Assessment of heavy metals contamination of paddy soil in Xiangyin county, China

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Abstract
A field survey was conducted to investigate the heavy metal contamination of paddy soils in Xiangyin County, China. The total concentration of Cd, Pb, Zn, Cu, Cr and Ni in paddy soil was measured, and the environmental quality of paddy soil was assessed using pollution index methods. Paddy soils were slightly polluted with the content of Cd exceeding the standard value. The moderately polluted soils were mainly distributed in the land-reclamation area from Dongting Lake, and slight pollution was found in the eastern hilly area.

Key Words
Paddy soil, heavy metal contamination, Xiangyin county.

Introduction
In China, heavy metal contamination in soils has attracted serious attention in recent years, which poses a considerable hazard to health (Cheng 2003; Zhao 2004). After long-term application of untreated wastewaters, significant amounts of heavy metals can accumulate in the soil at toxic levels. At present, heavy metals, such as Cr, Zn, Pb, Cd, Ni, etc., are commonly found in subsurface soil irrigated with wastewater. Once the adsorption sites of the soil for heavy metals is saturated, more heavy metals would be distributed in the aqueous phase and the bioavailability of heavy metals would subsequently be enhanced (Sridhara *et al.* 2008). The accumulation of heavy metals in agricultural soils has been a wide concern of the public as well as governmental agencies, due to the food safety issues and potential health risks as well as its detrimental effects on soil ecosystems (McLaughlin *et al.* 1999; Yanez *et al.* 2002). Combined pollution with heavy metals has frequently been reported in many contaminated sites in China, such as in Wenzhou, Zhejiang Province (Jin *et al.* 2002). As a very toxic element, Cd is of primary concern in soil and food contamination, particularly in the rice cropping system (Reeves and Chaney 2001). These potentially toxic elements accumulate in soils and induce a potential contamination of food chain and endanger the ecosystem safety and human health (Reynders *et al.* 2008). Sources of heavy metals in soils mainly include natural occurrence derived from parent materials and human activities (anthropogenic sources). Anthropogenic inputs are associated with industrialization and agricultural activities such as atmospheric deposition, waste disposal, waste incineration, urban effluent, vehicle exhausts, fertilizer application and long-term application of sewage sludge in agricultural land (Bilos *et al.* 2001; Hlavay *et al.* 2001; Koch and Rotard 2001). Pollution index methods have been widely used to assess soil environmental quality. The methods employ definite limit to differentiate and quantify the extent of soil pollution (He *et al.* 2007). Accordingly, the objectives of this study were to assess heavy metals contamination of paddy soil by using pollution index methods and to analyse the spatial distribution character of heavy metal contamination of paddy soil.

Methods
Soil sampling
The studying site is located in Xiangyin(Long. 112°30′—113°02′E and Lat. 28°30′—29°03′N), Hunan Province, China, and has an area of 1582 km^2, with Xiangjiang River, the largest river of this province, flowing across its centre. The river has seriously suffered from heavy metals pollution during the past decades due to the improper disposal of waste water from chemical factories and smelting plants. Although these factories have been legally closed by local governments in recent years, heavy metals accumulated in river sediments still poses a threat to the environment and health once they are released to soils by irrigation. To fully assess the status of heavy metals in agricultural soils, 99 soil samples, uniformly distributed in space, were collected from the 0-20 layer. The sample sites as seen in Figure 1 and positioned by use of hand-held GPS equipment. The soil samples were air-dried and ground in an agate mortar to pass through a 100-mesh sieve prior to chemical analysis.

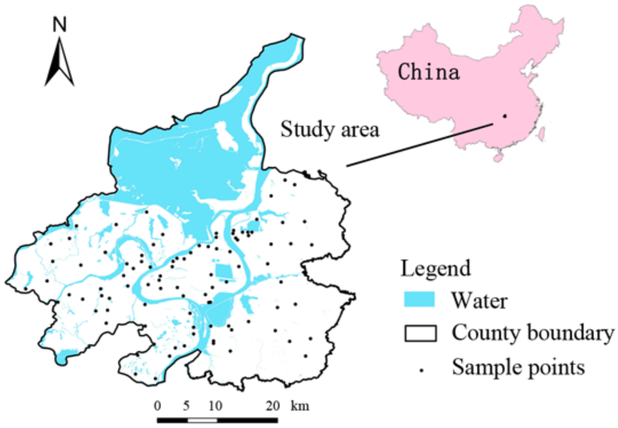

Figure 1. Location of the sample points.

Analytical Methods

Analysis of soil samples for total heavy metals were conducted based on the Environmental Monitoring of China Method. Briefly, 0.5g of soil were placed in a 50 mL Teflon crucibles, mixed with 10 mL of HCl and heated on a hot plate for about 2h until the digestion liquid has evaporated to approximately 3 mL. After cooling, 5 mL of concentrated HNO_3, 5 mL of concentrated HF and 2 mL of concentrated $HClO_4$ were consequently added and the digestion liquid was continuously reheated until no further oxidation of the sample was observed. After cooling, 1 mL of 1:5 HNO_3/H_2O was added to the digestion liquid and heated for 15 min at 95℃. The clear digests were diluted to 50 mL with distilled water and filtered into 100 mL plastic bottles. Metal concentrations were determined using a Hitachi atomic absorption spectrophotometer (Z-2300/2700). All standards and samples were analyzed in duplicate and mean values were shown.

Calculation formulas of pollution index methods

Two pollution index methods such as single-factor index and Nemero Comprehensive Index (NCI) method were employed to evaluate the environmental quality of the polluted soils. The calculation of the single factor index method can be expressed as:

$$P_i = C_i / S_i \qquad (1)$$

and the mathematical formula of the Nemero comprehensive index method is:

$$P = \sqrt{\frac{\left(\frac{1}{n}\sum_{i=1}^{n} P_i\right)^2 + \left[\max\left(P_i\right)\right]^2}{2}} \qquad (2)$$

where P_i is the pollution index of heavy metal i; C_i (mg/kg) is the actual monitoring data of heavy metal i; S_i (mg/kg) is the environmental value; P is the Nemero comprehensive pollution index.
Assessment criteria were established based on the National Environmental Quality Standards of China (GB15618-1995). The soil quality was classified on five levels: class I, excellent; class II, clean; class III, slightly polluted; class IV, moderately polluted; and class V, heavily polluted (Table 1).

Table 1. Assessment standard of soil Nemerow synthetical contamination index.

Class	NCI (P)	Pollution Level
I	$P \leq 0.7$	excellent
II	$0.7 < P \leq 1.0$	clean
III	$1 < P \leq 2.0$	slightly polluted
IV	$2.0 < P \leq 3.0$	moderately polluted
V	$P > 3.0$	heavily polluted

Results

Descriptive statistics and general variation in soil heavy metals

Table 2 shows the descriptive statistics for Pb, Cd, Cr, Ni, Cu and Zn on the analyzed sampling dates. All the mathematical and statistical computations were made using Statistical Package for Social Sciences (SPSS®) (Statistical Package for Social Studies) version 6.1, USA. Professional Statistics 6.1, 385, Marija J. Norusis/SPSS Inc., Chicago 1995). The CV_i of these data are in the following sequence: Zn > Cd > Pb > Cu > Ni > Cr.

Table 2. Statistical Table of heavy metal content of paddy soil in Xiangyin county (mg/kg).

Ci	Pb	Cd	Cr	Ni	Cu	Zn
criteria	250	0.3	250	40	50	200
Range	30.97-81.56	0.28-1.19	25.41-77.715	11.165-58.145	12.855-50.18	45.565-277.165
mean	55.7725	0.6608	53.6671	28.3569	30.4565	136.9583
max	81.5600	1.1900	77.7150	58.1450	50.1800	277.1650
min	30.9700	0.2800	25.4100	11.1650	12.8550	45.5650
Std. dev.	12.9312	0.2213	7.3771	4.2616	4.8265	47.6254
CV_i [a]	23.19%	33.49%	13.75%	15.03%	15.85%	34.77%
OCR[b]	0	95.96%	0	1.01%	1.01%	19.19%

a. Coefficient of variation, b. Over criteria rate.

Nemerow synthetical contamination index

According to the calculation of Eqs.1 and 2, the degree of heavy metals pollution indicated by the Nemero pollution comprehensive index were obtained based on actual heavy metal monitoring data (Ci) for the 99 soil samples. 1.01% of the sample points are at an excellent level, 17.17% at clean, 50.51% at slightly polluted, 31.31% at moderately polluted. The mean value of pollution comprehensive index is 1.65, indicating that the total pollution level is slightly polluted. Spatial analysis was carried out, Figure 2 is a map of the spatial distribution of heavy metal comprehensive pollution level, which was performed according to Kriging interpolation of Nemero pollution comprehensive index by using the geostatistical analyst extension of ArcGIS 9.2 (ESRI, Redlands, CA, USA).

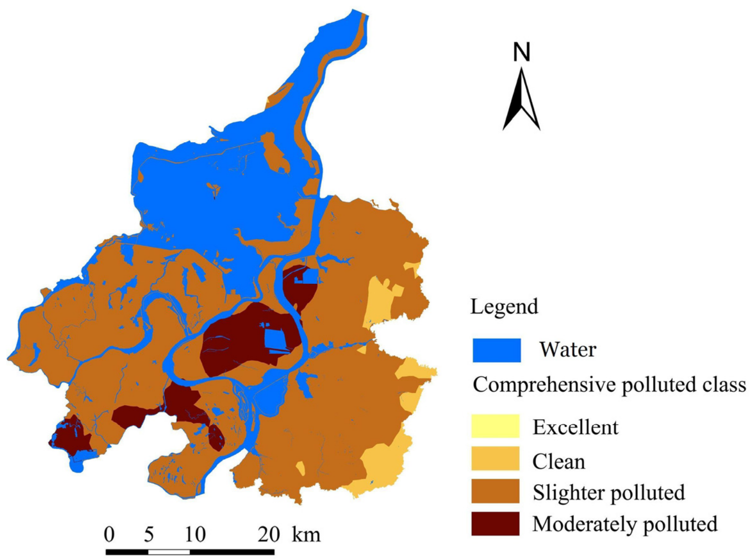

Figure 2. The spatial distribution of heavy metal comprehensive pollution class.

In the study area, the high level of heavy metal pollution was mainly found in the central area of land-reclamation from Dongting Lake which is consistent with the result of fairly high content of Cd in soils reported by Zhi-gang *et al.* (2006), and the low levels of heavy metal pollution found in the eastern hilly region. Lake pollution is high as parent material of the soil was river sediment with high levels of heavy metal contents, and on the other hand the heavy metals accumulated in the paddy soil by using water from Xiangjiang River for irrigation during the past 20 years. The relatively low level of heavy metal pollution in the eastern part of the study area could be explained by the following. Firstly, these metal contents may be representative of the local geochemical background in which the parent material of the soil is weathered slate residuals. Next, since the land altitude is higher, it is difficult to utilize water from Xiangjiang River for irrigation in the eastern hilly area.

With the development of modern agriculture and industry, increasing amounts of waste water and sewage may be discharged into Xiangjiang River, resulting in serious pollution of agricultural environments. Therefore, more emphasis should be put on heavy metal pollution in soils of the central part.

Conclusion

The total concentration of four heavy metals (Cu, Pb, Cr and Ni) in paddy soils of Xiangyin County was lower than the standard value of the National Environmental Quality Standards of China (GB15618-1995), which meet the environmental requirement of general farmland described in the above standards. In the study area, 1.01% of the sample points are at an excellent level and 17.17% at clean, while 50.51% are slightly polluted with 31.31% moderately polluted. The results show that most of the soils were suffering from slight pollution and attention should be focused on these areas. Additionally, heavy metal polluted soils were mainly distributed in the lake plain area, especially those directly reclaimed from the lake. The following points on soils heavy metal in the study area should be highlighted: 1) The Cd content exceeds the average background value of soil in China, and 2) irrigation with Xiangjiang River water is the main reason for high accumulations of heavy metal in paddy soils.

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