging interpolation method for heavy metal was used for the spatial distribution maps. The results showed that the concentrations of Cu, Zn and Pb had a similar spatial distribution, confirming the generally observed correlation among these variables (Figure 2a, b, and d), and that they tended to accumulate in greater abundance in the northeastern part of the study area, which has undergone rapid urbanization and industrial development. Approximately 44% of Cu samples, 30% of Zn samples, 25% of Ni samples, 16% of Pb samples, 9% of Cr samples, and 6% of Cd samples surpassed their corresponding average values.

As for Ni and Cr, abnormal values were observed in the eastern and western region (Figure 2c, e), which might indicate potential sources within industrial hot spots. The distribution of Cd concentration showed the highest values in the urbanized areas (Figure 2f), indicating that the abnormal values are due to urbanized rather than the agricultural activities.

Correlation between heavy metal concentrations and magnetic susceptibility

The correlations between low frequency magnetic susceptibility (χ_{lf}) against Cu (0.278) and PLI (0.249) were weak which is attributed to the fact that samples were collected from an agricultural area rather than industrial or urban one (Table 3). The results disagree with those reported from other industrial and urban areas. Previous studies have reported χ_{lf} to be significantly correlated to integrated pollution index, Fe, Cu, and Zn (Lu et al. 2008; Morton-Bermea et al. 2009). The lack of a relationship between χ_{lf} and Zn in this case can be attributed to arable soils. Furthermore, Cu showed a significant positive relationship with Zn (0.687), Ni (0.633), and Pb (0.673), which may reflect the fact that Cu, Zn, Ni, and Pb share similar sources. Cu exhibited no correlation with Cr and Cd. The Cr showed a weakly positive correlation with Cd (0.239); however, Cr showed negative correlations with Zn (−0.196) and Pb (−0.059) (Table 4).

Table 2. Average values of magnetic susceptibility and heavy metal concentrations in the agricultural soils.

	χ_{lf} 10^{-8} m^3kg^{-1}	χ_{fd} %	Cu $mgkg^{-1}$	Zn $mgkg^{-1}$	Ni $mgkg^{-1}$	Pb $mgkg^{-1}$	Cr $mgkg^{-1}$	Cd $mgkg^{-1}$	Reference
Taiyuan	–	–	32.11	90.76	29.74	27.87	74.10	0.25	Liu et al. (2015)
Beijing	–	–	22.40	69.80	–	20.40	–	0.14	Lu et al. (2012)
Nanjing	–	–	31.20	78.90	32.70	30.30	76.20	0.17	Liao et al. (2007)
Linfen	93.00	3.70	26.00	68.00	63.00	25.00	107.00	–	Cao et al. (2015)
Linfen	93.01	6.04	29.67	98.08	42.86	29.20	77.19	0.17	Present study

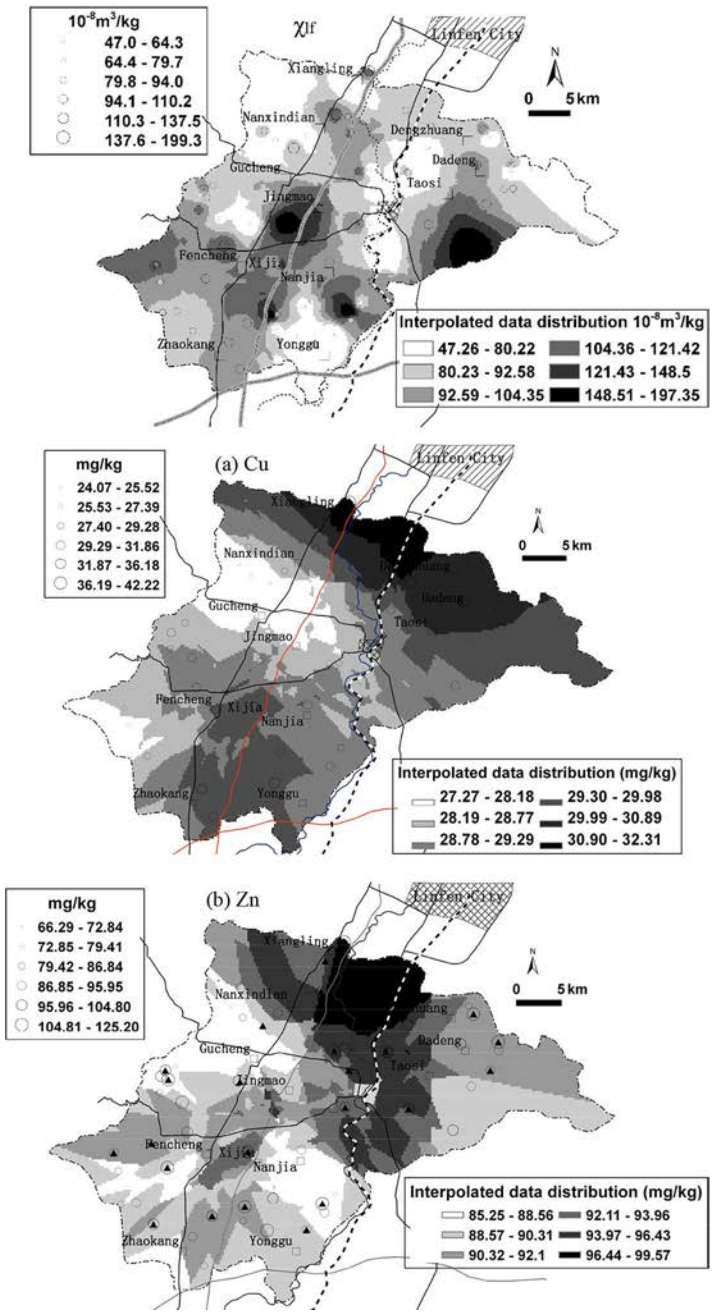
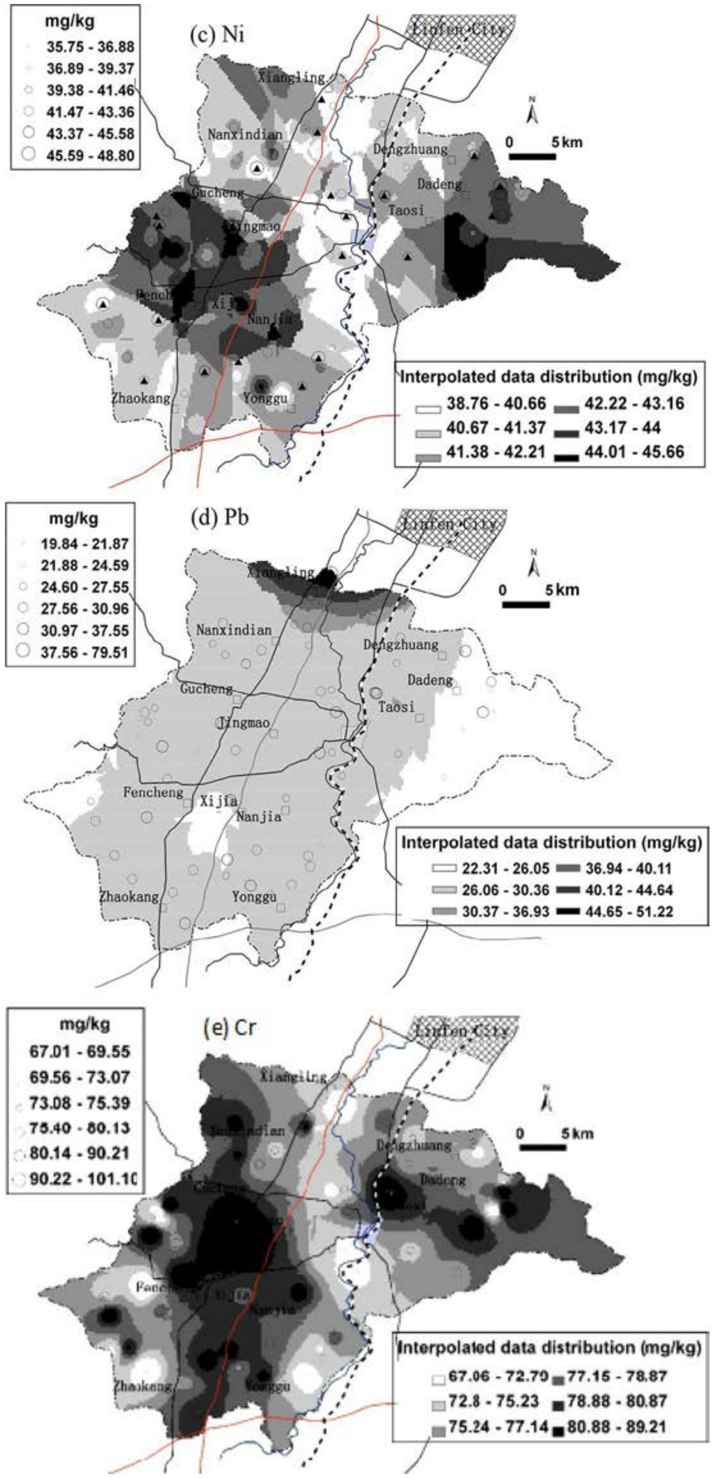


Figure 2. Spatial distribution of heavy metals in topsoil.

Identification of the sources of heavy metal pollution

Principal component analysis (PCA) was used to investigate the complex multivariate relationships among variables and to identify different groups of chemical elements with approximately the same geochemical pattern (Facchinelli, Sacchi, and Mallen 2001;



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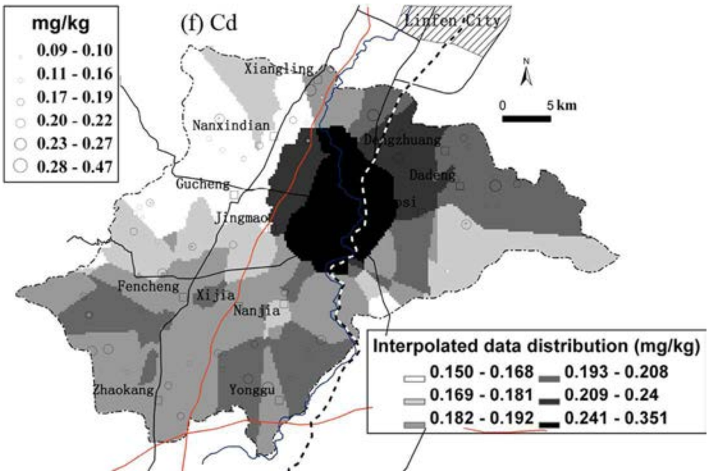

Figure 2. Continued.

Table 3. Pearson correlation coefficients among the variables.

	χ_{lf} 10^{-8} m^3kg^{-1}	χ_{fd} %	Cu $mgkg^{-1}$	Zn $mgkg^{-1}$	Ni $mgkg^{-1}$	Pb $mgkg^{-1}$	Cr $mgkg^{-1}$	Cd $mgkg^{-1}$
χ_{d}	0.017							
Cu	0.278*	−0.137						
Zn	0.141	−0.051	0.687**					
Ni	0.021	0.010	0.633**	0.639**				
Pb	0.225	−0.302*	0.673**	0.629**	0.284*			
Cr	−0.025	−0.263*	0.044	−0.196	0.186	−0.059		
Cd	0.145	−0.188	0.033	0.098	−0.204	0.050	0.239*	
PLI	0.249*	−0.291*	0.756**	0.776**	0.497**	0.733**	0.222	0.514**

*$p < 0.05$.
**$p < 0.01$.

Cicchella et al. 2014). If the concentrations of Cu, Zn, Ni, and Pb are significantly correlated, it suggests that these heavy metals have common sources which might depend on agricultural practices. According to the results of the initial eigenvalues, four principal components with eigenvalue >1, explained 82.12% of the total variance (Table 4, Figure 3). The first principal component (PC1), accounting for 31.21% of the total variance, was

Table 4. Total variance and component matrixes for soil properties.

	Rotation sums of squared loadings			
	PC1	PC2	PC3	PC4
χ_{lf}	0.109	0.989	0.025	−0.015
χ_{fd}	−0.172	0.104	−0.769	−0.119
Cu	0.878	0.204	0.048	0.103
Zn	0.890	0.047	0.020	−0.145
Ni	0.772	−0.060	−0.339	0.427
Pb	0.764	0.133	0.350	−0.229
Cr	−0.050	−0.008	0.271	0.923
Cd	−0.078	0.188	0.025	0.150
Eigenvalue	2.809	2.052	1.384	1.146
% Total Variance	31.212	22.802	15.378	12.728
Cumulative % Variance	31.212	54.014	69.392	82.120

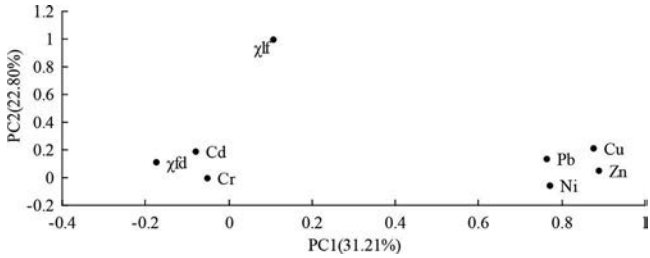

Figure 3. Loading plots of the first two PCs obtained for the data.

correlated (loading > 0.75) with Cu, Zn, Ni, and Pb, which can be primarily ascribed to anthropogenic influences such as intense agricultural activity. The χ_{lf} was mainly correlated with the second component (PC2), which explained 22.80% of the total variance. The third (PC3) and the fourth (PC4) principal components, accounting for the 15.38% and 12.73% of total variance, were correlated with χ_{fd}, Cd, and Cr, respectively (Table 4), which can be explained by industrial processes and urbanization activities.

Assessment of soil pollution

CF values ranged from 1.16 to 2.04 for Cu, from 0.97 to 2.85 for Zn, from 1.44 to 2.16 for Ni, from 0.84 to 3.38 for Pb, from 1.17 to 1.76 for Cr, and from 1.13 to 5.95 for Cd (Table 1). CF > 1 indicates that the soil pollution is anthropogenic in origin. The highest values of CF for Cd (5.95) and Pb (3.38) indicated topsoil contamination of Cd and Pb, whereas CF values from 1 to 3 revealed pollution signal with Cu, Zn, Ni, and Cr.

PLI values ranged from 1.27 to 2.18 with an average of 1.52 (Table 1). This indicates that there was a significant level of pollution in the study area as anticipated from other measurements.

Conclusion

This study has demonstrated the intense impact of human activities in the arable soil of the Linfen basin. The correlation coefficient and principal component analyses indicate a common source for Cu, Zn, Ni, and Pb, which is likely due to the use of agrochemicals in agriculture combined with atmospheric deposition. Enrichment factors and Pollution load index values larger than 1 indicated significant heavy metal contamination in the soils. This has significant implications for the potential danger that the soil might have towards the health of plant, animal, and human life.

The correlation between magnetic susceptibility and heavy metal concentration was also probed within this study and it is clear that it is complex within areas with multiple pollution sources. Nevertheless, magnetic susceptibility has a positive correlation with Cu and PLI that indicates that magnetic susceptibility could potentially be used as a parameter to measure heavy metal concentrations of soil, especially in areas considered to be exposed to anthropogenic sources of heavy metals. Understanding the types and sources of the heavy metal pollution is essential for decision-making policy in terms of sustainable future land use on. Through our investigation, we have shown that it is possible to correlate the use of pesticides and fertilizers with basic magnetic methods with minimal detrimental impact on the environment.

Acknowledgment

There is no conflict of interest regarding the paper. The authors are grateful to the editor, anonymous reviewers for their useful suggestions, Professor Dr. Renzhao Mao and Professor Dr. Erwin Appel for constructive discussions.

Funding

This work was supported by the National Natural Science Foundation of China (No. 31272258).

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