Pol. J. Environ. Stud. Vol. 26, No. 4 (2017), 1809-1823
DOI: *10.15244/pjoes/68533*

Original Research

Effects of Land Reclamation on Distribution of Soil Properties and Heavy Metal Concentrations, and the Associated Environmental Pollution Assessment

Xiaolu Yan[1, 2, 3], Yuanman Hu[1]*, Yu Chang[1], Yuehui Li[1], Miao Liu[1], Jingqiu Zhong[3], Danhua Zhang[1, 2], Wen Wu[1, 2]

[1]CAS Key Laboratory of Forest Ecology and Management, Institute of Applied Ecology, Chinese Academy of Sciences, No. 72, Wenhua Road, 110016, Shenyang, China
[2]University of Chinese Academy of Sciences, No. 19, Yuquan Road, 100049, Beijing, China
[3]College of Urban and Environmental Science, Liaoning Normal University, No. 850, Huanghe Road, 116029, Dalian, China

Received: 4 November 2016
Accepted: 16 January 2017

Abstract

We used the "space for time substitution" method to test the effects of reclamation history on evolution processes of soil properties and heavy metal concentrations. A total of 129 surface soil samples were collected in a reclamation zone of the Liaohe River Delta, which was divided into three reclaimed zones (1930s, 1960s, and 1990s), marsh, and intertidal flats. Soil metals (Fe, Mn, Cd, Cr, Cu, Ni, Pb, and Zn), soil particle size, pH, salinity, soil organic carbon (SOC), nitrate nitrogen (NO_3^--N), and available phosphorus (AP) were analyzed. Results obtained from correlation analysis and factor analysis showed that there were significant positive correlations ($p<0.01$) between the fine particle fractions (clay and silt) and metal concentrations. Using ANOVA, we found that soil erosion and leaching desalination had significant effects on the distributions of soil properties and heavy metal contents in short-term reclamation (1990s zone), and that sustained human activities have played an important role in long-term reclamation (1960s zone). Results of the enrichment factor (EF) and inverse distance weighting (IDW) indicated that Cd pollution had reached high pollution levels, followed by Cr pollution. Taking necessary measures is significant for sustainable utilization and management of coastal resources.

Keywords: land reclamation, soil properties, heavy metal, environmental pollution

*e-mail: hymlandscape@163.com, huym@iae.ac.cn

Introduction

Wetlands provide essential ecosystem services to people and the environment, including flood protection, water supply and purification, food productivity, erosion control, wave attenuation, shoreline stabilization, wildlife habitat, climate regulation, and amenity [1]. However, with the development of the economy and population growth, the contradiction between land supply and human demand is becoming increasingly acute. The reclamation of coastal wetlands has become a popular way to meet the increasing demand of new land for living and development [2]. In the past, many coastal countries around the world have obtained new land through reclamation [3-4]. China has now become the largest country that reclaims land from the sea, and the area of coastal wetlands has shrunk by 51.2% since 1949 [5]. Reclamation has been found to change shoreline evolution and wetland hydrology, deteriorate soil and water quality, alter vegetation succession, benthic animal and microbial communities and fisheries, and impair ecosystem functioning and services [6]. Therefore, this study on the effects of reclamation on wetland ecosystems is of great importance for effective management and sustainable development of coastal resources.

Soil is not only the material on which land organisms live; it also plays important roles in material cycles and energy exchange in terrestrial ecosystems. Large numbers of coastal wetlands around the world have been converted into reclaimed land in recent decades because of rapid economic development in coastal areas. Numerous studies of reclamation-induced changes in soil function have therefore been conducted. Portnoy and Giblin [7] and Sun et al. [8] found that soil bulk density tended to increase, while soil moisture, electrical conductivity, and particle size tended to decrease during reclamation. Li et al. [9] have suggested that soil nutrient distribution was significantly affected by land-use intensity after land had been reclaimed, with relatively high soil salinity in aquaculture ponds and high soil organic matter (SOM), available phosphorous (AP), and nitrate nitrogen (NO_3^--N) contents in agricultural fields. Soil organic carbon (SOC) plays important roles in the global carbon cycle, and small changes in SOC contents can strongly affect atmospheric CO_2 concentrations, leading to climate change [10]. Bai et al. [11] discovered that converting wetland to cropland and long-term reclamation projects significantly decrease the SOC contents of the top 50 cm in soil. Wetlands are important carbon pools and important sinks for heavy metals (through a range of physicochemical processes). It is difficult to remediate saline soil polluted with heavy metals because the heavy metals are very mobile. Activities on reclaimed land (e.g., agriculture, industry, and urban expansion) affect soil properties and promote the release of heavy metals, increasing heavy metal concentrations in water bodies and organisms [12]. Sources of heavy metals have been identified and the effects of land use on heavy metal behaviors in reclaimed soils have previously been studied. Li et al. [13] revealed that different land-use structures and industry types affected heavy metal concentrations in soils. They found significantly higher As, Cu, Hg, Pb, and Zn concentrations in industrial areas than the background concentrations, and moderate As and Hg pollution in forested land. Some studies have been focused on heavy metals in wetland and agricultural soils, including heavy metal enrichment, spatial distributions, and the potential hazards posed to public health [14-15]. At present, heavy metal pollution has yet to be controlled effectively, especially in developing countries, and the attention of policymakers is desperately needed.

Little historical data for soil is available, but the space-for-time substitution method has been used to determine spatial and temporal variabilities in the evolution of soil function after reclamation [16]. Reclamation history is often used as a representative indicator when using this method, and is an important factor regarding the effects of land reclamation on soil property evolution and heavy metal accumulation [15]. There may be uncertainties when determining land use history and management practices, but in previous studies these uncertainties were found not to fundamentally change the direction of the soil changes [17]. Reclamation history data for coastal areas in eastern and southern China have recently been gathered, but it has not been surveyed in northeastern China. The Liaohe River Delta, in Liaoning Province, China, is adjacent to the northern part of Liaodong Bay in the Bohai Sea, with a total area of about 5,000 km^2. The Delta is formed by sediment deposited by the Daliaohe, Dalinghe, Xiaolinghe, and Liaohe (or Shuangtaizihe) rivers. The second largest permanent reed (*Phragmites australis*) marsh in the world is around the delta, which was designated the Shuangtaihekou National Nature Reserve in 1986 and, by the Ramsar Convention, a Wetland of International Importance in 2005. Panjin is the largest city in the Liaohe River Delta, and Dawa County in Panjin City is the main reclaimed area. The reclamation history was determined from historical records, remote sensing images, and information provided by local inhabitants. *Dawa County Annals* and the *Panjin Water Conservancy Annals* indicated that the study area has had three periods of reclamation: massive reclamation before 1949, all-round development and reclamation of state-owned land after 1949, and comprehensive large-scale development since the 1990s. A number of studies of soil function evolution in the study area have been performed [18-20], but the effects of reclamation history on soil function evolution processes are yet to be investigated.

This study was undertaken in Dawa County with the aim of exploring the effects of reclamation history on biogeochemical changes in soils. Specifically, the objectives of this study were 1) to reveal how reclamation history affects soil property evolution and heavy metal concentrations and 2) to delineate the levels of soil heavy metal pollution posed by reclamation in the study area.

Material and Methods

Study Site

We investigated an area (121°31′-122°10′E, 40°39′ -41°12′N) of about 1,249 km^2 located in southwest Dawa County, including intertidal flats, permanent reed marsh (i.e., Shuangtaihekou National Nature Reserve) and reclaimed zones (Fig. 1). The region experiences a semi-humid temperate monsoon climate, where the annual average temperature is 8.3-8.4ºC, annual average precipitation is 611.6 mm, and annual evaporation is between 1,390 and 1,705 mm [21]. In this study, the area was divided into three different zones according to reclamation history (they were reclaimed during the 1930s, 1960s, and 1990s, respectively). Soil sampling sites were selected from each zone.

Sample Collection and Preparation

As physicochemical properties and heavy metal elements of the surface soil are more sensitive to anthropogenic activities than that of the deeper layers [22], 129 samples in the top 20 cm of soil – including 33 samples from the 1930s zone, 27 samples from the 1960s zone, 33 samples from the 1990s zone, 30 samples from the reed marshes, and six samples from the intertidal flats – were collected from the study area during October 2014, after the paddy rice fields had been drained and reaped (Fig. 1). In each reclamation zone, the location of sampling sites was determined at random, with special consideration for different land use, vegetation, and soil types to ensure even coverage of the whole study area. The position of each sample site was identified with a portable GPS receiver to an accuracy of 3-5 m. In order to obtain a representative soil sample at each site, three sub-samples 50-100 m apart were collected and then mixed fully to form a composite sample. All soil samples were packed in polyethylene bags and returned to the laboratory.

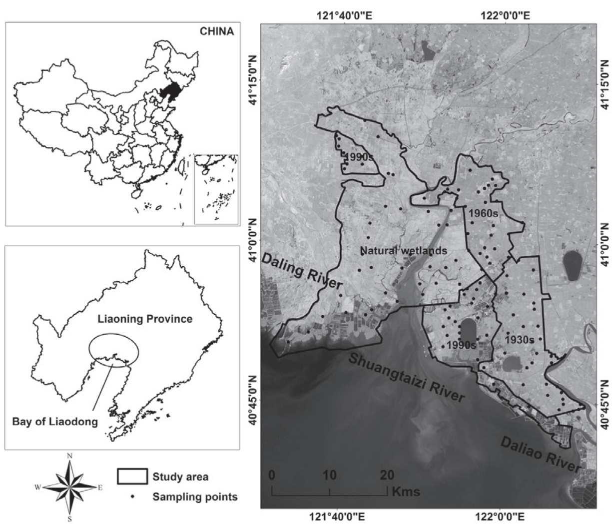

Fig. 1. Location of the study area and distribution of sampling points.

Sample Chemical Analysis

These soil samples (average weight of 1 kg) transported to the laboratory were air-dried under normal temperature for three weeks. After air drying naturally, all samples were sieved through a 2-mm nylon sieve to remove the roots and coarse debris. Portions of the soil samples were then ground with a pestle and mortar until all particles passed through a 0.149-mm nylon sieve. Fourteen indices were selected to represent soil quality and the average conditions of heavy metal concentrations. Soil pH was analyzed in soil slurry with a pH meter at a 1:2.5 (g g^{-1}) soil-to-water suspension. Soil particle size distribution was obtained using a laboratory's laser particle size analyzer (Microtrac S3500). Soil salinity was determined directly during the field survey using a salt-water sensor (SDI-12/RS485, Australia). Soil organic carbon (SOC), nitrate nitrogen (NO_3^--N), and available phosphorus (AP) are important indicators of the soil nutrient content. SOC was measured by a TOC analyzer (HT1300, Analytikjena, Germany) after removing soil carbonates with 1 M HCI. Soil AP was analyzed using the photoelectric colorimetry method, while the NO_3^--N was extracted using the deoxidization photoelectric colorimetry method.

For analysis of total concentrations of soil metals, including iron (Fe), manganese (Mn), Pb, Cu, Cd, Zn, Cr and Ni, soil samples (0.149 mm) were digested using an HNO_3-HF-$HClO_4$ mixture in Teflon tubes at 160ºC for 6 h and then measured by inductively coupled plasma-atomic emission spectroscopy (ICP-AES) (HITACHI P-4010, Japan). Parallel analysis and standard reference materials (GBW-07401, Chinese Academy of Measurement Sciences) were used to assess quality assurance and control. The obtained recovery rates for the standards were between 95.12 % and 104.47 %. The analytical results met the standard requirements of Technical Specification for Soil Environmental Monitoring HJ/T 166-2004 (National Environmental Protection Administration of China 2004).

Statistical Analysis and Enrichment Factor

Descriptive statistics were used to calculate the statistical parameters of soil properties and metals to evaluate the data distribution. The Kolmogorov-Smirnov (K-S) test was used to examine the normality of the probability distributions of soil variables. When these variables were not passing the normality test at the 0.05 significance level, they were normalized by logarithmic transformation or Box-Cox transformation. Pearson's correlation analysis and factor analysis were carried out to investigate the relationships between the soil properties and selected heavy metals. The soil properties and total metal concentrations from different zones were compared using multivariate analysis of variance (ANOVA), followed by post hoc least significant difference test

(p<0.05). All statistical analyses were processed with SPSS 13.0 (SPSS Inc., USA). Data transformation was performed using MiniTab 17.0.

The EF (enrichment factor) is often used to evaluate enrichment status of soil heavy metals and was calculated by the following formula [23]:

$$EF = \frac{C_n}{C_{ref}} \Big/ \frac{A_n}{A_{ref}} \quad (1)$$

…where C_n and A_n are the measured and background values of a given heavy metal (unit: mg kg^{-1}), respectively, and C_{ref} and A_{ref} are the measured and background values of the reference metal (unit: mg kg^{-1}), respectively. To identify the potential spatial distribution of soil heavy metal pollution, the inverse distance weighting (IDW) was used for interpolation and for producing maps with ArcGIS software for desktop (ver. 10.1, ESRI). The process of the interpolation was performed as described in this reference [24]. Figures were drawn by OriginPro 8.0 and ArcGIS 10.1.

Results and Discussion

Descriptive Statistics

Descriptive statistics for the soil properties and heavy metal concentrations are shown in Table 1. The Fe and Mn concentrations were high (>100 mg kg^{-1}), and the mean Cr and Zn concentrations were relatively high, at 154.95 and 102.21 mg kg^{-1}, respectively. The heavy metal concentrations and the soil properties apart from pH varied widely in different samples. The pH values varied in a range from 6.65 to 8.80, indicating the soil samples were weakly acidic to weakly alkaline. The salinity was 0.01-0.62 ms m^{-1}, and the SOC, NO_3^--N, and AP contents (representing soil fertility) were 0.05-2.88%, 0.25-10.72 mg kg^{-1}, and 0.72-26.46 mg kg^{-1}, respectively. The coefficients of variation (CV) for the heavy metal elements varied in a range of 12.63-58.23%. Among them, Cd, Cr, Cu, Mn, and Pb had relatively high CVs (>30%), suggesting that their distributions might be influenced by human activities. Fe, Ni, and Zn had lower CVs (<30%), indicating they were distributed in the study area more evenly. Salinity, NO_3^--N content, and AP contents had high CVs at 97.33%, 94.68%, and 74.80%, respectively, indicating that they varied widely in the study area and implying that human activities have affected them. For the soil fractions, statistical results indicated that the major soil texture was from silt loam to sandy loam in the study area (Table 1).

Correlation Analysis of Soil Physicochemical Properties and Metals

The Pearson correlation matrix shown in Table 2 was used to identify correlations between the metal concentrations and soil properties. The six anthropogenic heavy metal (Cd, Cr, Cu, Ni, Pb, and Zn) concentrations significantly positively correlated (p<0.01) with each other. The Cu concentration strongly correlated with the Ni and Zn concentrations (the correlation coefficients were 0.52 and 0.71, respectively), and the Cr-Ni, Cr-Zn,

Table 1. Descriptive statistics for soil properties and heavy metal concentrations in topsoil.

Variable	Minimum	Median	Maximum	Mean	S.D.	C.V. (%)
Cu (mg kg^{-1})	4.55	15.65	31.67	17.23	6.05	35.10
Cr (mg kg^{-1})	50.65	153.43	275.79	154.95	52.16	33.66
Cd (mg kg^{-1})	0.26	1.63	3.46	1.41	0.82	58.23
Ni (mg kg^{-1})	20.87	39.33	59.34	40.71	9.13	22.44
Zn (mg kg^{-1})	53.40	99.59	149.50	102.21	19.44	19.02
Pb (mg kg^{-1})	2.95	11.26	19.57	11.97	3.66	30.54
Mn (mg kg^{-1})	25.78	96.42	211.90	100.46	30.94	30.80
Fe (g kg^{-1})	0.63	0.93	1.18	0.93	0.12	12.63
Salinity (ms m^{-1})	0.01	0.11	0.62	0.15	0.14	97.33
pH	6.65	7.95	8.80	7.89	0.48	6.07
SOC (%)	0.05	0.84	2.88	0.93	0.48	51.81
NO_3^--N (mg kg^{-1})	0.25	1.56	10.72	2.35	2.23	94.68
AP (mg kg^{-1})	0.72	5.61	26.46	7.23	5.41	74.80
Clay (%)	1.04	2.83	6.02	3.04	1.04	34.09
Silt (%)	9.59	37.19	66.58	37.63	10.46	27.81
Sand (%)	25.60	59.07	89.25	58.72	11.63	19.81

Table 2. The correlation of soil properties and heavy metal concentrations

	Cu	Cr	Cd	Ni	Zn	Pb	Mn	Fe	Salinity	pH	SOC	NO_3^--N	AP	Clay	Silt	Sand
Cu	1															
Cr	0.47**	1														
Cd	0.22**	0.20**	1													
Ni	0.52**	0.71**	0.30**	1												
Zn	0.71**	0.52**	0.19**	0.57**	1											
Pb	0.43**	0.25**	0.23**	0.35**	0.37**	1										
Mn	0.29**	-0.11	0.05	-0.10	0.16	0.13	1									
Fe	0.57**	0.35**	0.24**	0.27**	0.28**	0.20**	0.39**	1								
Salinity	0.03	0.25**	0.07	0.20*	-0.02	0.07	0.01	-0.03	1							
pH	-0.14*	-0.09	-0.13	-0.17	-0.04	-0.33**	0.24**	0.18**	0.38**	1						
SOC	0.32**	0.24**	0.34**	0.26**	0.25**	0.13	0.02	0.07	-0.30	-0.32**	1					
NO_3^--N	0.01	-0.07	-0.06	-0.07	-0.08	0.18	0.09	-0.05	-0.26**	-0.22*	0.33**	1				
AP	-0.10	-0.01	0.12	-0.06	-0.05	0.16	-0.03	-0.01	-0.09	-0.10	0.05	0.15	1			
Clay	0.29**	0.37**	0.13**	0.29**	0.23**	0.13	0.20*	0.16	0.27**	0.18	0.37**	-0.21*	-0.22*	1		
Silt	0.35**	0.43**	0.23**	0.35**	0.32**	0.05	0.17	0.06	0.13	0.05	0.23**	0.04	-0.22*	0.78**	1	
Sand	-0.37**	-0.44**	-0.15**	-0.36**	-0.27**	-0.06	-0.17	-0.12	-0.07	0.01	-0.26**	0.03	0.28**	-0.82**	-0.99**	1

*p<0.05; **p<0.01

and Ni-Zn correlation coefficients were 0.71, 0.52, and 0.57, respectively, suggesting that these metals could have similar anthropogenic origins. The Pb concentration correlated (p<0.01) with the Cu, Ni, and Zn concentrations, but weakly correlated with the Cd and Cr concentrations, implying that Cd, Cr, and Pb probably had different sources. Turer et al. [25] has reported that atmospheric deposition is an important source of Pb-to-soil and sediment. As the major components of rock-forming, the Fe and Mn concentrations significantly positively correlated (p<0.01), but the Fe concentrations more significantly correlated (p<0.01) with the concentrations of the other heavy metals than with the Mn concentration.

In all measured soil properties, some typical correlations could be identified based on the correlation matrix analysis. As shown in Table 2, soil pH significantly positively correlated with salinity (p<0.01) and negatively correlated with SOC content (p<0.01). In previous studies, soil salinity and pH were found to strongly affect soil structure and SOM decomposition [26-27]. In this study, therefore, although the correlation between soil salinity and SOC was not significant, a weak negative correlation could be observed between them. Variations in SOC content will affect crop productivity and soil fertility and also regional and global carbon cycles [28]. It was obviously observed that SOC had significantly positive correlation with clay and silt particles and negative correlation (p<0.01) with sand content. This result kept in line with Li et al. [29] and Baritz et al. [30], who reported that the positive effect between SOC and clay and silt content were due to the stabilization of SOM by fine particles and physical protection of SOC from oxidation by the relatively smaller spaces in soils. Moreover, a significant correlation was observed between SOC and NO_3^--N, which might imply that soil organic matter was also likely one of the main sources of soil N supply [31].

It has been found that SOM, pH, salinity, and soil texture directly and indirectly affect heavy metal mobilities and solubilities in soil [15, 24]. As shown in Table 2, apart from Cr (p<0.01) and Ni (p<0.05), no other metals showed significant correlation with salinity. Although the correlations between pH and heavy metals (except Cu and Pb) were not significant, we found that pH had a weak reverse impact on heavy metal concentrations and were in agreement with Li et al. [13] and Zhang et al. [32], who reported that the low-pH soil could facilitate the migration and availability of heavy metals. No significant correlations were observed between NO_3^--N, AP, and metals. The Cd, Cr, Cu, Ni, and Zn (but not the Pb) concentrations significantly positively correlated (p<0.01) with the SOC, clay, and silt contents, and significantly negatively correlated (p<0.01) with the sand content. These findings were supported by previous research and suggested that SOM and finer particle-size fractions (clay and silt) could act as major sinks for heavy metals due to their strong absorption capacity and increased surface areas [33-34]. In general, the correlation coefficients shown in Table 2 indicated that the particle fractions affected the heavy metal concentrations more than did the other soil properties.

Table 3. Rotation sums of squared loadings for soil properties and heavy metals (Factor analysis).

Raw data			
Component	Total	Percent of Variance (%)	Cumulative (%)
1	3.14	19.61	19.61
2	2.96	18.53	38.14
3	1.99	12.43	50.57
4	1.78	11.12	61.69
Fe-normalization data			
Component	Total	Percent of Variance (%)	Cumulative (%)
1	2.90	19.31	19.31
2	2.63	17.51	36.82
3	1.86	12.43	49.25
4	1.63	10.87	60.12

Extraction method: principal component analysis
Rotation method: varimax with Kaiser normalization.
Marked loadings are ≥0.60

Factor Analysis

Normalization with reference elements can remove the metal concentration variance caused by natural processes and allow anthropogenic effects to be described quantitatively [35]. Iron is widely used as a reference metal because it is naturally abundant and is mainly supplied through rock and soil weathering [24,36]. The Fe concentration had a low coefficient of variation and significantly positively correlated with the concentrations of the other metals (Tables 1 and 2), suggesting that Fe could be used to normalize the data and correct for the influence of the soil particle size. Factor analysis was performed on the raw data and Fe-normalized data to further identify relationships between the parameters and the heavy metal sources (Table 3 and Fig 2).

Four factors, accounting for 61.69% of the total variance, were extracted from the raw data (Table 3). The rotated component matrix indicated that the first factor, called F1, accounted for 19.61% of the total variance and was positively and strongly related to the Cr, Cu, Ni, and Zn concentrations, soil particle size fractions, and SOC content, confirming the correlation matrix analysis results. The second factor included salinity, pH and NO_3^--N, with loadings >0.60 (Fig. $2a_1$). The Fe and Mn were in F3, with a factor loading >0.7 (Fig. $2a_2$), suggesting Fe and Mn mainly had common sources. This result was in agreement with Zhou et al. [37] and Mico et al. [38], who stated that the variability of Fe and Mn was controlled by soil parent materials in the reclaimed wetlands of estuaries. The fourth factor accounted for 11.12% of the total variance and

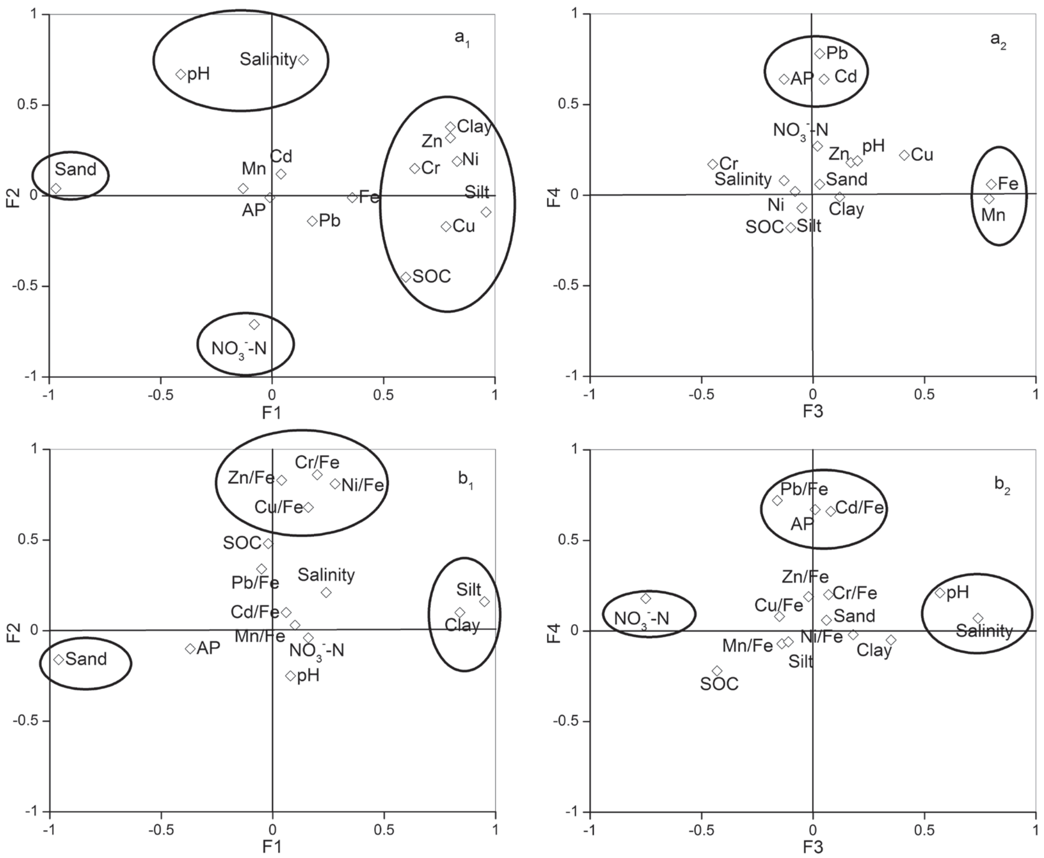

Fig. 2. Scatter plots of rotated component matrix for soil properties and heavy metals (a: Raw data, b: Fe-normalization).

was strongly related to the Cd, Pb and AP content (with loadings of 0.64, 0.78, and 0.64, respectively). Similarly, four factors, explaining 60.12% of the total variance, were also extracted from the Fe-normalized data (Table 3). The first factor included grain-size variables (loadings >0.80) and the second factor included the Fe-normalized Cr, Cu, Ni, and Zn concentrations (loadings >0.60) (Fig. $2b_1$). This indicated that soil particle size affected the Fe-normalized heavy metal concentrations little and that normalization was necessary. The coefficients were different, but the third and fourth factors extracted from the Fe-normalized data matched the second and fourth factors extracted from the raw data (Fig. $2b_2$).

Factor analysis gave information on three aspects of the relationships between the heavy metal concentrations and soil properties. The soil fractions can significantly affect the soil properties and heavy metal distribution. The finer soil particle-size display higher metal concentrations because of increased surface areas, higher clay-silt mineral and organic matter contents, and the presence of Fe-Mn oxides and sulphides [39]. The reverse effects of salinity on NO_3^--N (with the loading >0.70) indicated that high soil salinity may result in decreasing N mineralization [40]. However, Mladenoff [41] indicated that N mineralization increased as the pH increased in acidic soil, which was the opposite of what we found. This could have been because the relationship between pH and N mineralization involves relatively complex processes in weakly alkaline soils under anaerobic conditions [42]. The factor analysis results showed that Cr, Cu, Ni, and Zn could have common sources related to anthropogenic activities, particularly industrial activities [43]. The strong relationships between the Cd, Pb and AP content indicated that persistent fertilizer application in the study area may have increased the Cd, Pb, and P concentrations in soil [44]. Shan et al. [45] found that agrochemicals applied to agricultural land are the main anthropogenic sources of Cd and Pb.

Variance Analysis for Soil Properties in Different Zones

As indicated in Fig. 3, the box plots for soil fractions, pH, salinity, SOC, NO_3^--N, and AP contents in different zones of the study area showed different variation trends and significant differences. Soil particle size distribution is one of the most important physical attributes of soils. Although there were no significant differences (p>0.05) with reclaimed history increasing, there was a decreasing trend for clay/silt and an increasing trend for sand from the latest intertidal flats to the 1930s. Our results were consistent with previous studies and revealed that soil erosion occurred, and preferentially detaching and transporting clay and silt after long-term reclamation [31, 46]. In addition, reclamation might result in a higher percentage of macro-aggregate of coastal reclaimed

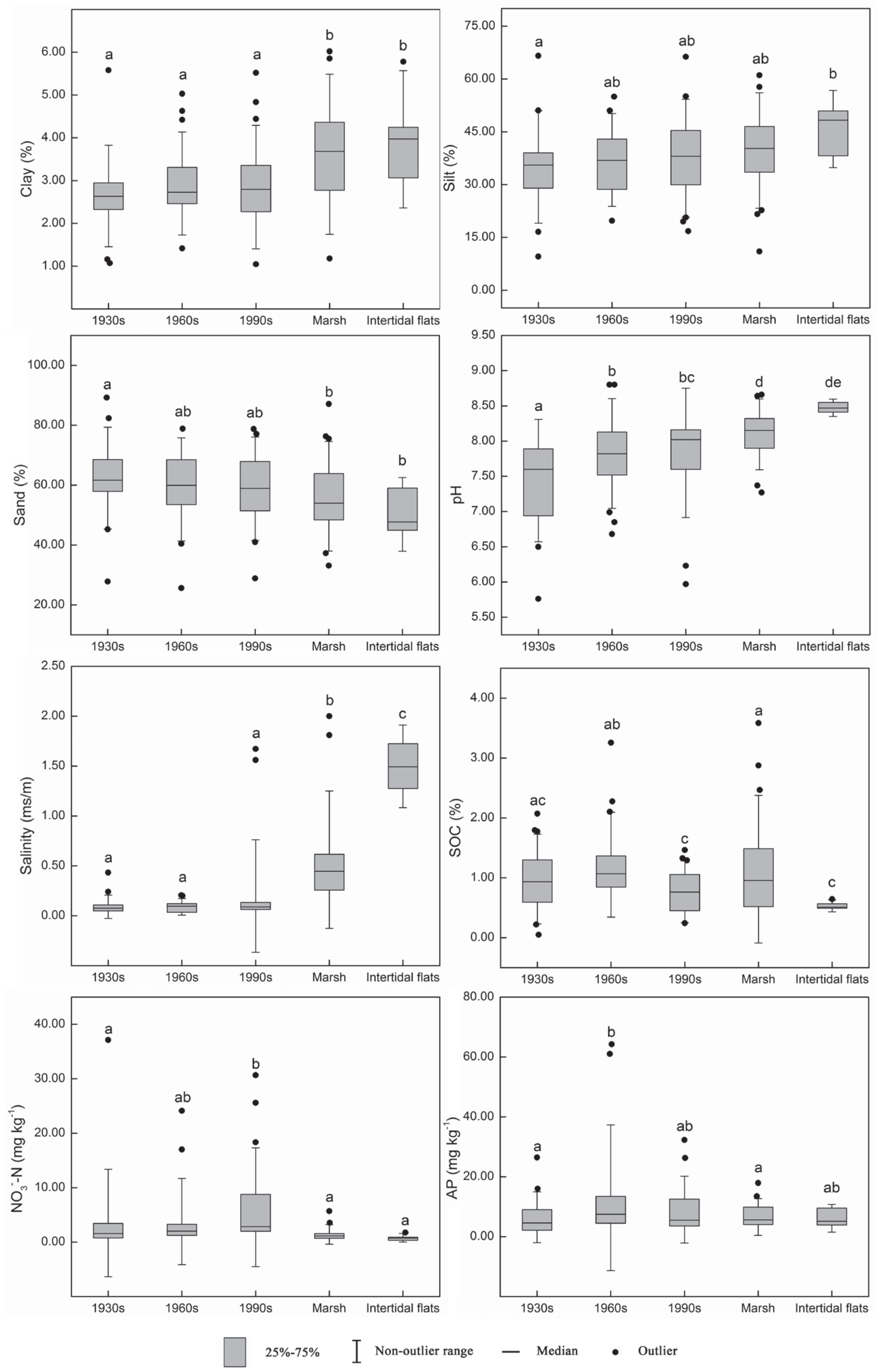

Fig. 3. Multiple comparison analysis of ANOVA and box plots for soil properties in different zones. Different letters marked for bars represent significant differences (p<0.05).

soils, especially for old zone 1930s that was closer to the residential zone. Previous studies have presented soil quality as possibly being improved by increasing vegetation coverage to prevent soil erosion and decrease the loss of fine particles, especially in the rainy seasons after the crops are harvested [47]. For soil pH, the median value displayed a slightly declining trend from 8.47 to 7.60 with the increase of reclamation time, but the ANOVA showed a significant difference from the intertidal flats to the 1930s soils (p<0.05).

Generally, soil pH values of > 7 indicated that weakly alkaline conditions at most sampling profiles that were supported by He et al. [48], who suggested that the primary soils were considerably influenced by the coastal deposits and marine water. However, a study in Hangzhou Bay [49] reported soil pH of 6.3 after 700 years of reclamation, implying that future soil acidification is possible for the study area if chemical fertilizers are overused. As expected, soil salinities decreased by 94.0% from the intertidal flats to the early reclamation period, and

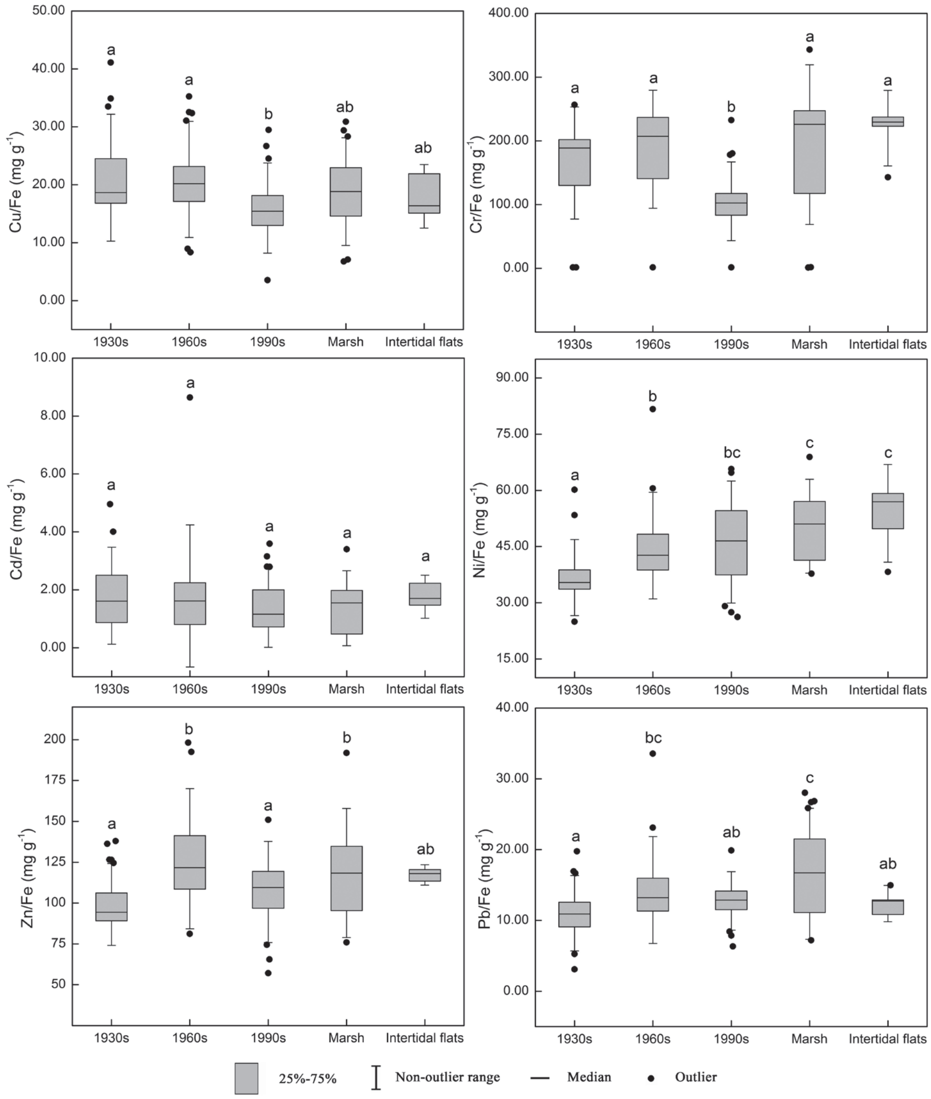

Fig. 4. Multiple comparison analysis of ANOVA and box plots for Fe-normalized heavy metal concentrations in different zones. Different letters marked for bars represent significant differences ($p<0.05$).

subsequently dropped slowly in the 1930s. The difference between the coastal wetlands and the reclaimed zones was significant ($p<0.05$), while no pronounced difference was observed for the three reclamation areas. The rapid decline in soil salinity was attributed to the effective leaching of exchangeable ions caused by dewatering after reclamation [50].

SOC, NO_3^--N, and AP contents are important indices for assessing soil nutrients. It was notable that SOC initially decreased during the early period of reclamation but then recovered to stable levels close to that of wetlands within 55 years, and the difference between the intertidal flats and the 1960s was significant ($p<0.05$). The temporal pattern of SOC was similar as shown by Cui et al. [31]. In the study area, large amounts of SOC were stored in soil covered by *Phragmites australis*, which might have been caused by the large inputs from higher plants, human activities (i.e., oil exploration), irrigation of paper wastewater, and also as consequence of the slow turnover rates of organic material under anaerobic conditions [51-52]. However, the SOC in surface soil was subject to rapid decomposition once exposed to air, suggesting that the outputs induced by mineralization were much larger than the inputs. Bai et al. [11] found that, on a regional basis, SOC loss through reclamation can be counteracted by accumulation in ditch marshes. Therefore, building some ditch in reclaimed wetlands can facilitate the retention of SOC loss posed by reclamation. For nitrate nitrogen and available P, the contents showed an initial increase, followed by a decrease over time, with 55 years being the turning point. Similar results were also reported in coastal reclaimed soils of Eastern China [8]. Overall, fertilizer application may also cause the soil nutrient contents to increase obviously, especially in the first 25 years of reclamation, while the tendency is to remain unchanged or even decrease after the 85 years in our study. Zhang and He [53] reported that the observed changes of soil nutrient over time were mainly attributed to the combined effect of pedogenetic processes and management factors. However, it is worth mentioning that it is necessary to investigate and monitor N and P in soils, especially in the estuaries of a semi-closed sea, because eutrophication occurs easily in these areas through the loss of soil N and P via drainage systems and soil erosion [54].

Fe-Normalized Heavy Metal Concentrations in Different Zones

Elevated heavy metal concentrations in soil and water have been found in many coastal areas around the world because of rapid development (of agriculture, industry, and transport infrastructure) on reclaimed land [55]. Heavy metal concentrations have been found to increase over time in reclaimed areas used for agriculture [15, 56], but not all of the heavy metals followed this trend in our study area. The Fe-normalized heavy metal concentrations and significant differences among them in different reclamation zones are shown in Fig. 4. For Cd, Cr, and Cu, there was a decreasing trend in the early period of reclamation (zone 1990s) and an increasing trend after 85 years of reclamation (the 1960s and 1930s zones). This result was inconsistent with a recent study conducted by Bai et al. [15], where the median values of normalized Cr in the 1990s zone were significantly lower than those in coastal wetlands, 1960s, and 1930s ($p<0.05$), but no significant difference was found among them. The same trend was also identified for normalized Cu and Cd, but the only significant difference for normalized Cu between the 1960s and the 1990s, and no significant difference was detected in those zones, which might be because of the disturbance of some outliers. It was observed that the value of normalized Ni tended to decline with the increase of reclamation ages, and significant differences ($p<0.05$) were found between wetlands and reclamation zones (1960s and 1930s). The same result was also reported in the Chongming Dongtan of the Yangtze River Estuary [14]. For Zn and Pb, it was clear that the values were in increasing trends from the early years of reclamation to the 1960s, while the contents of Zn and Pb significantly decreased in the 1930s ($p<0.05$). A recent study conducted by Wei et al. [57] revealed that the contents of Cd, Zn, and Pb were implicated with phosphorus fertilizer application. This conclusion proved the factor analysis in the study, and the ANOVA analysis for soil properties also indicated that the 1930s had the lower content of AP than other zones. This might be one of the reasons why the concentrations of Zn and Pb significantly decreased in the 1930s. Additionally, Du and Li et al. [58] showed that leaching irrigation promoted the movement of heavy metals from the surface soils to the deeper soils with reclaimed development, and the maximum reduction ratio was heavy metal Pb followed by Zn. Consequently, in spite of the effects of reclamation on soil properties and heavy metals of topsoil being conspicuous, further studies on soil properties and heavy metals from different soil layers will need to be performed.

In the 1990s zone, the concentrations of six Fe-normalized heavy metals were lower than those of natural wetlands, which suggested that the early reclamation significantly affected their distributions. In the study area, there were huge areas of saline soil caused by water evaporation in the latest reclaimed land. The majority of these were used for aquaculture and then abandoned. *Phragmites australis* grew widely on abandoned land and effectively improved soil quality by enriching nutrient materials, such as SOM. A large number of studies have reported that SOM has a strong capacity for binding metallic contaminants [59-60]. However, leaching and soil erosion related to agricultural cultivation can cause rapid desalinization, and most heavy metals accumulated in wetland soils will be lost after reclamation [61]. These processes can disrupt soil structure under natural conditions and increase the O_2 and CO_2 fluxes from the soil, ultimately resulting in organic matter and soluble metals being released [62]. Our results were consistent with the results just summarized. In the 1960s zone, metals other than Ni have been increasing since the 1990s, suggesting that these metals were being enriched in the older reclamation zone. Taking into consideration the fact that leaching processes had finished in the young reclamation zone, agricultural activities could be the main cause for the increasing Cd, Cr, Cu, Pb, and Zn concentrations in the old reclamation zone.

Since the founding of the People's Republic, large sums of water conservancy projects and farmland capital construction have been energetically carried out, and some state farms such as Qingshui Farm and Xinxing Farm have been established in the study area. It has been reported that higher rate of pesticides and fertilizers was the main source giving rise to the heavy metal enrichment in reclaimed soils [63]. Nevertheless, some studies on the reclamation history effect on concentrations of anthropogenic metals indicated that these metals transform slowly over time due to having their highest bioavailability [64], which might be one of the principal factors for distribution characteristics of heavy metals except for Ni, Zn, and Pb in the 1930s.

Assessing Heavy Metal Pollution and its Implications for Environmental Management

Heave metal pollution is an inorganic chemical hazard. With the rapid development of China's economy in the past 30 years, the environmental risk posed by heavy metal pollution has become increasingly significant [13]. In this study, the levels of soil heavy metal pollution were calculated using EF (Fig. 5) and inverse distance-weighted (IDW) interpolation was applied to show spatial distribution of pollution levels in different zones (Fig. 6). Previous studies often used Fe, Al, and Mn as reference metals to assess pollution levels [32]. As shown in Table 1, the CV value of iron was relatively low, indicating that anthropogenic activities had less impact on soil Fe concentrations, and thus Fe was used as the reference metal in this study. In addition, the corresponding background values for Cd, Cr, Cu, Fe, Ni, Pb, and Zn

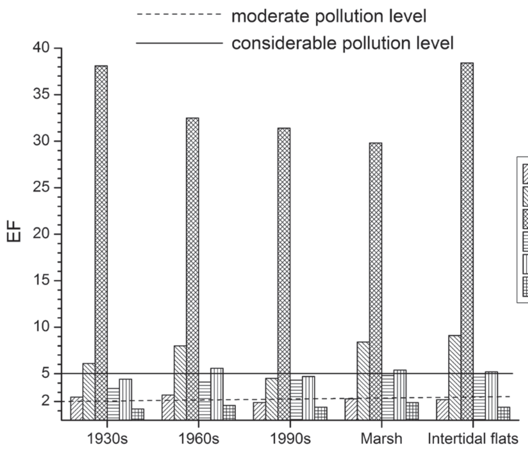

Fig. 5. The EFs of heavy metals in different zones.

were 0.11, 57.9, 19.8, 2400, 25.6, 21.4, and 54.2 (unit: mg kg^{-1}), respectively [65]. According to Liaghati et al. [66], heavy metal pollution can be divided into five levels: low (EF<2), moderate (2≤EF<5), considerable (5≤EF<20), high (20≤EF<40), and very high (EF≥40).

As shown in Figs 4 and 5, the mean EF values of Cu, Ni, and Pb were 2.3, 4.2, and 1.5, respectively, which indicates that the pollution levels for these heavy metals were relatively low and moderate. Among them, most EFs of Pb were less than 2, revealing that Pb contamination was not very severe in this study area and might be correlated with phasing out leaded gasoline by Chinese government, but in some hotspots closed to human settlement, the EF values of Pb were more than 2, and Pb contamination reached levels of moderate pollution. Previous studies have suggested that Pb poisoning for human beings can

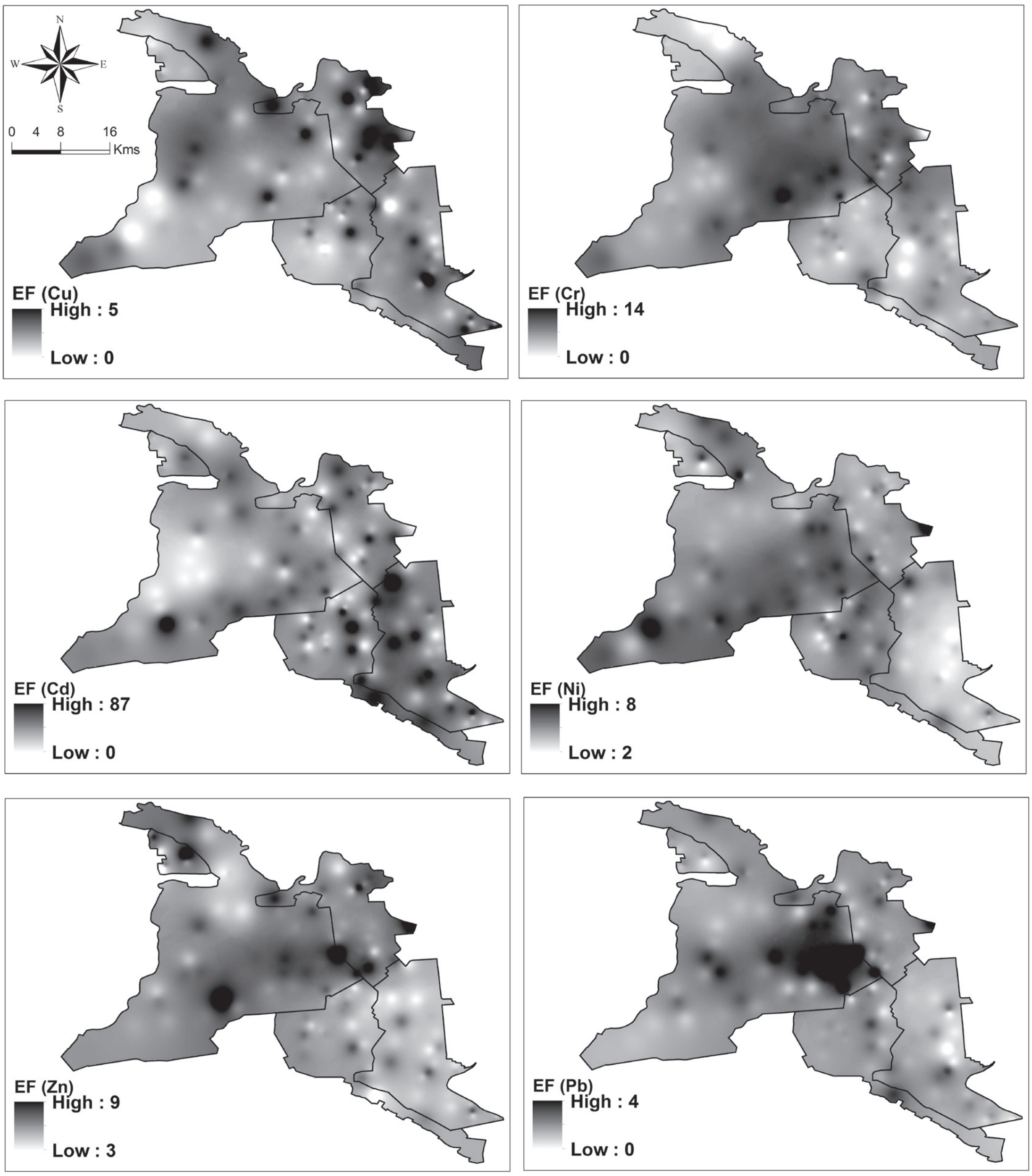

Fig. 6. The spatial distribution of heavy metal pollution in different zones.

cause hematological, gastrointestinal, and neurological dysfunctions [67]. A similar spatial distribution pattern was also found for Ni, with moderate or considerable pollution levels in natural wetlands and low pollution levels in the 1930s zone. The present study obtained similar findings posed by Ma et al. [14]. According to previous reports, the heavy metal pollution especially for Ni and Pb in soils might be caused by oilfield exploitation [68]. That could explain the basically similar patterns for the two metals due to the development of Liaohe Oilfield in the study area. However, it was worth noticing that the EF of Cd ranged between 29.8 and 38.4 in most areas of the study region, indicating high pollution level (20≤EF<40). This finding was in agreement with the result observed by Yang et al. [69], who reported Cd was the main pollutant in Liaodong Bay, adjoining this study area. Cadmium is considered one of the most mobile and strongest toxic heavy metals in the environment, and excess cadmium ions not only constrain plant growth, but also threaten human health through the food chain [70]. The spatial distribution of Cd showed that the hotspots were mostly located in the old reclamation zone and near the river mouths (of the Daling , Shuangtaizi, and Daliao), suggesting that riverine inputs caused by reclamation might be the most important source of Cd pollution, and that the pollution level had posed a serious environmental hazard. It is recommended that the local government pay enough attention to dealing with this serious issue. Cd was the most abundant pollutant in this study area and, furthermore, the EF of Cr (except for in the 1990s zone) was also relatively high, reaching considerable pollution levels. Moreover, for Zn contamination, apart from the old reclamation zone, the EFs of Zn in the young reclamation zones (1960s and 1930s) and natural wetlands were closer to or more than 5, suggesting that the Zn pollution level could also not be neglected. Previous studies have reported that chemical fertilizer, plastic film, and wastewater discharge from the industrial zone and rural-residential zone are major sources of Cu, Cr, and Zn pollution [71].

Generally, wetland ecosystems had higher pollution levels than did the other areas for all of the metals. In addition to the influence of deposition background, the anoxic condition might be the main environmental factor resulting in the heavy metal adsorption in wetlands. Because the wetland was generally near the river mouths, it has become an accumulative pool that gathers heavy metal pollutants resulting from petroleum hydrocarbons and upstream inputs of the Liaohe River, which may induce the wetlands an important secondary pollution source. Heavy metals in wetland soils may accumulate to toxic levels and have the potential to pose an environmental risk [72], which influences the sustainable development of wetland reclamation. Therefore, it is necessary for local governments to take measures to control heavy metal pollution, such as increasing ecological lands, developing ecological agriculture (e.g., rice-crab culture) and constructing natural reserves in key sites. Additionally, artificial ditches are a remarkable feature in the landscape of this study area, where reclamation always promotes the release of heavy metals from combination patterns in soils to aggregate to ditch and riparian wetlands. Wei et al. [57] has shown that ditch wetland serving as a temporary sink of heavy metals was found to pose the lowest dispersion risk of heavy metals. Periodic ditch dredging could effectively decrease the pollution input into the river and thereby reducing the threat to the regional water environment. These findings, observed by previous studies, are important implications for the environmental management of the study area. Although reclamation history is an important factor related to the changes of soil properties and heavy metal concentrations, admittedly these changes were also significantly influenced by land use intensity. Thus in order to better understand the impact of reclamation on soil function evolution processes, the effects of land use intensity on soil properties and heavy metal behaviors are required in future studies.

Conclusions

This study discusses the distributions of soil properties and heavy metal concentrations and associated environmental pollution levels in different zones. Results obtained from correlation analysis and factor analysis showed that soil particle size was a principal factor controlling the distribution of heavy metal concentrations. Using ANOVA, it was found that reclamation history could well reflect the evolution processes of soil properties and heavy metal concentrations. Soil erosion and leaching desalination had significant effects on the distributions of fine particle fractions, pH, salinity, SOC, and heavy metal contents in the early period of reclamation (1990s zone). With the time of land reclamation (1960s zone), persistent fertilizer application and enhanced human activities (such as papermaking, oil exploration, and urban expansion) had greatly increased the concentrations of soil nutrients and heavy metals. However, after 55 years of reclamation (1930s zone), the contents of soil properties and heavy metals basically remained unchanged or even decreased because they had their highest bioavailability and were influenced by the management factors. Due to the rapid development of the coastal economy in Liaohe River Delta, the environmental risk posed by heavy metal pollution has become increasingly significant. The enrichment factor (EF) and the inverse distance weighting (IDW) were used to show the distribution of soil heavy metal pollution.

Results showed that Cd pollution had reached high pollution levels, and the hotspots were mostly located in the old reclamation zone and near the river mouths. Taking measures to control heavy metal pollution, such as increasing ecological lands, developing ecological agriculture (e.g., rice-crab culture), constructing natural reserves in key sites, and periodic ditch dredging could effectively decrease pollution and thereby reduce the threat to the regional environment. For 2010 to 2020, the State Council of China has approved more than 200 km^2 of coastal wetlands to be reclaimed in Liaoning Province, and the study area will be viewed as one of the key areas for development. Severe environmental problems may occur in the study area. Therefore, researching the effects of land reclamation on soil environment evolution can provide valuable information for the sustainable development and management of the coastal zone.

Acknowledgements

This research is supported by the Special Foundation for the State Major Basic Research Program of China (Nos. 2013FY111800 and 2013FY111100-02). We thank Mr. Zhong for his hard work during the field investigation.

References

1. SHEPARD C.C., CRAIN C.M., BECK M.W. The protective role of coastal marshes: A systematic review and meta-analysis. PloS one **6**, e27374, **2011**.
2. WANG W., LIU H., LI Y.Q., SU J.L. Development and management of land reclamation in China. Ocean Coast. Manage. **102**, 415, **2014**.
3. SUZUKI T. Economic and geographic backgrounds of land reclamation in Japanese ports. Mar. Pollut. Bull. **47**, 226, **2003**.
4. DIPSON P.T., CHITHRA S.V., AMARNATH A., SMITHA S.V., HARINDRANATHAN NAIR M.V., SHAHIN A. Spatial changes of estuary in Ernakulam district, Southern India for last seven decades, using multi-temporal satellite data. J. Environ. Manage. **148**, 134, **2015**.
5. AN S.Q., LI H.B., GUAN B.H., ZHOU C.F., WANG Z.S., DENG Z.F., ZHI Y.B., LIU Y.H., XU C., FANG S.B., JIANG J.H., LI H.L. China's Natural Wetlands: Past Problems, Current Status, and Future Challenges. Ambio **36**, 335, **2007**.
6. CUI B.S., HE Q., GU B.H., BAI J.H., LIU X.H. China's coastal wetlands: Understanding environmental changes and human impacts for management and conservation. Wetlands **36**, 1, **2016**.
7. PORTNOY J.W., GIBLIN A.E. Effects of historic tidal restrictions on salt marsh sediment chemistry. Biogeochemistry **36**, 275, **1997**.
8. SUN Y.G., LI X.Z., MANDER Ü., HE Y.L. Effect of reclamation time and land use on soil properties in Changjiang River Estuary, China. Chin. Geogra. Sci. **21**, 403, **2011**.
9. LI X.Z., SUN Y.G., MANDER Ü., HE Y.L. Effects of land use intensity on soil nutrient distribution after reclamation in an estuary landscape. Landscape Ecol **28**, 699, **2012**.
10. BRUCE J.P., FROME M., HAITES E., JANZEN H., LAL R., PAUSTIAN K. Carbon sequestration in soils. J. Soil Water Conserv. **54**, 382, **1999**.
11. BAI J.H., XIAO R., ZHANG K.J., GAO H.F., CUI B.S., LIU X.H. Soil organic carbon as affected by land use in young and old reclaimed regions of a coastal estuary wetland, China. Soil Use Manage. **29**, 57, **2013**.
12. SHEORAN A.S., SHEORAN V. Heavy metal removal mechanism of acid mine drainage in wetlands: A critical review. Miner. Eng. **19**, 105, **2006**.
13. LI J.G., PU L.J., LIAO Q.L., ZHU M., DAI X.Q., XU Y., ZHANG L.F., HUA M., JIN Y. How anthropogenic activities affect soil heavy metal concentration on a broad scale: A geochemistry survey in Yangtze River Delta, Eastern China. Environ. Earth Sci. **73**, 1823, **2015**.
14. MA C., ZHENG R., ZHAO J.L., HAN X.M., WANG L., GAO X.J., ZHANG C. S. Relationships between heavy metal concentrations in soils and reclamation history in the reclaimed coastal area of Chongming Dongtan of the Yangtze River Estuary, China. J. Soil Sediment **15**, 139, **2015**.
15. BAI J.H., XIAO R., CUI B.S., ZHANG K.J., WANG Q.G., LIU X.H., GAO H.F., HUANG L. B. Assessment of heavy metal pollution in wetland soils from the young and old reclaimed regions in the Pearl River Estuary, South China. Environ. Pollut. **159**, 817, **2011**.
16. LI X.R., HE M.Z., DUAN Z.H., XIAO H.L., JIA X.H. Recovery of topsoil physicochemical properties in revegetated sites in the sand-burial ecosystems of the Tengger Desert, northern China. Geomorphology **88**, 254, **2007**.
17. HU J.L., LIN X.G., YIN R., CHU H.Y., WANG J.H., ZHANG H.Y., CAO Z.H. Comparison of fertility characteristics in paddy soils of different ages in Cixi, Zhejiang. Plant Nutr. Fertil. Sci. **14**, 673, **2008**.
18. YANG X.L., YUAN X.T., ZHANG A.G., MAO Y.Z., LI Q., ZONG H.M., WANG L.J., LI X. D. Spatial distribution and sources of heavy metals and petroleum hydrocarbon in the sand flats of Shuangtaizi Estuary, Bohai Sea of China. Mar. Pollut. Bull. **95**, 503, **2015**.
19. ZHANG Z.S., SONG X.L., LU X.G., XUE, Z.S. Ecological stoichiometry of carbon, nitrogen, and phosphorus in estuarine wetland soils: Influences of vegetation coverage, plant communities, geomorphology, and seawalls. J. Soils Sediments. **13**, 1043, **2013**.
20. LI G.L., LANG Y.H., YANG W., PENG P., WANG X.M. Source contributions of PAHs and toxicity in reed wetland soils of Liaohe estuary using a CMB-TEQ method. Sci. Total Environ. **490**, 199, **2014**.
21. LI X.W., LIANG C., SHI J.B. Developing Wetland Restoration Scenarios and Modeling Its Ecological Consequences in the Liaohe River Delta Wetlands, China. CLEAN - Soil, Air, Water **40**, 1185, **2012**.
22. ELLIS S., ATHERTON J.K. Properties and development of soils on reclaimed alluvial sediments of the Humber estuary, eastern England. Catena **52**, 129, **2003**.
23. BUAT-MENARD P., CHESSELET R. Variable influence of the atmospheric flux on the trace metal chemistry of oceanic suspended matter. Earth Planet. Sci. Lett. **42**, 399, **1979**.
24. WEI J., WEI O.Y., HAO F.H., LIU B., WANG F.L. Geochemical variability of heavy metals in soil after land use conversions in Northeast China and its environmental applications. Environ. Sci. Proc. Impacts **16**, 924, **2014**.
25. TURER D., MAYNARD J.B., SANSALONE J.J. Heavy metal contamination in soils of urban highways comparison between runoff and soil concentrations at Cincinnati, Ohio. Water, Air, Soil Pollut. **132**, 293, **2001**.
26. SPARKS D.L. Environmental soil chemistry. Academic Press: **2003**.
27. WONG V.N.L., DALAL R.C., GREENE R.S.B. Salinity and sodicity effects on respiration and microbial biomass of soil. Biol. Fert. Soils **44**, 943, **2008**.
28. POST W.M., KWON K.C. Soil carbon sequestration and land-use change: Processes and potential. Global Change Biol. **6**, 317, **2000**.
29. LI D.F., SHAO M.A. Soil organic carbon and influencing factors in different landscapes in an arid region of northwestern China. Catena **116**, 95, **2014**.
30. BARITZ R., SEUFERT G., MONTANARELLA L., RANST E.V. Carbon concentrations and stocks in forest soils of Europe. Forest Ecol. Manag. **260**, 262, **2010**.

31. CUI J., LIU C., LI Z.L., WANG L., CHEN X.F., YE Z.Z., FANG C.M. Long-term changes in topsoil chemical properties under centuries of cultivation after reclamation of coastal wetlands in the Yangtze Estuary, China. Soil Till. Res. **123**, 50, **2012**.
32. ZHANG H., WANG Z.F., ZHANG Y.L., HU Z.J. The effects of the Qinghai-Tibet railway on heavy metals enrichment in soils. Sci. Total Environ. **439**, 240, **2012**.
33. LAING G.D., RINKLEBE J., VANDECASTEELE B., MEERS E., TACK F.M.G. Trace metal behaviour in estuarine and riverine floodplain soils and sediments: A review. Sci. Total Environ. **407**, 3972, **2009**.
34. ZHANG W., YU L., HUTCHINSON S.M., XU S., CHEN Z., GAO X. China's Yangtze Estuary: I. Geomorphic influence on heavy metal accumulation in intertidal sediments. Geomorphology **41**, 195, **2001**.
35. DUAN D.D., RAN Y., CHENG H.F., CHEN J.A., WAN G. Contamination trends of trace metals and coupling with algal productivity in sediment cores in Pearl River Delta, South China. Chemosphere **103**, 35, **2014**.
36. SCHIFF K.C., WEISBERG S.B. Iron as a reference element for determining trace metal enrichment in Southern California coastal shelf sediments. Mar. Environ. Res. **48**, 161, **1999**.
37. ZHOU H.Y., PENG X.T., PAN J.M. Distribution, source and enrichment of some chemical elements in sediments of the Pearl River Estuary, China. Cont. Shelf Res. **24**, 1857, **2004**.
38. MICO C., RECATALA L., PERIS M., SANCHEZ J. Assessing heavy metal sources in agricultural soils of an European Mediterranean area by multivariate analysis. Chemosphere **65**, 863, **2006**.
39. QIAN J., SHAN X.Q., WANG Z.J., TU Q. Distribution and plant availability of heavy metals in different particle-size fractions of soil. Sci. Total Environ. **187**, 131, **1996**.
40. ROSENBERG O., PERSSON T., HÖGBOM L., JACOBSON S. Effects of wood-ash application on potential carbon and nitrogen mineralisation at two forest sites with different tree species, climate and N status. Forest Ecol. Manag. **260**, 511, **2010**.
41. MLADENOFF D.J. Dynamics of nitrogen mineralization and nitrification in hemlock and hardwood treefall gaps. Ecology **68**, 1171, **1987**.
42. SAHRAWAT K.L. Mineralization of soil organic nitrogen under waterlogged conditions in relation to other properties of tropical rice soils. Soil Res. **21**, 133, **1983**.
43. HAN Y.M., DU P.X., CAO J.J., POSMENTIER E.S. Multivariate analysis of heavy metal contamination in urban dusts of Xi'an, Central China. Sci. Total Environ. **355**, 176, **2006**.
44. TANG W.Z., SHAN B.Q., ZHANG H., MAO Z.P. Heavy metal sources and associated risk in response to agricultural intensification in the estuarine sediments of Chaohu Lake Valley, East China. J. Hazard. Mater. **176**, 945, **2010**.
45. SHAN Y.S., TYSKLIND M., HAO F.F., WEI O.Y., CHEN S.Y., LIN C.Y. Identification of sources of heavy metals in agricultural soils using multivariate analysis and GIS. J. Soil Sediment **13**, 720, **2013**.
46. QUINTON J.N., CATT J.A., HESS T.M. The selective removal of phosphorus from soil: Is event size important? J. Environ. Qual. **30**, 538, **2001**.
47. YAN Y.C., XIN X.P., XU X.L., WANG X., YANG G.X., YAN R.R., CHEN B.R. Quantitative effects of wind erosion on the soil texture and soil nutrients under different vegetation coverage in a semiarid steppe of northern China. Plant Soil **369**, 585, **2013**.
48. HE X.Q., GU C.J. Study on reclamation and sustainable development of Chongming wetland. Ter. Nat. Resour. Stud. **4**, 39, **2003** [In Chinese].
49. CHEN L.M., ZHANG G.L. Parent material uniformity and evolution of soil characteristics of a paddy soil chronosequence derived from marine sediments. Acta Pedol. Sin. **46**, 753, **2009** [In Chinese].
50. WANG Y.D., WANG Z.L., FENG X.P., GUO C.C., CHEN Q. Long-term effect of agricultural reclamation on soil chemical properties of a coastal saline marsh in Bohai Rim, Northern China. PloS One, **9**, e93727, **2014**.
51. BATIONO A., KIHARA J., VANLAUWE B., WASWA B., KIMETU J. Soil organic carbon dynamics, functions and management in West African agro-ecosystems. Agric. Syst. **94**, 13, **2007**.
52. LIN T., YE S. Y., MA C.L., DING X.G., BRIX H., YUAN H.M., CHEN Y.J., GUO Z.G. Sources and preservation of organic matter in soils of the wetlands in the Liaohe (Liao River) Delta, North China. Mar. Pollut. Bull. **71**, 276, **2013**.
53. ZHANG M.K., HE Z.L. Long-term changes in organic carbon and nutrients of an ultisol under rice cropping in southeast China. Geoderma **118**, 167, **2004**.
54. BOESCH D.F. Challenges and opportunities for science in reducing nutrient over-enrichment of coastal ecosystems. Estuar. Coast **25**, 886, **2002**.
55. ACOSTA J.A., FAZ A., MARTÍNEZ-MARTÍNEZ S., AROCENA J.M. Enrichment of metals in soils subjected to different land uses in a typical Mediterranean environment (Murcia City, southeast Spain). Appl. Geochem. **26**, 405, **2011**.
56. GEIGER G., FEDERER P., STICHER H. Reclamation of heavy metal-contaminated soils: Field studies and germination experiments. J. Environ. Qual. **22**, 201, **1993**.
57. WEI J., WEI O.Y., HAO F.H., WANG F.L., LIU B. Long-term cultivation impact on the heavy metal behavior in a reclaimed wetland, Northeast China. J. Soil Sediment **14**, 1, **2014**.
58. LI Q.S., LIU Y.N., DU Y.F., CUI Z.H., SHI L., WANG L.L., LI H.J. The behavior of heavy metals in tidal flat sediments during fresh water leaching. Chemosphere **82**, 834, **2011**.
59. YANG S.L., ZHOU D.Q., YU H.Y., WEI R., PAN B. Distribution and speciation of metals (Cu, Zn, Cd, and Pb) in agricultural and non-agricultural soils near a stream upriver from the Pearl River, China. Environ. Pollut. **177C**, 64, **2013**.
60. NIAZI N.K., SINGH B., MINASNY B. Mid-infrared spectroscopy and partial least-squares regression to estimate soil arsenic at a highly variable arsenic-contaminated site. Int. J. Environ. Sci. Te. **12**, 1735, **2014**.
61. OVERESCH M., RINKLEBE J., BROLL G., NEUE H.U. Metals and arsenic in soils and corresponding vegetation at Central Elbe river floodplains (Germany). Environ. Pollut. **145**, 800, **2007**.
62. MOHAMED I., AHAMADOU B., LI M., GONG C.X., CAI P., LIANG W., HUANG Q.Y. Fractionation of copper and cadmium and their binding with soil organic matter in a contaminated soil amended with organic materials. J. Soil Sediment **10**, 973, **2010**.
63. SHOMAR B.H. Trace elements in major solid-pesticides used in the Gaza Strip. Chemosphere **65**, 898, **2006**.
64. JALALI M., KHANLARI Z.V. Effect of aging process on the fractionation of heavy metals in some calcareous soils of Iran. Geoderma **143**, 26, **2008**.
65. WU Y.Y., LI J., WANG X. Soil element background values of Liaoning Province in China. China Environmental Science Press: Beijing, China, 1, **1994**.

66. LIAGHATI T., PREDA M., COX M. Heavy metal distribution and controlling factors within coastal plain sediments, Bells Creek catchment, southeast Queensland, Australia. Environ. Int. **29**, 935, **2004**.
67. GILLIAN L. Perspectives on lead toxicity. Clin.Biochem. **26**, 371, **1993**.
68. OSUJI L.C., ONOJAKE C.M. Trace heavy metals associated with crude oil: A case study of Ebocha-8 oil-spill-polluted site in Niger Delta, Nigeria. Chem. Biodivers. **1**, 1708, **2004**.
69. YANG X.L,, YUAN X.T., ZHANG A.G., MAO Y.Z., LI Q., ZONG H.M., WANG L.J., LI X. D. Spatial distribution and sources of heavy metals and petroleum hydrocarbon in the sand flats of Shuangtaizi Estuary, Bohai Sea of China. Mar. Pollut. Bull. **95**, 503, **2015**.
70. JARUP L. Hazards of heavy metal contamination. Brit. Med. Bull. **68**, 167, **2003**.
71. LIU E.F., BIRCH G.F., SHEN J., YUAN H.Z., ZHANG E.L., CAO Y.M. Comprehensive evaluation of heavy metal contamination in surface and core sediments of Taihu Lake, the third largest freshwater lake in China. Environ. Earth Sci. **67**, 39, **2012**.
72. PERKINS S.M., FILIPPELLI G.M., SOUCH C.J. Airborne trace metal contamination of wetland sediments at Indiana Dunes National Lakeshore. Water Air Soil Poll. **122**, 231, **2000**.