



## Spatial characteristics of cadmium in topsoils in a typical e-waste recycling area in southeast China and its potential threat to shallow groundwater



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### HIGHLIGHTS

- We characterize the Cd<sub>total</sub> in topsoils, pH and SOM in a typical e-waste recycling area.
- The relationships between Cd<sub>total</sub> in topsoils, pH, and SOM were studied.
- Impact of topsoil Cd accumulation and acidification on shallow groundwater quality was evaluated.

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### ABSTRACT

Informal electrical and electronic waste (e-waste) recycling often creates secondary sources of cadmium (Cd) pollution. To characterize the total Cd concentration (Cd<sub>total</sub>) in topsoil and evaluate the threat of Cd in topsoils to shallow groundwater, 187 topsoil samples and 12 shallow groundwater samples were collected in a typical e-waste recycling area in southeast China. Soil organic matter content, soil pH and Cd<sub>total</sub> in topsoil, pH and dissolved Cd concentration in shallow groundwater were measured. Cd<sub>total</sub> in the topsoils showed an inverse distribution trend with soil pH in that high Cd concentrations (and low pH values) were found in the surrounding area of the metal recycling industrial park where there were many family-operated e-waste recycling facilities before the industrial park was established and with low concentrations (and high pH values) in other areas, and they had similar spatial correlation structures. Cd accumulation and acidification were synchronous in topsoils, and soil pH was significantly correlated with Cd<sub>total</sub> in topsoils with low to moderate negative correlation coefficient ( $r = -0.24$ ), indicating that both of them maybe correlated with informal recycling. The shallow groundwater in the surrounding area of the metal recycling industrial park was seriously contaminated by Cd, and topsoil Cd accumulation and acidification in the surrounding area of e-waste recycling sites significantly increase the risk of shallow groundwater contaminated by Cd. Action is urgently required to control Cd accumulation and acidification by improving the recycling operations of e-wastes in order to reduce the risk of Cd leaching from topsoils and shallow groundwater contamination.

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**Abbreviations:** E-wastes, electrical and electronic wastes; HM, heavy metal; Cd, cadmium; Cd<sub>total</sub>, total Cd concentration; GPS, global positioning system; SOM, soil organic matter content; ICP-MS, inductively coupled plasma mass spectrometry; HNO<sub>3</sub>, nitric acid; HClO<sub>4</sub>, perchloric acid; Cd<sub>dissolved</sub>, dissolved Cd concentration; RSD, relative standard deviation; ln, natural logarithmic; BV, background value; GLV, guideline value; MOH, Ministry of Health of China; NSMC, National Standardization Management Committee; SEPAC, State Environmental Protection Administration of China.

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### 1. Introduction

With the rapid development of information technology and the accelerated update and upgrade of electronic products, the elimination and recycling of electrical and electronic wastes (e-wastes) become a problem of the world currently. E-wastes not only contain recyclable resources, such as iron, aluminum, copper and precious metals, but also contain persistent organic pollutants (e.g. polycyclic aromatic hydrocarbons, polychlorinated biphenyls, polybrominated diphenyl ethers) and heavy metals (e.g. cadmium, lead, zinc, mercury). The potential economical benefit is promoting the development of e-waste disassembly industry, particularly for developing countries including China (Williams et al., 2008; Liu et al., 2009; Ni and Zeng, 2009). In China, a

large proportion of e-waste is transported to family-operated recycling facilities. In these recycling facilities, strong acid leaching and the open burning of dismantled components are used often to extract precious metals contained in e-wastes. These informal dismantling, recycling, and disposal practices caused severe heavy metal (HM) pollution in air, dust, soil, river water and sediment in e-waste recycling area (Deng et al., 2006; Ha et al., 2009; Zhang and Min, 2009). Informal recycling is currently the prevalent e-waste recycling practice in China, especially in some coastal regions (Liu et al., 2006; Terazono et al., 2006; Yang, 2008).

In southeast China, e-waste processing sites are usually located in fields adjacent to land used for agricultural purposes (Tang et al., 2010; Luo et al., 2011). The informal recycling process may cause serious agricultural soil contamination with HMs in the vicinity of e-waste processing sites (Wong et al., 2007; Shen et al., 2008; Zhang and Min, 2009). Agricultural soil contamination with HMs had become a serious environmental problem because it poses a serious threat to human health by entering into food chains and to environmental security by washing into surface water by rain and leaching into shallow groundwater (Shen and Chen, 2000; Romić and Romić, 2003). Cadmium (Cd) is one of the toxic HMs that was used widely in the electrical and electronic industry, and they were most widely detected in e-waste contaminated surroundings (Shen et al., 2008; Guo et al., 2009; Wang et al., 2009; Chen et al., 2010). Therefore, it is necessary to quantify the spatial characteristics of HM in agricultural soils and evaluate the threat of toxic HMs in topsoils to shallow groundwater by leaching in order to be able to formulate a policy to control or eliminate Cd pollution and ensure shallow groundwater safety.

There is a large body of literature on HM contamination in soils in the vicinity of e-waste processing site (e.g., Ha et al., 2009; Tang et al., 2010; Luo et al., 2011). However, few studies have been conducted on the threat of Cd in topsoils to shallow groundwater in the vicinity of e-waste processing sites. In this study, a typical e-waste recycling area with many family-operated recycling facilities in southeastern China was chosen as our study area. The main objectives of this study

were: (1) to characterize the total Cd concentration ( $Cd_{total}$ ) in topsoil in a typical e-waste recycling area; (2) to study the relationship between  $Cd_{total}$  in topsoils and pH, SOM; and (3) to evaluate the threat of Cd in topsoils to shallow groundwater.

## 2. Materials and methods

### 2.1. Descriptions of study area

The study area is part of a town in Taizhou city, Zhejiang Province, Southeast China with a total area of approximately 10 km<sup>2</sup>. In this town, there are many family recycling facilities to process a large quantity of e-wastes from domestic supplies and developed countries from the late of 1980s. A metal recycling industrial park with a total area of 107 ha has been established in southeastern of the study area with the purpose of promoting efficient and environmentally-friendly recovery of original and imported metal scraps (Fig. 1), and most of e-waste recycling activities were carried on in the metal recycling industrial park in recent years. The study area has become an important e-waste recycling area in southeastern China.

The study area is in the northern subtropical zone of monsoonal climate with a temperate and humid climate throughout the year with four distinct seasons. The average annual temperature is 17.1 °C and the mean annual precipitation is approximately 1523 mm. Paddy fields and uplands are two major land use types of arable land in the study area. Gleysols is the major type of anthropogenic soils in the study area. According to hydrogeological data, the thickness of the porous unconfined aquifer is approximately 2–3 m, and the shallow groundwater level is approximately 0.5–2.5 m being deeper in the west and more shallow in the eastern region of the study area. The permeability of the unconfined aquifer was uneven and very low. The vertical flow is the major movement of groundwater and the velocity of horizontal flow was very low due to the small hydraulic gradient in the study area.

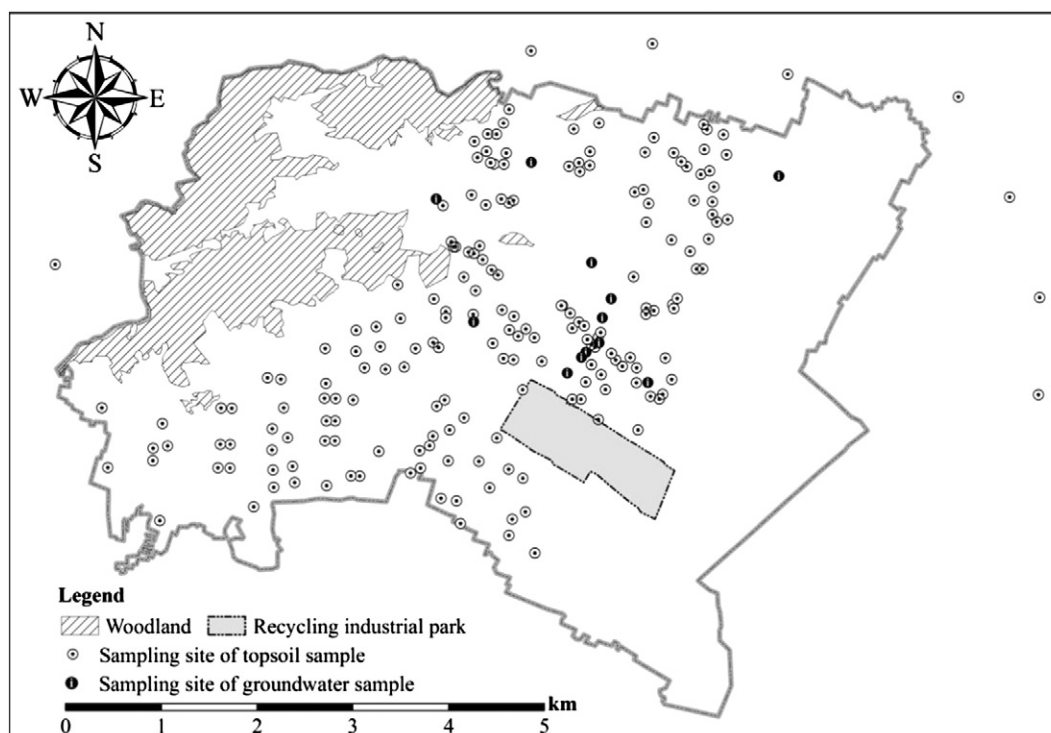


Fig. 1. The location of study area and distribution of sampling sites.

## 2.2. Sampling design and analysis

To characterize  $Cd_{total}$  in agricultural soils, 187 topsoil samples (0–15 cm) were collected from agricultural lands in the study area on approximate grid sampling methodology with consideration of the locations of pollution source and agricultural land use type during September 2007 (Fig. 1). To know the rough background value of Cd, 8 reference topsoil samples were collected from agricultural lands in the surrounding areas on random sampling methodology with consideration of potential pollution source and agricultural land use type. Individual samples were collected from 6 to 8 points at each site of area 0.1–0.2 ha, mixed thoroughly to give a composite sample, then divided into ~1–2 kg portion, and one portion was taken to the laboratory for analysis. All sampling sites were geo-referenced using a handheld global positioning system (GPS) instrument. Soil samples were air-dried, sieved at a diameter of 2 mm, and then analyzed in laboratory. Soil organic matter content (SOM) was measured by dry ash method and soil pH was measured by pH meter (Sartorius Basic pH meter PB-10) with a soil:water ratio of 1:2.5. The  $Cd_{total}$  in agricultural soils was measured by inductively coupled plasma mass spectrometry (ICP-MS) after soil samples had been digested with a mixture of nitric acid ( $HNO_3$ ) and perchloric acid ( $HClO_4$ ). The validity of the whole analyzing procedure was checked using the certified reference materials GBW07404 (GSS-4). Analyses of GSS-4, replicate samples and blanks were performed after every ten samples and were carried through the entire soil sample preparation and analytical process.

The shallow nature of some unconfined aquifers makes them highly vulnerable to superficial events such as rain, irrigation, and dumping of toxic substances. To evaluate the impact of topsoil Cd accumulation and soil acidification on shallow groundwater quality, nine groundwater samples were collected from nine private tube-wells in the surrounding area of the metal recycling industrial park and another three reference groundwater samples were collected from three private tube-wells which were in the northern part of the study area and far away from the metal recycling industrial park (>1.8 km) in April 2009. During the period of groundwater sampling, groundwater levels of twelve tube-wells were approximately 1–1.5 m. Since the groundwater depth of all twelve tube-wells was approximately 10 m, groundwater was considered to be from the shallow aquifer. Groundwater samples were collected from tube-wells following this sequence: (1) pump the tube-well for several minutes; (2) wash out a clean polyethylene bottle with the well water; (3) collect groundwater from 5 cm below the groundwater table using a polyvinyl chloride (PVC) Baylor tube and filter the water through a 0.45  $\mu m$  glass-fiber filter (Whatman, Clifton, NJ, USA) to remove sand and debris during field sampling; (4) fill a 250 ml polyethylene bottle with filtered groundwater and acidify with ultra-pure nitric acid to  $pH < 2$  to inhibit adsorption of the bottle wall and biological activity; and (5) store the sample at 4 °C. The pH of groundwater was measured on-site using a portable pH meter, and the dissolved Cd concentration ( $Cd_{dissolved}$ ) in groundwater was measured by ICP-MS. The percentage relative standard deviation (RSD) values determined from repeated analyses of standards and duplicate or triplicate samples were less than 5%.

## 2.3. Data transformation

In multivariate statistics and linear geostatistics, the variables under study should be normally distributed (Webster and Oliver, 2001; Gallego et al., 2002). To avoid distortion of the results and low levels of significance, data transformations are generally performed on all measured values. Of numerous data transformation methods, natural logarithmic (ln) transformation is widely applied (Webster and Oliver, 2001). In our study, ln-transformation was used to transform the data to increase normality and reduce skewness.

## 2.4. Geostatistical analysis

Geostatistical methods can be used with unbiased prediction and minimum variance for soil properties of interest (Stein and Corsten, 1991; Sun et al., 2012). Kriging is a commonly used geostatistical prediction method. The semivariogram, the main component of kriging, is an effective tool for evaluating spatial variability and structure (Boyer et al., 1991; Cahn et al., 1994; Guo et al., 2001; Iqbal et al., 2005). The variogram provides a clear description of the spatial structure of variables and some insight into possible processes affecting variable distribution (Webster and Oliver, 1990; Paz González et al., 2001).

In this study, anisotropy of variogram was not found for all data. All semivariograms in isotropic form were fit using a spherical model, exponential model, Gaussian model or linear model, and the best-fit model was applied to kriging interpolation. Ordinary kriging was chosen to create the spatial distribution maps of  $Cd_{total}$  after data transformation, soil pH and SOM, using the nearest 16 sampling points and a maximum search distance equal to the range distance of the variable. The resolution of the interpolated grid was 20 m. More technical descriptions of kriging and the semivariogram are available in the literature (see Webster and Oliver, 2001). Software version 7.0 of GS+ Geostatistics for the Environmental Sciences (Gamma Design Software, Plainwell, MI) was used to perform all the geostatistical computations.

## 2.5. Correlation analysis

Correlation analysis was applied to identify the relationships between  $Cd_{total}$  and related soil properties (pH and SOM) in the study area. The Spearman nonparametric correlation coefficient was used for the most positively skewed variables such as  $Cd_{total}$ . Correlation analysis was performed using SPSS for Windows version 13.

## 3. Results and discussion

### 3.1. Descriptive statistics and status of Cd accumulation in topsoils

A descriptive summary of  $Cd_{total}$  in topsoil of the study area, and selected soil properties (pH and SOM) are listed in Table 1. The results show that  $Cd_{total}$  in topsoil of the study area had a wide range (48–11,389  $\mu g kg^{-1}$ ) coupled with high coefficient of variation (198%), suggesting that extrinsic factors affect  $Cd_{total}$  of agricultural soils in the study area. According to the background value (BV) at Zhejiang Province scale (Zhejiang Soil Survey Office, 1994; Cheng et al., 2006), the BV of Cd is 170  $\mu g kg^{-1}$ , and the  $Cd_{total}$  in the 8 reference topsoil samples had a small range (92–176  $\mu g kg^{-1}$ ) with a mean of 126  $\mu g kg^{-1}$ . There were 124 topsoil samples (66.3%) for Cd exceeding the local BV and 173 topsoil samples (92.5%) exceeding the mean concentration of  $Cd_{total}$  in the 8 reference topsoil samples, indicating that Cd enrichment existed in most of the area. According to the Chinese

**Table 1**

Summary statistics for total cadmium concentrations ( $Cd_{total}$ ,  $\mu g kg^{-1}$ ), soil pH and soil organic matter content (SOM,  $g kg^{-1}$ ) in topsoils of the study area and pH and dissolved cadmium concentrations ( $Cd_{dissolved}$ ,  $\mu g l^{-1}$ ) in shallow groundwater.<sup>a</sup>

		Min	Max	Mean	CV (%)	Skew
Topsoil (n = 187)	$Cd_{total}$	48	11,389	1255.5	198	2.57
	pH	3.94	6.01	4.95	8.1	-0.31
	SOM	16.1	79.4	49.4	20.2	0.12
Groundwater <sup>I</sup> (n = 9)	$Cd_{dissolved}$	9.0	39.1	17.1	53.8	1.92
	pH	7.0	7.4	7.2	2.2	0.43
Groundwater <sup>II</sup> (n = 3)	$Cd_{dissolved}$	0.6	1.0	0.8	27.2	1.29
	pH	6.9	7.3	7.1	29.2	-1.29

<sup>a</sup> Min, minimum; Max, maximum; CV, coefficient of variation; Skew, skewness; Groundwater<sup>I</sup>, groundwater samples were collected from nine private tube-wells in the neighbor of the recycling industrial park; Groundwater<sup>II</sup>, groundwater samples were collected from 3 private tube-wells far away the recycling industrial park.

Environmental Quality Standard for Soils (GB 15618-1995) (SEPA, 1995), the guideline value (GLV) of Cd under acidic soil ( $\text{pH} < 6.5$ ) is  $300 \mu\text{g kg}^{-1}$ . There were 70 topsoil samples (37.4%) exceeding the GLV of Cd, indicating that topsoil Cd contamination existed in partial area. The SOM was in the range of  $16.1\text{--}79.4 \text{ g kg}^{-1}$  with a mean of  $49.4 \text{ g kg}^{-1}$  and the soil pH in the study area was in the range of  $3.94\text{--}6.01$  with a mean of 4.95, indicating that topsoil acidification in this area was very serious. The soil pH in the 8 reference topsoil samples was in the range  $4.74\text{--}6.69$  with a mean of 5.82. The low topsoil pH in the study area and the surrounding areas may be partly correlated with acid rain. According to the meteorologic data of 1999–2008 from the local observatory, the rate of acid rain increased from 47.5% to 85.3% and the mean value of pH decreased from 4.99 to 4.48 (Zhou et al., 2011).

### 3.2. Spatial characteristics of Cd, pH and SOM in topsoils

Soil HMs and soil properties in environmental science have spatial structures, including spatial autocorrelation. In this study, the geostatistics method was used to spatial structure analysis. The fitted models and parameters of semivariogram for  $\ln\text{Cd}_{\text{total}}$ , pH and SOM were summarized in Table 2. Semivariograms showed that  $\ln\text{Cd}$  and pH were all fitted for spherical model, whereas SOM was best fitted for an exponential model. Range value is a measure of extension where autocorrelation exists (Webster and Oliver, 1990; Li et al., 2007). The effective range of Cd was very close to that of pH, suggesting that they had similar spatial correlation structures and their distribution was affected by similar influence factors. Whereas, SOM had shorter effective range than pH, indicating that SOM has worse spatial structure and more variation caused by extrinsic factors.

In order to know distribution patterns of pH, SOM, and  $\text{Cd}_{\text{total}}$  in agricultural soils, kriging interpolation was used to obtain the filled contour maps (Fig. 2). According to spatial maps,  $\text{Cd}_{\text{total}}$  in topsoil had an obvious distribution trend with high concentration in the surrounding area of the metal recycling industrial park where there were many family-operated e-waste recycling facilities before the park was established and with low concentration in other areas. High concentration of  $\text{Cd}_{\text{total}}$  in the surrounding area of the metal recycling industrial park maybe correlated with the previous and recent informal recycling operations of e-wastes. The pH of agricultural soils had an almost inverse distribution trend with  $\text{Cd}_{\text{total}}$  in agricultural soils, indicating that soil acidification and Cd accumulation were synchronous, and both of them maybe correlated with informal recycling operations of e-wastes. This indicates the low pH of the topsoil in the study area partly correlated with informal e-waste recycling such as acid baths that are used to dissolve lead, silver, and other metals contained in the e-wastes. The spatial maps of SOM showed local spatial variability and its distribution pattern seemed more irregular than pH and  $\text{Cd}_{\text{total}}$ , suggesting that SOM expressed different spatial variability with pH and  $\text{Cd}_{\text{total}}$  due to varying influence factors.

### 3.3. Correlations between $\text{Cd}_{\text{total}}$ in topsoils and related soil properties

Previous studies indicated that SOM and pH are the most important parameters controlling the accumulation of HMs in soil environment (Msaky and Calvet, 1990; Nyamangara and Mzezewa, 1999; Covelo et al., 2007; Kashem et al., 2007). To obtain the valuable information,

**Table 2**

The isotropic semivariogram model parameters of natural logarithmic transformed total Cd concentrations ( $\ln\text{Cd}_{\text{total}}$ ), pH and soil organic matter content (SOM).

Variable	Model	Nugget/sill (%)	Effective range (m)	$r^2$
$\ln\text{Cd}_{\text{total}}$	Spherical	11.5	1789	0.927
pH	Spherical	49.7	1818	0.847
SOM	Exponential	8.9	366	0.632

the Spearman nonparametric correlation coefficients were calculated between  $\text{Cd}_{\text{total}}$  in topsoils and related soil properties (pH and SOM), and the results were shown in Table 3.  $\text{Cd}_{\text{total}}$  in topsoils was poorly correlated with SOM, and significantly correlated with soil pH at the 0.01 level with low to moderate negative correlation coefficient ( $r = -0.24$ ). It indicated that the influence of SOM on Cd accumulation in the study area was relatively limited, and the influence of soil pH on Cd accumulation was not ignorable.

### 3.4. Impact of Cd in topsoils and pH on shallow groundwater quality

A descriptive summary of  $\text{Cd}_{\text{dissolved}}$  in the shallow groundwater and pH of groundwater was also listed in Table 1. The results showed that  $\text{Cd}_{\text{dissolved}}$  of all nine shallow groundwater samples that were collected from wells in the surrounding area of the metal recycling industrial park was significantly higher than that of three groundwater samples that were collected from well far away the metal recycling industrial park. The guideline value (GLV) of Cd is  $5 \mu\text{g l}^{-1}$  according to the Chinese standard for drinking water (GB5749-2006) (MOH and NSMC, 2006), nine shallow groundwater samples (100%) collected from wells in the surrounding area of the metal recycling industrial park and no shallow groundwater sample (0%) collected from wells that were far away the metal recycling industrial park exceeded the GLV. This shows that the groundwater in the surrounding area of the metal recycling industrial park was seriously contaminated by Cd. The pH of all shallow groundwater was in the range of 6.9–7.4, indicating that the shallow groundwater was neutral and the impact of soil acidification on the pH of shallow groundwater was limited.

From Figs. 1 and 2, we can conclude that the topsoil in the surrounding area of the metal recycling industrial park was also highly contaminated by Cd and acid, whereas the topsoils in the surrounding area of the three groundwater sample sites far away the metal recycling industrial park were not polluted by Cd. This indicates that there were close correlations between shallow groundwater contamination with Cd and topsoil Cd contamination and soil acidification. A previous study showed that a large proportion (>50%) of Cd in the topsoil in surrounding area of e-waste recycling sites was in the form of exchangeable Cd (Wang et al., 2006). It is well known that the exchangeable Cd has high mobility, easy to entering to the shallow groundwater. Action is urgently required to control the Cd accumulation and acidification in order to reduce the risk of shallow groundwater contamination with Cd.

## 4. Conclusions

In the study area, Cd was enriching in majority area, and serious soil acidification existed in whole study area. The  $\text{Cd}_{\text{total}}$  in agricultural soils of the study area had an inverse distribution trend with soil pH in that high Cd concentration (low pH value) was found in the surrounding area of the metal recycling industrial park where there were many family-operated e-waste recycling facilities before the industrial park was established and with low concentration (high pH value) in other areas. Soil pH was significantly correlated with  $\text{Cd}_{\text{total}}$  in topsoils with low to moderate negative correlation coefficient, and the influence of soil pH on Cd accumulation was not ignorable. The concentration of  $\text{Cd}_{\text{dissolved}}$  in the shallow groundwater in the surrounding area of e-waste recycling sites was significantly higher than that of in the other area; topsoil Cd accumulation and acidification in the surrounding area of e-waste recycling sites significantly increase the risk of shallow groundwater contaminated by Cd.

## Conflict of interest

We are the authors of the manuscript entitled “Spatial characteristics of cadmium in topsoils in a typical e-waste recycling area in southeast China and its potential threat to shallow groundwater”, and we declare that no conflict of interest existed among us.

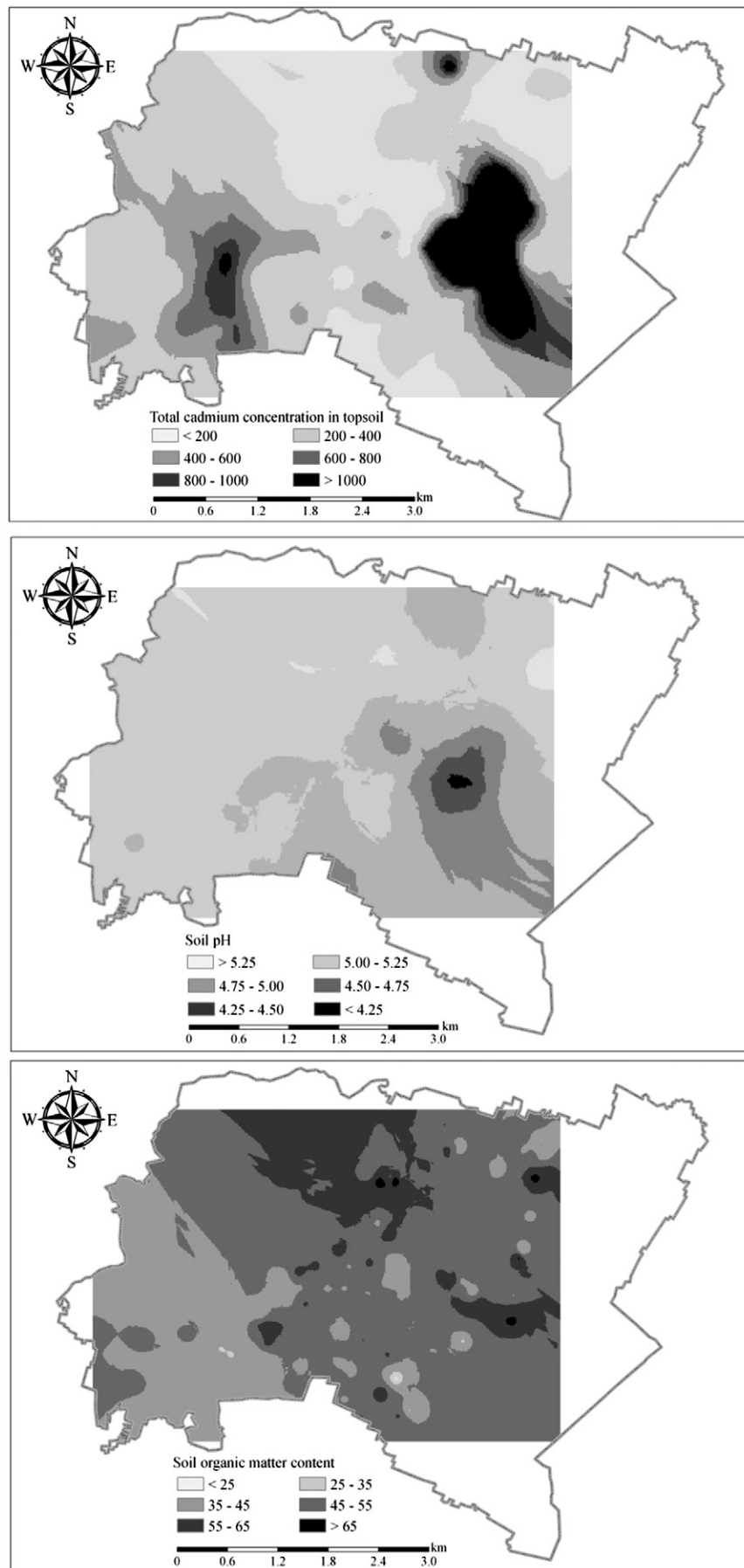


Fig. 2. The prediction map of total cadmium concentration ( $\mu\text{g kg}^{-1}$ ), soil pH and soil organic matter content ( $\text{g kg}^{-1}$ ) in topsoils of the study area by ordinary kriging.

**Table 3**

Nonparametric correlations among soil pH, soil organic matter content (SOM), and total cadmium concentration ( $Cd_{total}$ ) in topsoils ( $n = 187$ ), respectively.

	$Cd_{total}$	pH	SOM
$Cd_{total}$	1	−0.24**	0.12
pH	−0.24**	1	−0.14
SOM	0.12	−0.14	1

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

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