



Full Length Article

Spatio-temporal Variability of Heavy Metal Concentrations in Soil-rice System and Its Socio-environmental Analysis

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Abstract

Ninety nine surface soils and the corresponding rice grain samples from Wenling County in 2011 were used for the spatio-temporal variability investigation and evaluation of Cd, Cu, Ni, Pb and Zn concentrations to compare with values found in 2006. All heavy metals pose potential pollution risks in soil, especially Cd and Cu. The trend of increasing Cd contamination in rice was obvious. There were large percentage increases in Cd and Cu concentrations in soil and Cd and Ni in some rice regions. The regions with increased soil Cd concentrations were located in the north of Wenling with presence of intensive private small E-waste recycling companies. Concentrations of Cu in soil increased mainly in the east. Increases in the concentrations of Cd and Ni were observed for rice in the southwest and significantly different. Redundancy analysis (RDA) was used to provide a survey of variations in soil metal concentrations and then interpreted by consideration of soil properties and socio-economical observations. It was concluded that Zn concentration was related to the rapid development of local industry. The socio-environmental factors explained 43.3% of the variations in metal concentrations in soil. This study demonstrates the situation and accumulating trend of heavy metal concentrations in a soil-rice system caused by intensive human activities in Wenling County. © 2016 Friends Science Publishers

Keywords: Heavy metals; Soil-rice system; Spatio-temporal variation; RDA analysis; Socio-environmental factors

Introduction

Increasing soil pollution is of much concern worldwide including China, because of rapid increase in urbanization, industrialization and increasing trend of using agrochemicals. And with vehicle exhausts it has become a pivotal environmental problem over past decades (Tam *et al.*, 1987; Manzoor *et al.*, 2006; Cai *et al.*, 2012). The concentration and accumulation of heavy metals may lead to the functional disorder of agricultural soils, and crops cultivated accumulate both in the plants and grain (Wu and Chen, 2013). The consumption of heavy metals in several foods and human diet through crops can surely damage human health (Lee *et al.*, 2006; Li *et al.*, 2009; Franco-Uría *et al.*, 2009). Heavy metal contamination is extremely dangerous because of their non-biodegradability and biological long half-lives during the elimination from the body (Adriano, 2001; Gallego *et al.*, 2002; Cui *et al.*, 2005).

Two main factors of heavy metals, found in agricultural soils are natural and anthropogenic with earlier associated with weathering of parent rocks and pedogenesis.

In the last decades, because of human activities several heavy metals are being added to soil and excessive of natural inputs have been recorded even on a regional scale (Facchinelli *et al.*, 2001; Cai *et al.*, 2012). Commercial fertilizers and sewage sludge used in intensive agriculture are usually contaminated with significant concentrations of different heavy metals (Nicholson *et al.*, 2003; Franco-Uría *et al.*, 2009). In addition, the rapid expansion of industry in many areas, uncontrolled discharges and emissions from industrial plants and land application of industrial effluents, have caused widespread heavy metal concentration and contamination in agricultural soils (Zhao *et al.*, 2007; Hu *et al.*, 2013). It is therefore of due importance to quantify and evaluate the heavy metal concentrations in soils for defining the sources for monitoring soil quality, to aid policy designed to decrease heavy metal inputs and ensure food safety (Micó *et al.*, 2006).

Geostatistics is often used to discriminate human contributions from natural background concentrations, and to determine the regional distribution and possible sources of heavy metals (Chen *et al.*, 2008). Redundancy Analysis

(RDA) usually used in ecological studies, is one form of principal component analysis (PCA) and enable focus on the analysis of pollutant variances directly explained by external variables (Van den Brink *et al.*, 2003).

Wenling County is progressing and has become a developed region in China. Its economy is well-developed and ranked 21 nationwide in 2010. The private economy in this county is prosperous, with many small family-sized factories involved in machinery manufacturing, metal production, clothing and foot wear manufacturing. Rapid social and economic development may lead to many environmental problems. The most developed and industrialized regions such as Europe with intensive human activities, may have higher soil metal concentrations than less developed areas (Han *et al.*, 2002). Additionally, due to the lack of intensification and modernization, private enterprises in Wenling may readily produce numerous severe and negative environmental effects (Liu *et al.*, 2013). Further, the typical pattern of rice cultivation in south China uses small areas of paddy fields scattered throughout villages or surrounding individual households. These paddy soils are at high risk of contamination by family-sized local private industry. In addition, the northwest of Wenling County is especially famous countrywide for the recycling of electronic waste (E-waste), which may contain high concentrations of Cd, Cu, Pb, Hg, Zn and other metals.

Zhao *et al.* (2010) reported that due to the rapid social and economic development and intensive agriculture practices, increases in heavy metal concentrations in soil-rice systems in recent years deserve further investigation. In this study, we focus on the temporal variations of heavy metals in soil and rice grain between 2006 and 2011. This will help us to determine changes and trends in heavy metal contamination and provide the necessary information for effective environment management in the region. We also combine this data with socio-environmental parameters, providing an interpretation of the variations in heavy metal concentrations in rice paddy soils.

Materials and Methods

Study Area and Samples

Wenling County, southeast of Zhejiang Province, China has an area of 926 km² (121°10'-121°44' E, 28°13'-28°32' N) with population of more than 1.3 million. Wenling has a subtropical monsoon climate and weather with an annual average temperature and rainfall of 17.3°C and 1660 mm respectively. Wenling County contains 11 towns and 5 urban sub-districts in the administrative division (Fig. 1). Rice is the main grain crop in this area. Fertilizers including urea, calcium superphosphate and potassium chloride are being used and pesticides are also applied when necessary. Many industries including E-waste recycling workshops are distributed in this region.

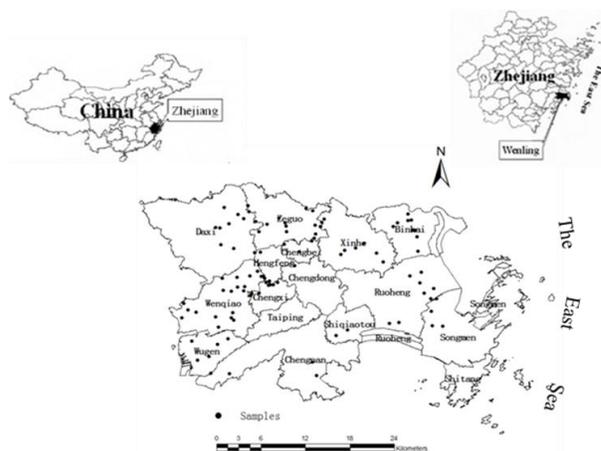


Fig. 1: Administrative divisions of Wenling and distribution of sampling points

Ninety nine rice grain samples and their corresponding soil samples (0-15 cm in depth) were collected from Wenling in October, 2011 (Fig. 1) following a completely randomized design. At least 5 sub-samples within distances of 10 m were taken from each field and then mixed to obtain composite samples. A Differential Global Position System (GPS) was utilized in recording of the coordinates of sampling locations. Soil samples were air-dried in the laboratory at room temperature, later on stones and debris were removed and passed through a polyethylene sieve (<2 mm). Portions of the sieved soil samples (50 g) were ground in an agate mortar and passed through a 0.149 mm mesh. Rice samples were oven-dried at 105°C for 1 h, and then at 70°C to constant weight. After removing the hulls, rice samples were comminuted and ground to pass through a 0.149 mm mesh. All the prepared samples were stored in polyethylene bottles for analysis.

Chemical Analysis

The total heavy metals concentrations in the soils were determined by acid digestion with a mixture of HNO₃-HF-HClO₄. Rice samples were digested with HNO₃ and H₂O₂. The concentrations of Cd, Cu, Ni, Pb and Zn were measured by flame-atomic absorption spectroscopy (FAAS, Jena novAA300, Germany) and graphite furnace atomic absorption spectroscopy (GFAAS, Perkin Elmer AA800, USA). Soil pH and electrical conductivity (EC) were measured in aqueous soil suspensions (1:2.5 and 1:5 soil water ratio, respectively). Soil organic matter (OM) was determined by the potassium dichromate wet combustion procedure (Agricultural Chemistry Committee of China, 1983). Soil amorphous and free Fe oxides were extracted by ammoniumoxalate (pH 3.0-3.5) and sodium citrate-sodium dithionite respectively. Soil particle size distribution (sand, silt and clay content) was analyzed by the hydrometer method (Agricultural Chemistry Committee of China, 1983).

Geo-statistical Analysis

Kriging is a linear geostatistical interpolation technique that provides the best linear unbiased estimates of quantities varying in space (Liu *et al.*, 2008). Among kriging techniques, ordinary kriging is probably the most familiar univariate interpolation method, and has been widely applied in environmental science (Wu *et al.*, 2006; Chen *et al.*, 2008; Zhao *et al.*, 2010). Based on the interpolation maps of heavy metals, the raster calculator is used to produce temporal variation maps. The raster calculator in the Spatial Analyst tool allows the use of arithmetic operators for the addition, subtraction, multiplication and division of two raster layers (ESRI, 2001). This method was used to calculate temporal changing percentages of each metal between 2006 and 2011 from the formula: $[(X_{2011}-X_{2006})/X_{2006}]$ (Olmos and Brich, 2010), where X is the raster layer converted from the interpolation map of each metal.

Redundancy (RDA) Analysis

Canonical ordination is usually used in studies of the relationships between ecological communities and environmental factors (Ter Braak, 1994). In our study, this method was applied to interpret the relationships between soil heavy metal concentrations in 2011 and the socio-environmental variables.

Redundancy analysis (RDA) is a linear canonical community ordination method. It was used to quantify the relationship between the response variable values (heavy metals), the environmental variable gradients and samples. The resulting ordination diagram shows sites as points, and heavy metals and environmental factors as vectors whose magnitude and angle indicate statistical significance and magnitude of the correlations with an ordination axis or another vector (Liang *et al.*, 2012). The proportion of heavy metal variations explained by the environmental variables is the canonical equivalent of the regression coefficient of determination, R^2 (Peres-Neto *et al.*, 2006).

Data Analysis

Correlation analysis was conducted between metal concentrations in soils and environmental factors to interpret the variations of heavy metal concentrations in the soil-rice system in 2011 in Wenling. Correlation analysis and descriptive statistics were performed with SPSS 16.0. Temporal change maps of heavy metals were prepared using the ArcGIS 10.1 version. RDA analysis and its resulting ordination diagram were generated using CANOCO for Windows, version 4.5.

Results

Descriptive Statistics of Heavy Metals

The coefficient of variation (CV) showed difference between 2006 and 2011 in soil heavy metals (Table 1).

The CVs of Cd and Cu changed considerably from 2006 to 2011 and those of all other heavy metals were much less variable, but higher in 2011 than 2006. Heavy metal concentrations in 2006 and 2011 in soil was compared on the basis of the background values of previously analyzed soils of Zhejiang province (China National Environmental Monitoring Centre, 1990) and the Environmental Quality Standard for Soils in China (GB 15618-1995 Ministry of Environmental Protection of China, 1995). In the study area, the average concentrations of Cd, Cu, Pb and Zn in 2006 and 2011 exceeded the background values. However, Ni concentrations were still below the background values. The average concentration of Cd in 2006 (0.31 mg kg^{-1}) and 2011 (0.32 mg kg^{-1}) exceeded the second grade standardized values (GB 15618-1995 Ministry of Environmental Protection of China, 1995) and it was little higher in 2011. The mean concentrations of Cu changed from below the threshold values in 2006 (41.13 mg kg^{-1}) to above the threshold in 2011 (55.09 mg kg^{-1}). The proportions of samples exceeding the second grade standardized values for Cd, Cu, Ni, Pb and Zn ranged from 0 to 27.08% in 2006, and from 1.01 to 22.22% in 2011, respectively. Cadmium was the most common heavy metal above the threshold in the two periods (27.08% in 2006, 22.22% in 2011). The percentages of samples of Cu exceeding the limit were relatively high in the two years (14.58% in 2006, 19.19% in 2011) and there was an increasing trend during these years.

Heavy metal concentration in rice was below the standardized values (Ministry of Health of China, 2012) in 2006 and 2011 (Table 2). Thus, the quality of rice in the study area was acceptable until 2011. Among these metals, the concentration of Cd changed greatly between 2006 and 2011. The mean Cd concentration nearly doubled from 0.072 mg kg^{-1} (2006) to 0.140 mg kg^{-1} (2011) during this period. The CV showed greater variability of Cd in 2011 than in 2006. The percentage of samples exceeding the standardized value of Cd increased from 9.38% in 2006 to 22.22% in 2011.

Temporal Changes of Spatial Patterns

The temporal variation maps of soil and rice grain are shown in (Fig. 2 and 3). The concentrations of Cd and Cu in soil increased in some regions during 2006-2011 (Fig. 2). The increased Cd concentrations in soil mainly occur in north of the study area. In the administrative divisions of Daxi and Zeguo towns, Cd concentrations increased by more than 20%. Parts of these regions had pollution growth rates exceeding 60% over this period. There were no significant increases in Ni in the study area, but in many regions, such as centre and east, the growth rates reached 20%. Concentrations of Cu increased mainly in the east of the study area by more than 14%, and the growth rates in Shiqiaotou reached more than 80%. Although there were increased concentrations of Pb and Zn in some regions but not significant over a wide concentration range.

Table 1: Descriptive statistics for heavy metals in the paddy fields of Wenling in 2006 and 2011(mg kg⁻¹)

Element and sampling year	Sample number	Mean±SD	Min	Max	C.V. (%)	Skewness	kurtosis	Background value ^a	Number exceeding background value	Percent	Second grade standardized value ^b	Number exceeding standardized value	Percent
Cd-2006 ^c	96	0.31±0.38	0.11	3.46	121.74	6.45	49.55	0.129	91	94.79	0.3	26	27.08
Cd-2011	99	0.32±0.54	0.12	5.29	168.75	8.29	75.52	0.129	97	97.98	0.3	22	22.22
Cu-2006	96	41.13±19.74	15.78	160.11	47.99	3.31	15.05	30.54	77	80.21	50	14	14.58
Cu-2011	99	55.09±95.76	17.96	934.20	173.82	8.23	74.30	30.54	86	86.87	50	19	19.19
Ni-2006	96	33.89±12.69	9.21	68.16	37.43	-0.11	-0.12	36.48	47	48.96	40	6	6.25
Ni-2011	99	28.64±16.88	5.93	165.16	51.51	5.35	43.64	36.48	16	16.16	40	5	5.05
Pb-2006	96	48.30±15.99	27.07	140.49	33.10	2.70	11.34	30.46	94	97.92	250	0	0
Pb-2011	99	50.23±35.08	31.23	362.25	69.84	7.53	65.30	30.46	99	100.00	250	1	1.01
Zn-2006	96	137.03±33.83	64.97	275.97	24.69	1.10	2.57	107.79	81	84.38	200	5	5.21
Zn-2011	99	129.11±59.40	68.73	649.67	46.01	7.09	61.16	107.79	77	77.78	200	3	3.03

^aThe background values of soil in Zhejiang province (China National Environmental Monitoring Centre, 1990)

^bChinese Environmental Quality Standard for Soils (GB 15618-1995 Ministry of Environmental Protection of China, 1995)

^cData of 2006 were from Zhao *et al.* (2010)

Table 2: Descriptive statistics for heavy metals in rice of Wenling in 2006 and 2011(mg kg⁻¹)

Element and sampling year	Sample number	Mean	SD	Min	Max	C.V. (%)	Skewness	Kurtosis	Standardized value ^b	Number exceeding standardized value	Percent
Cd-2006	96	0.072	0.105	0.002	0.467	146.0	2.18	4.47	0.2	9	9.38
Cd-2011	99	0.140	0.226	0.002	1.72	161.4	4.64	27.48	0.2	22	22.22
Cu-2006	96	3.09	0.96	0.71	5.79	31.1	0.30	0.02	10	0	0
Cu-2011	99	2.73	0.747	1.07	5.05	27.4	0.511	0.888	10	0	0
Ni-2006	96	0.221	0.234	0.045	1.717	105.9	3.74	18.81	nd ^c	nd	nd
Ni-2011	99	0.209	0.156	0.00140	0.959	74.6	1.77	4.96	nd	nd	nd
Pb-2006	96	ND ^a	ND	ND	ND	ND	ND	ND	0.2	0	0
Pb-2011	99	ND	ND	ND	ND	ND	ND	ND	0.2	0	0
Zn-2006	96	20.69	4.71	11.45	35.39	22.7	0.52	0.21	50	0	0
Zn-2011	99	18.68	4.22	11.32	31.39	22.6	0.509	0.0560	50	0	0

^aND: not detected

^bThe maximum levels of contaminants in foods in China recommended by Ministry of Health of China (2012)

^cnd: the standardized value for Ni is not defined by Ministry of Health of China

Concentrations of Cd and Ni in rice increased significantly in some regions during 2006-2011 (Fig. 3). Compared to 2006, Cd increased by more than 70% in most regions of the study area, especially in the west and south east, where the percent reached about 200-300%. Wenqiao town had the largest percentage increase in Cd, of more than 300%. Nickel concentration increased significantly in the central and western regions of the study area. In Wenqiao, Daxi, Chengbei and Songmen, the percentage increase was more than 7%, and exceeded 20% in Chengnan, Taiping and Chengxi town. Concentrations of Cu and Zn in rice did not increase significantly from 2006 to 2011. Zinc concentrations increased in the southeast of the study area, mainly by less than 20%.

Correlations between Soil Heavy Metal Concentrations and Environmental Variables

There were significant positive correlations ($P < 0.05$) between the pairs of heavy metals except Ni-Pb (Table 3).

Besides the properties of soils, anthropogenic factors were probably related to the study obtained from the Wenling Statistical Yearbook (2010) (Statistical Bureau of Wenling, 2010). Parameters involved in anthropogenic

factors were: population density, factory density, total industrial output value, rice output, and farmer income.

The correlations between soil heavy metals and soil properties indicated that Cd and Cu were positively related to organic matter (OM). Nickel was positively correlated with all the properties except OM and sand which had negative associations with it. Lead (Pb) showed positive correlations with OM and sand whereas it was negatively correlated with pH, clay and free Fe oxide. Zinc was positively correlated with EC, OM, silt and amorphous Fe oxide, and negatively with sand. Heavy metals had distinct relationships with the anthropogenic factors. Nickel had a positive relationship with population density and negative with rice output. Lead was negatively correlated with population density. Likely, zinc was positively correlated with farmer income, factory density and total industrial output value (scaled). In contrast, Cd and Cu were not significantly correlated with the anthropogenic factors.

RDA Analysis

The RDA analysis model examines the relationships between samples, species and environmental gradients together on the same diagram (McKinley *et al.*, 2005).

Table 3: The correlation between the contents of soil heavy metals in 2011

	Cd	Cu	Ni	Pb	Zn
Cd	1				
Cu	0.796**	1			
Ni	0.233*	0.335**	1		
Pb	0.813**	0.777**	-0.027	1	
Zn	0.670**	0.623**	0.407**	0.610**	1

*Significant at the 0.05 probability level

**Significant at the 0.01 probability level

Table 4: The correlation between soil heavy metals and socio-environmental variables

	pH	EC	OM	Sand	Silt	Clay	FR-Fe	AM-Fe	PD	FI	FD	TIOV(S)	TIOV	RO
Cd	-0.192	0.111	0.397**	0.097	-0.040	-0.143	-0.143	0.022	-0.069	0.004	0.075	0.076	0.010	0.137
Cu	-0.083	0.163	0.327**	0.118	-0.093	-0.092	-0.055	-0.006	-0.124	-0.098	-0.027	-0.014	-0.075	0.067
Ni	0.469**	0.414**	-0.094	-0.474**	0.329**	0.451**	0.612**	0.388**	0.221*	0.025	-0.040	-0.0004	0.001	-0.273**
Pb	-0.293**	-0.038	0.393**	0.202*	-0.129	-0.212*	-0.319**	-0.103	-0.199*	-0.006	0.086	0.097	-0.027	0.175
Zn	0.109	0.216*	0.310**	-0.241*	0.231*	0.113	0.173	0.331**	0.136	0.228*	0.246*	0.241*	0.119	0.039

*Significant at the 0.05 probability level

**Significant at the 0.01 probability level

OM: organic matter; EC: electrical conductivity; FR-Fe: free Fe oxide; AM-Fe: amorphous Fe oxide; PD: population density

FD: factory density; TIOV(S): total industrial output value (scaled); TIOV: total industrial output value

The above data were obtained from total values of towns in 2010 divided by the areas (square kilometer)

RO: rice output, the total values divided by sown area of each town (ton per mu); FI: farmer income (yuan per capita)

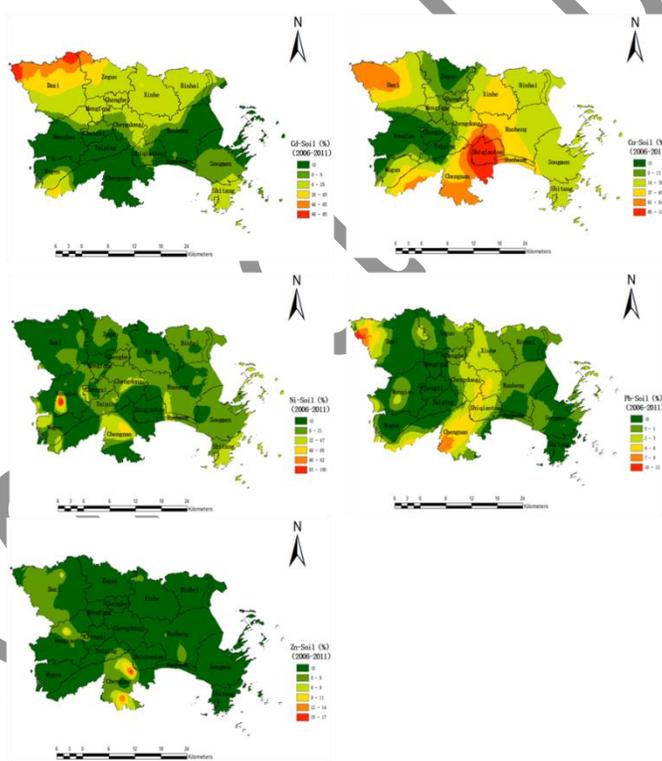


Fig. 2: Temporal variation maps of heavy metal concentrations in soil during 2006- 2011

The two groups of environmental variables, soil properties and anthropogenic factors, explained 36.3% and 16.6% respectively of the variations in soil heavy metal concentrations. Based upon interaction between the factors, a total of 43.3% of the variation in heavy metal concentrations could be explained by the model (from the canonical sum of the eigen values).

Soil heavy metals and environmental variables are shown by arrows in Fig. 4 and the sampling sites in 2011 belonging to each town in Wenling are indicated by symbols of different shapes and colors. Zinc and Cu occurred in the upper right quadrant. Socio-environmental factors were mostly contained in this part of diagram, including total industrial output value, factory density and farmer income.

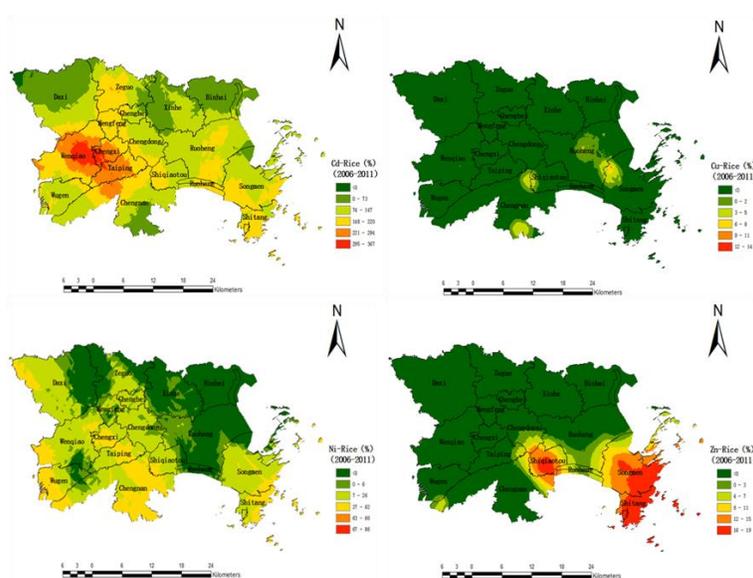


Fig. 3: Temporal variation maps of heavy metal concentrations in rice grain during 2006-2011

Also, there were soil properties such as EC, OM and amorphous Fe oxide in this part. Sampling sites were mainly from Zeguo and Chengxi towns. Lead and Cd were in the upper left quadrant where rice output was important and both organic matter and sand were in high quantity. Sampling sites which affected by these two metals were primarily located in Daxi and Wenqiao. High Ni concentration occurred in the lower quadrant separately in close relationship with many soil properties including silt, pH, clay and free Fe oxide. Soils from Binhai and Songmen had higher Ni concentrations than other sites.

Discussion

The average concentrations of Cd in soil in 2006 and 2011 exceeded second grade standardized values, with 2011>2006. This suggests a high potential pollution risk to soil environmental quality by Cd in the study area. This accumulation of Cd in soil increased the contamination of rice. Although quality of rice in the study area is currently below the national threshold, the increasing trend of Cd contamination in rice is significant and needs careful monitoring. The increased concentrations of Cd in soil were distributed in the northern regions to a significant degree. This may be due to the intensive E-waste recycling activities. Thus, monitoring and protection measures should be adopted to prevent further increases in Cd pollution. The concentrations of Cu in soils increased significantly, mainly in the east, between 2006 and 2011. Although there was no increase in Cu in rice until now, these increases in soils may lead to higher concentrations in rice in future. Although the concentrations of Zn and Pb in soil did not exceed the second grade standardized values and were above normal soil background. It is therefore critical to prevent further

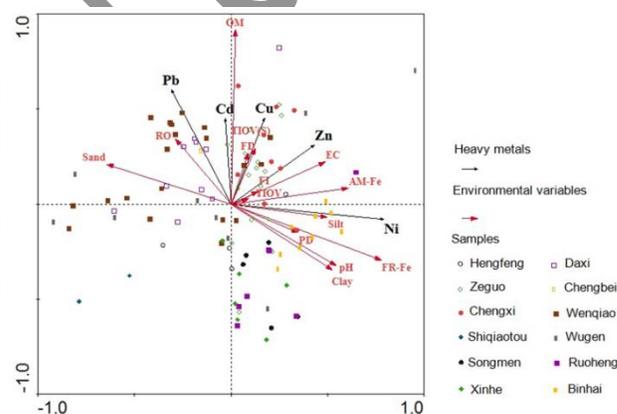


Fig. 4: Redundancy analysis (RDA) ordination diagram

PD: population density; FD: factory density; TIOV(S): total industrial output value (scaled); TIOV: total industrial output value. The above data were obtained from total values of towns in 2010 divided by the areas (square kilometer). RO: rice output, the total values divided by sown area of each town (ton per mu); FI: farmer income (yuan per capita); OM: organic matter; EC: electrical conductivity; FR-Fe: free Fe oxide; AM-Fe: amorphous Fe oxide

accumulation in soils. The coefficient of variation is the most discriminating factor for describing variability (Zhang *et al.*, 2007). The increasing trend of CVs in soil from 2006 to 2011 indicated that heavy metal concentrations in the paddy soils were probably affected by increasing human activities over this period (Liu *et al.*, 2008; Chen *et al.*, 2009). The proportions of soils exceeding threshold values of heavy metals (especially Cd and Cu) in the two periods certainly indicated that study area was non-uniformly contaminated with heavy metals. Heavy metals in soil and rice samples showed different increasing trends especially

with Cd, Cu and Ni. This suggests that heavy metal concentrations in rice correlate not only to heavy metals in soil but also to soil properties and other factors, such as cultivation methods and rice varieties etc. (Zhao *et al.*, 2010; Hu *et al.*, 2011).

Heavy metal concentrations showed spatial variations in the study area. Greater soil concentrations of Cd, Cu, Zn and Pb mainly occurred in 2011, in towns on north and west of the study area (Fig. 4). The distributions of these metals were probably caused by the large numbers of E-waste recycling workshops in Wenqiao, Daxi and Zeguo towns. In these workshops, simple techniques such as burning and smelting, combined with a lack of efficient prevention of pollutant emission losses, would certainly result in heavy metal contamination of the local environment (Fu *et al.*, 2008). The distribution of Ni differed from other metals because of their different sources (Biasioli *et al.*, 2006).

Inter-element relationships can provide some information on the sources and pathways of the heavy metals (Lu *et al.*, 2010; Li and Feng, 2012). There was strong correlations recorded between Cd, Cu, Zn and Pb concentrations in soil in 2011 (Table 3), based on comparison with the background values, which indicated similar anthropogenic sources of the metals, including industrial discharges, application of agrochemicals, animal manure, etc. (Chen *et al.*, 2008). Ni had significant positive correlations with Fe oxides, one of the major components of natural soils, suggesting that natural factors control its distribution (Yang *et al.*, 1999). The positive correlations between Cd, Cu, Zn, Pb and soil OM demonstrated the capacity of the organic matter to immobilize metals and minimize their losses (Vega *et al.*, 2004). Heavy metals may be added to certain livestock feeds as supplementary trace elements for health or as growth promoters (Nicholson *et al.*, 2003). Therefore, applications of livestock manure can result in elevated heavy metal concentrations in soils (Franco *et al.*, 2006). This may explain the positive correlations between heavy metals and OM. The correlations between heavy metals and anthropogenic factors (i.e. relevant economic and social parameters in the study area) (Statistical Bureau of Wenling, 2010) indicating the influence of economic and social development on the accumulation of heavy metals in paddy soils. Socio-environmental factors had significant impacts on Zn concentrations. This may be because Zn is one of the main industrial age metals leading to the contamination of soil (Han *et al.*, 2002). The correlations between Zn and soil properties (sand, silt and amorphous Fe oxide) showed that Zn seem to have both natural and anthropogenic origins (Cai *et al.*, 2012). Cadmium and Cu had no statistical correlations with the anthropogenic factors. This may indicate that anthropogenic factors identified in this study were not detailed enough to indicate their relationships rather than for any relationship between them. Additionally, because most of the industry in this area consisted of family workshops, the impact of E-waste recycling on heavy metal

accumulation was probably not reflected in the anthropogenic factors.

RDA analysis indicates the relationship between soil concentrations of heavy metals and environmental variables at the town scale. Daxi and Wenqiao soils contained high concentrations of Cd and Pb, where intensive agriculture practices (OM and rice output) were followed. Previous studies also reported that Cd and Pb concentrations in soil were due to the application of commercial fertilizers and livestock manure (Taylor, 1997; Mann *et al.*, 2002; Franco-Uría *et al.*, 2009). High concentrations of Cu and Zn in Zeguo and Chengxi soils were recorded, where intensive industrial plants were found and relatively farmers have high incomes. This suggests that high heavy metal concentrations may occur in developed and industrialized areas, due to intensive human activities (Han *et al.*, 2002; Loska *et al.*, 2004; Velea *et al.*, 2009). Many private enterprises which are insufficiently modernized can cause soil pollution in the surrounding areas. Soils from Binhai and Songmen had higher Ni concentrations, with the heavy metal variation being accounted for by soil properties (silt, clay and Fe oxide). However, the close relationship between Ni and population density and rice output also suggests a considerable influence of social factors on Ni concentrations in these soils. Due to the relatively low variations in heavy metal concentrations interpreted by the environmental variables (43.3%), it is also possible that socio-economic factors other than we analyzed may be involved. Therefore, methods to provide better characterization of anthropogenic factors are required.

Our research gave a general indication of heavy metal variability in soil and rice and the socio-environmental interpretation of concentrations of heavy metals in soils of the study area. Isotopic fingerprinting, which has a high discriminatory power, may also be useful for the identification of source of heavy metals in future studies (Duzgoren-Aydin *et al.*, 2004; Cheng and Hu, 2010; Sun *et al.*, 2011).

Conclusion

Between 2006 and 2011, high potential risks to soil environmental quality and rice caused by Cd pollution were suggested in the study area. The increased risk of Cu contamination in soils was also confirmed. Increasing trends of soil Cd and Cu concentrations in paddy soils were probably caused by intensive agricultural practices and distribution of industries in the regions. In addition, E-waste activities may be an important cause of the spatial and temporal variation of Cd and Cu. There was a close relationship between economic and social development and the accumulation of Zn in paddy soils. Our study provides reliable data to assist the development of strategies for reducing heavy metal accumulation and concentration in agricultural land and soils and to effectively target policies for preserving soil quality and food safety in the long-term.

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