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# The impacts of land reclamation on the accumulation of key elements in wetland ecosystems in the Sanjiang Plain, northeast China☆

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ABSTRACT

The Sanjiang Plain, which is located in northeastern China, given the distribution of temperate freshwater wetlands there and this region has considerable significance in ensuring food security in China. Two periods of farmland reclamation that occurred during the last 100 years led to the loss of nearly 80% of the area of the native wetlands, and the development of agriculture has also increased the potential environmental risks to the residual wetlands. To evaluate the effects of farmland reclamation on the accumulation of key elements within the residual wetland ecosystems, six wetland profiles in the Sanjiang Plain are selected in this study. Using age-depth models and the concentrations of key elements, the historical accumulation rates (ARs) of carbon (C), nutrient elements (N and P) and potentially toxic elements (Hg, As, Pb, Cu, and Zn) over the last 150 years are reconstructed. The results show that the ARs of the potentially toxic elements in two of the wetland profiles begin to increase during the first reclamation period (AD 1900–1930). The ARs of both of the key elements clearly increase in all of the wetland profiles during the second reclamation period (AD 1950–1980). After land reclamation had ceased, increases in population and the development of industry became major factors that caused the potential environmental risks to wetlands to continue to increase from AD 1980 to the present. During the last 100 years, reclamation has increased the potential environmental risks and has led to the storage of additional carbon in the residual wetlands of the Sanjiang Plain.



## 1. Introduction

Wetlands, which provide critical habitats for many species, are widely distributed and cover 5–8% of the land surface of the Earth (Mitsch and Gosselink, 2007; Rydin and Jeglum, 2013). Because of the nutrient-rich soils and flat topography, freshwater wetlands are frequently reclaimed for food production and have been strongly influenced by human activities during the last 150 years (Thiere et al., 2009; Van Dyke and Wasson, 2005). The Sanjiang Plain, which is located in northeastern China, is one of the most important regions of temperate freshwater wetlands in the world (Brinson and Malvárez, 2002); it is also one of the most productive agricultural regions in China and produces large amounts of rice and soybeans (Liu et al., 2010). From the end of the 19th century to the present, the area of farmland in the Sanjiang Plain increased by more than 300 times (from ca. 130 $km^2$ before AD 1900 to 36,533 $km^2$ in AD 1996) (Group, 1998). Not surprisingly, this increase in farmland area occurred through the reclamation of native grasslands, wetlands and other ecosystems (Zhang et al., 2010). From the 1950s to the present, the area of wetlands decreased from 35,270 $km^2$ to 8100 $km^2$, and nearly 91% of the lost wetlands were converted to cultivated areas; small areas of wetlands remain in several nature reserves (Wang et al., 2011; Song et al., 2014). Despite the significant loss of the native wetlands, the Sanjiang Plain is still one of the most important regions in China for breeding, nesting and migratory birds, given the wetlands that remain (Kamiya and Ozaki, 2002; Xue et al., 2008). Additionally, the residual wetlands in the Sanjiang Plain provide other important ecosystem services (e.g., sequestering carbon, attenuating floodwater, and maintaining biodiversity) and are important for the

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ecological security of the region (Bao et al., 2011; Liu, 2007; Lu and Liu, 2008). However, the rapid development of agriculture and the economy has resulted in increases in the potential environmental risks to the residual wetland ecosystems and even to the ecological security of the region (Zhou et al., 2009).

Two periods of reclamation have occurred during the last 100 years (i.e., AD 1900–1930 and AD 1950–1980), and the area of farmland devoted to rice cultivation has clearly increased in the Sanjiang Plain (Group, 1998; Zhang et al., 2006). Slash-and-burn practices are widely followed in the Sanjiang Plain for farmland reclamation; the frequency and intensity of fire increased considerably during these two farmland reclamation periods (Gao et al., 2014a). The high frequency of biomass burning released potentially toxic elements (e.g., Hg and Pb) and was a key source of these potentially toxic elements in these periods (Cordeiro et al., 2002). Because pesticides and phosphorus fertilizers contain varying amounts of heavy metals and other rare earth elements as contaminants, the increasing consumption of pesticides and fertilizers has also led to the transfer of increased amounts of potentially toxic elements (e.g., Pb, As, and Hg) and nutrient elements (e.g., N and P) into the surrounding natural ecosystems (Braskerud, 2002; Jiao et al., 2015; Novotny, 1999). The development of agriculture in the Sanjiang Plain occurred at the same time as increases in population and the development of the regional economy (Statistics Office of Heilongjiang Province, 2012). In addition to the accumulation of greater amounts of potentially toxic elements from anthropogenic sources in the natural ecosystems (Gao et al., 2014b; Gu et al., 2014; Singh et al., 2005), the development of agriculture and the increases in population have also increased the supply of water and decreased the water table in the wetlands. These developments have caused the environmental risks to the surrounding native ecosystems and the residents living nearby to increase (Brinson and Malvárez, 2002; Connor et al., 2012; Johnson et al., 2001).

Because slow rates of decomposition occur under anaerobic conditions, combined with the continuous depositional inputs that occur in wetland ecosystems (Martini et al., 2007), wetland sedimentary records are widely used to reconstruct the historical impacts of regional climate changes and human activities on natural ecosystems (Drexler et al., 2016; Wang et al., 2004b). Potentially toxic elements are often considered to be conservative pollutants in wetlands because they are inert in the sedimentary environment (Olivares-Rieumont et al., 2005). Large amounts of pollutants from anthropogenic sources have accumulated in the eastern part of the Sanjiang Plain; the accumulation of these pollutants finally resulted in clear increases in the historical ARs of Pb and Cu beginning in approximately AD 1950 (Gao et al., 2014b; Liu et al., 2018). In addition to Cu and Pb, Hg and As are two other important potentially toxic elements that are produced by human activities and accumulate in natural ecosystems (Daga et al., 2016). In addition to potentially toxic elements, the large amounts of fertilizer that are used to increase agricultural productivity have also caused increased amounts of nutrient elements (e.g., N and P) to accumulate in the surrounding wetland ecosystems; these elements influence the biogeochemical processes (e.g., the carbon cycle) that occur within wetland ecosystems (Cabezas et al., 2014). However, few studies have evaluated the impacts of farmland reclamation on the historical ARs of these elements in the Sanjiang Plain using wetland sedimentary records.

Consequently, the goal of this study is to evaluate the impact of farmland reclamation on the surrounding wetland ecosystem in the Sanjiang Plain during the last 100 years. To achieve this goal, we summarize the ARs of Cu, Pb and Zn reported by previous studies (Gao et al., 2014b; Liu et al., 2018); additionally, the potentially toxic and nutrient elements (such as Hg, As, N and P) in six wetland profiles in the Sanjiang Plain are analyzed in this study. Based on age-depth models constructed using $^{210}$Pb data, the historical ARs of the potentially toxic elements and nutrient elements in the Sanjiang Plain during the last 150 years are reconstructed. Based on principal component analysis (PCA) and heatmap analysis, the impacts of historical reclamation on wetland ecosystems at different sites and in different periods are assessed, and the influence of land reclamation on the wetland ecosystems during the last 100 years is evaluated.

## 2. Materials and methods

### 2.1. Sites and sampling

The Sanjiang Plain is a low and flat alluvial plain that is crossed by the Heilong River, the Songhua River and the Wusuli River. It experienced an annual mean temperature of 1.3–4.3 °C and received 400–700 mm of precipitation per year, on average, between the 1950s and the 2000s (Fig. 1). The Heilong River and the Wusuli River make up part of the boundary between China and Russia. The southern origin of the Wusuli River is Xingkai Lake, and the Wusuli River flows 492 km from south to north and merges into the Heilong River. Because of the special topographic and climate conditions in this area, a large number of wetland patches exist there. Six wetlands located within the Sanjiang Plain are selected in this study, and the locations of important cities in this region are also shown in Fig. 1. Detailed information on these six wetlands have been provided by previous studies and are summarized in Table 1 (Gao et al., 2014b; Liu et al., 2018). Briefly, the Heixiazi (HXZ; 134°44′ E, 48°19′ N) wetland is located in a delta region where the Heilong and Wusuli Rivers converge, and the Qindeli (QDL; 133°20′ E, 48°9′ N) wetland is located near the HXZ wetland and upstream along the Heilong River. The Dongfanghong (DFH; 133°37′ E, 46°23′ N) wetland, the Zhenbaodao (ZBD; 133°45′ E, 46°13′ N) wetland and the Zhuaji (ZJ; 134°37′ E, 48°19′ N) wetland are located along the Wusuli River. The Shenjiadian (SJD; 130°39′ E, 46°34′ N) wetland is a typical valley peatland that is located in Huachuan County, approximately 34 km from the major city (i.e. Jiamusi). Samples were collected using a Russian peat corer or taken from a profile in the central portion of each wetland, and the length of each core is over 50 cm. The profiles were cut into contiguous 1-cm samples in the field using a stainless-steel knife, and the samples were sealed in tagged plastic bags for transport to the laboratory and refrigerated at −20 °C for further analysis.

### 2.2. Chronology

The reconstruction of wetland sedimentation over the last 150 years is frequently based on $^{210}$Pb data (Turetsky et al., 2004). We assume constant sedimentation in the wetland profiles; the constant rate of supply (CRS) model is frequently used to generate age-depth models (Binford, 1990). Samples collected at 1-cm intervals are analyzed for $^{210}$Pb by measuring the gamma-ray emissions from the samples using a highly purified germanium semiconductor and a low-background gamma spectrometer (OTEC Instruments Ltd., USA). More than thirty samples in each wetland profile were selected for geochronological analyses, which were performed at the State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Science. The dry bulk density was determined by oven drying at 105 °C for 12 h (He et al., 2017). The wetland sedimentation rates are calculated from the age-depth models and the dry bulk density at each depth within the profiles. Based on the history of farmland reclamation, four periods (i.e., AD 1900–1930, AD 1930–1950, AD

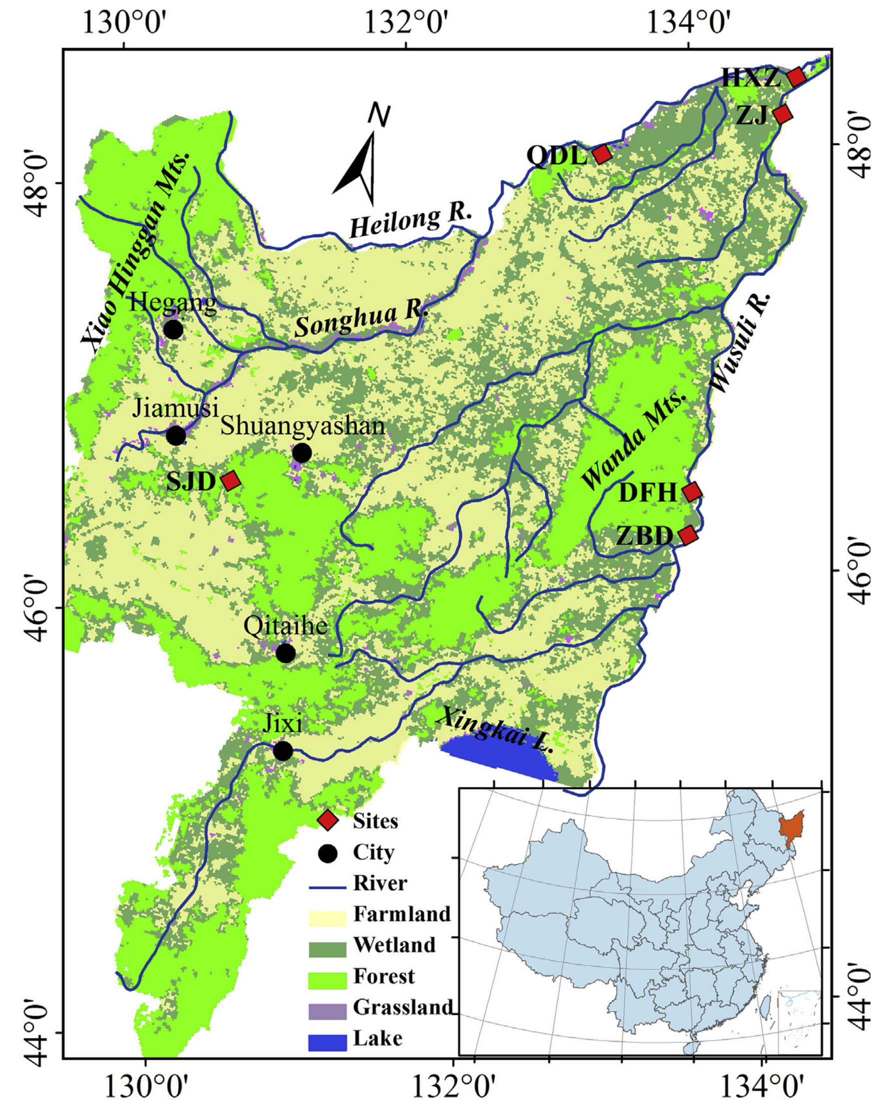


**Fig. 1.** Locations of wetland profiles in the Sanjiang Plain; the landscape in the Sanjiang Plain in AD 2000 is also shown. Landscape of Sanjiang Plain from 1954 to 2000 could be found in Wang et al. (2011). The landscape dataset was provided by the Environmental and Ecological Science Data Center for West China, National Natural Science Foundation of China (http://westdc.westgis.ac.cn).

**Table 1**
Information of sampling sites in this study.

| Name | Location | Plant communities | Depth (dating depth[a]) | Soil types | Reference |
|---|---|---|---|---|---|
| Zhenbaodao (ZBD) | 133°45′ E, 46°13′ N | Deyeuxia angustifolia/ Carex appendiculata | 50 (23) | Sediment | This study |
| Zhuaji (ZJ) | 134°37′ E, 48°19′ N | Deyeuxia angustifolia | 50 (26) | Sediment | Gao et al., 2014b |
| Dongfanghong (DFH) | 133°37′ E, 46°23′ N | Carex lasiocarpa/ Carex pseudocuraica | 50 (28) | Sediment | Gao et al., 2014b |
| Heixiazi (HXZ) | 134°44′ E, 48°19′ N | Deyeuxia angustifolia | 50 (27) | Sediment | Liu et al., 2018 |
| Qindeli (QDL) | 133°20′ E, 48°9′ N | Carex lasiocarpa | 50 (16) | Peat | Liu et al., 2018 |
| Shenjiadian (SJD) | 130°39′ E, 46°34′ N | Betula fruticosae/ Calamagrostis angustifolia | 50 (33) | Peat | Liu et al., 2018 |

[a] Dating depth was obtained from CRS model and $^{210}Pb$ data.

1950–1980, and after AD 1980) are identified and used to evaluate the ARs of the key elements in the wetland ecosystems of the Sanjiang Plain during the different farmland reclamation periods.

### *2.3. Elemental analyses*

The samples were milled and completely digested in a concentrated mixture of $HNO_3$, $HClO_4$ and HF, which is the same procedure used for the elemental analyses (Bai et al., 2011). The contents of Pb, Zn, Cu, P, and Al were determined using an inductively coupled plasma atomic emission spectroscope (ICPS-7500). The contents of Hg and As were measured in freeze-dried samples after digestion in a mixture of $HNO_3$ and $H_2SO_4$ using a Leco AMA-254 thermal decomposition-atomic absorption spectrometer. The recovery rates of the elements, as determined using State Standard Reference Material GBW07401, are approximately 93–109%. The total nitrogen (N) in the soils was measured after digestion in concentrated $H_2SO_4$ and a catalyst using a continuous flow chemistry analyzer. The carbon (C) contents were determined using LOI (loss-on-ignition), in which the samples were combusted at 550 °C for 4 h; the organic matter content is multiplied by 0.5 (Heiri et al., 2001). All of the samples were analyzed at the Analysis and Test Center of the Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences.

### 2.4. Statistical methods

#### 2.4.1. Enrichment factors

The variations in the elemental concentrations with depth within the profiles are studied to assess the environmental oscillations and the degree of anthropogenic influence. For these purposes, the enrichment factor (EF) of a particular element Me is calculated as:

$$EF= ([Me]/ [Al])_{samples}/ ([Me]/ [Al])_{reference}$$

where [Me] is the concentration of the trace element in question (i.e., Pb, Cu, Zn, Hg, or As). In the calculation of the EFs of particular elements, Al is commonly used as the conserved element because the Al in soil is resistant to chemical weathering (Wang et al., 2004a). Therefore, the EFs of the trace elements are calculated by normalizing the concentrations of Al in this study. The normalization of data involves a reference (a baseline); it does not influence the historical trend of the [Me]/[Al] ratios in the samples. Because it is difficult to identify a period without anthropogenic effects, the minimum values of the ratios $([Me]/[Al])_{lithogenic}$ (e.g., Me = Pb) in all of the wetland profiles are taken to represent the pre-anthropogenic inorganic geochemical background to clarify the historical trends in the [Me]/[Al] ratios in the samples and to increase their comparability (Gao et al., 2014b).

#### 2.4.2. Principal component analysis

Principal component analysis (PCA) is used to identify the associations among multiple variables and to identify the processes that may control the distributions of elements (Biester et al., 2012; Gao et al., 2014c). For these purposes, PCA is applied to the ARs of typical elements (i.e., C, N, P, Cu, Pb, Zn, Hg, and As) at all of the sites during the last 150 years. The ARs of the typical elements at each depth in all of the wetlands are regarded as variables in each sample. The Z-score transformation is applied to the data to reduce all of the variables to a similar range of variation and to avoid scaling effects while not changing the original trends in the data. All of the statistical computations are carried out in the R statistical computing environment. PCA is implemented using the “princomp” function within the “stats” package (Team, 2015).

#### 2.4.3. Heatmap analysis

Cluster analysis is used to classify the objects in a system into categories or clusters based on their nearness or similarity, and dendrograms are used to show the results of cluster analyses (Varol et al., 2012). Heatmap analysis displays a false-color image with two dendrograms for two different objects and can divide these two objects into several groups. The different influence factors in these two objects are reordered according to their nearness or similarity by the cluster analysis. In this study, heatmap analysis is used to evaluate the spatial and temporal effects of human activities on the wetlands in the Sanjiang Plain. To evaluate the spatial and temporal variations in the accumulation of key elements in the wetlands in the Sanjiang Plain, average PC1 scores predicted using a spline model with a 1-year interval and the average values of the PC1 scores every ten years are regarded as reflecting the effects of human activities on the wetlands and are used in the heatmap analysis. The six wetlands and the different periods (ten-year intervals) are employed as the two objects of the heatmap analysis. The PC1 scores are predicted using the “interpSpline” function in the “splines” package, and the heatmap analysis is carried out using the “heatmap.2” function in the “gplots” package of the R statistical computing environment (Team, 2015; Warnes et al., 2009).

## 3. Results

### 3.1. Carbon accumulation rates

The average C ARs in the six wetland profiles are between $136 \pm 77$ g m$^{-2}$ yr$^{-1}$ in the ZBD wetland and $391 \pm 342$ g m$^{-2}$ yr$^{-1}$ in the ZJ wetland (Table 2). At the beginning of the 1900s, the C ARs in the six wetlands are all lower than 100 g m$^{-2}$ yr$^{-1}$, and the lowest C AR value (14.6 g m$^{-2}$ yr$^{-1}$) is identified in the ZJ wetland. The C ARs in the six wetland sites display obvious increases from the beginning of the 1900s to the present, and the highest C ARs in all of the wetlands appear after AD 1950. In the DFH, ZBD and ZJ wetlands, the highest C ARs occur after AD 1980; in the other three wetland profiles, the highest C ARs occur from AD 1950 to AD 1980. The highest C ARs in the six wetlands ($563 \pm 298$ g m$^{-2}$ yr$^{-1}$) appear in the ZJ wetland after AD 1980, and these values are nearly two times greater than the lowest C ARs in this period, which are only $180 \pm 49$ g m$^{-2}$ yr$^{-1}$ and occur in the HXZ wetland.

### 3.2. Accumulation rates of the nutrient elements

The average N ARs in the six wetland sites in the Sanjiang Plain during the last 150 years are between $7.9 \pm 5.2$ and $22.7 \pm 17.9$ g m$^{-2}$ yr$^{-1}$ (Table 2). As with the C ARs, the N ARs at the beginning of the 1900s are also the lowest in all of the wetland profiles, and the lowest N ARs are only 0.9 g m$^{-2}$ yr$^{-1}$ in the DFH wetland between AD 1930 and AD 1950. After AD 1950, the N ARs in all of the wetlands display clear increases, and the highest N ARs in the Sanjiang Plain are $34.0 \pm 20.9$ g m$^{-2}$ yr$^{-1}$ in the SJD wetland between AD 1950 and AD 1980. The average P ARs are between $0.9 \pm 0.4$ g m$^{-2}$ yr$^{-1}$ in the ZBD wetland and $3.6 \pm 2.2$ g m$^{-2}$ yr$^{-1}$ in the DFH wetland. The P ARs also increase from the beginning of the 1900s to the present, and the highest P ARs ($4.6 \pm 1.8$ g m$^{-2}$ yr$^{-1}$) occur in the DFH wetland after AD 1980.

### 3.3. Accumulation rates of the potentially toxic elements

Five potentially toxic elements (i.e., Cu, Zn, Pb, Hg, and As) are selected for investigation in this study, and the average ARs of these five elements are shown in Table 2. During the last 150 years, the highest average Pb AR ($275 \pm 147$ mg m$^{-2}$ yr$^{-1}$) occurs in the DFH wetland, and the highest average Hg AR ($298 \pm 218$ μg m$^{-2}$ yr$^{-1}$) occurs in the ZJ wetland. Low average Pb ARs are found in the QDL, SJD and ZBD wetlands, which display values of only $26.1 \pm 16.1$ to $39.5 \pm 40.7$ mg m$^{-2}$ yr$^{-1}$. For the Hg ARs, the lowest values of only $40.3 \pm 21.6$ μg m$^{-2}$ yr$^{-1}$ appear in the HXZ wetland. Higher ARs of the potentially toxic elements mainly occur from AD 1950 to AD 1980 in the HXZ and SJD wetlands and after AD 1980 in the other

**Table 2**
The accumulation rates of carbon, N, P, Cu, Zn, Pb, Hg, and As in four different periods (i.e., AD 1900–1930, AD 1930–1950, AD 1950–1980, and after AD 1980) during the Sanjiang Plain reclamation and during the whole last 150 years. Cu, Pb, and Zn in the DFH and ZJ wetlands are recalculated from Gao et al. (2014b).

| Site | Period AD | Accumulation rates | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Carbon $g\ m^{-2}\ yr^{-1}$ | N $g\ m^{-2}\ yr^{-1}$ | P $g\ m^{-2}\ yr^{-1}$ | Cu $mg\ m^{-2}\ yr^{-1}$ | Zn $mg\ m^{-2}\ yr^{-1}$ | Pb $mg\ m^{-2}\ yr^{-1}$ | Hg $\mu g\ m^{-2}\ yr^{-1}$ | As $mg\ m^{-2}\ yr^{-1}$ |
| DFH | 1900–1930 (n = 1) | 42.6 | 2.2 | 1.2 | 27.9 | 115 | 113 | 81.4 | 11.2 |
| | 1930–1950 (n = 1) | 16.4 | 0.9 | 0.4 | 10.1 | 43.1 | 44.0 | 27.9 | 3.5 |
| | 1950–1980 (n = 6) | 107 ± 55 | 5.8 ± 3.6 | 2.1 ± 1.1 | 53.6 ± 28.6 | 230 ± 118 | 205 ± 100 | 134 ± 68 | 14.7 ± 6.6 |
| | After 1980 (n = 19) | 435 ± 216 | 26.7 ± 11.0 | 4.6 ± 1.8 | 96.9 ± 35.3 | 388 ± 152 | 331 ± 130 | 276 ± 85 | 23.7 ± 9.6 |
| | Average (n = 28) | 321 ± 247 | 19.5 ± 14.1 | 3.6 ± 2.2 | 78.8 ± 42.6 | 319 ± 175 | 275 ± 147 | 221 ± 114 | 19.8 ± 10.4 |
| HXZ | 1900–1930 (n = 3) | 53.9 ± 27.5 | 3.1 ± 1.6 | 1.1 ± 0.6 | 27.6 ± 16.6 | 50.5 ± 30.0 | 49.7 ± 28.9 | 12.0 ± 5.6 | 10.0 ± 5.1 |
| | 1930–1950 (n = 4) | 150 ± 87 | 5.5 ± 5.2 | 3.7 ± 3.4 | 68.6 ± 48.6 | 159 ± 136 | 156 ± 127 | 36.3 ± 16.9 | 27.8 ± 17.4 |
| | 1950–1980 (n = 11) | 222 ± 108 | 14.0 ± 8.2 | 3.7 ± 1.9 | 64.6 ± 32.2 | 139 ± 65 | 158 ± 75 | 48.0 ± 23.3 | 27.3 ± 13.1 |
| | After 1980 (n = 9) | 180 ± 49 | 11.6 ± 3.1 | 1.7 ± 0.8 | 31.0 ± 10.2 | 88.4 ± 53.6 | 74.4 ± 35.8 | 42.2 ± 18.2 | 12.3 ± 4.86 |
| | Average (n = 27) | 178 ± 94 | 11.2 ± 6.6 | 2.8 ± 2.0 | 49.9 ± 32.4 | 115 ± 77 | 118 ± 81 | 40.3 ± 21.6 | 20.4 ± 13.2 |
| QDL | 1900–1930 (n = 1) | 32.6 | 2.0 | 0.07 | 3.25 | 2.6 | 3.5 | 4.3 | 0.4 |
| | 1930–1950 (n = 2) | 106 ± 1 | 7.0 ± 0.1 | 0.2 ± 0.0 | 6.2 ± 1.2 | 4.5 ± 0.4 | 10.7 ± 1.5 | 16.0 ± 1.0 | 1.0 ± 0.1 |
| | 1950–1980 (n = 6) | 270 ± 176 | 17.9 ± 13.7 | 1.0 ± 0.7 | 18.3 ± 13.0 | 15.3 ± 10.4 | 22.6 ± 16.8 | 37.8 ± 23.9 | 2.4 ± 1.6 |
| | After 1980 (n = 7) | 261 ± 68 | 15.2 ± 3.4 | 1.3 ± 0.3 | 22.1 ± 4.8 | 31.7 ± 12.7 | 36.6 ± 10.4 | 67.2 ± 21.0 | 2.8 ± 0.5 |
| | Average (n = 16) | 231 ± 134 | 14.4 ± 9.5 | 1.0 ± 0.6 | 17.5 ± 10.3 | 20.3 ± 15.0 | 26.1 ± 16.1 | 45.9 ± 29.0 | 2.3 ± 1.3 |
| SJD | 1900–1930 (n = 3) | 79.8 ± 14.5 | 4.83 ± 1.4 | 0.2 ± 0.1 | 6.6 ± 2.5 | 14.2 ± 5.0 | 10.3 ± 5.9 | 8.8 ± 2.3 | 0.4 ± 0.1 |
| | 1930–1950 (n = 3) | 184 ± 62 | 10.9 ± 3.7 | 0.6 ± 0.2 | 12.1 ± 4.1 | 22.0 ± 6.6 | 13.7 ± 5.4 | 19.3 ± 6.7 | 0.6 ± 0.4 |
| | 1950–1980 (n = 14) | 523 ± 286 | 34.0 ± 20.9 | 2.8 ± 2.2 | 37.2 ± 23.3 | 61.5 ± 38.5 | 58.8 ± 53.1 | 90.0 ± 56.3 | 3.2 ± 2.0 |
| | After 1980 (n = 11) | 341 ± 94 | 20.3 ± 6.4 | 2.2 ± 0.8 | 19.5 ± 12.2 | 47.9 ± 21.8 | 36.2 ± 19.7 | 140 ± 72 | 3.4 ± 1.7 |
| | Average (n = 33) | 361 ± 256 | 22.7 ± 17.9 | 2.0 ± 1.8 | 24.2 ± 20.4 | 45.7 ± 33.2 | 39.5 ± 40.7 | 87.7 ± 73.0 | 2.6 ± 2.0 |
| ZBD | 1900–1930 (n = 3) | 61.6 ± 34.0 | 2.9 ± 1.7 | 0.6 ± 0.4 | 33.1 ± 16.5 | 112 ± 57 | 41.4 ± 18.8 | 109 ± 58.1 | 6.4 ± 3.3 |
| | 1930–1950 (n = 2) | 72.1 ± 62.0 | 3.3 ± 2.9 | 0.6 ± 0.5 | 27.4 ± 22.2 | 91.0 ± 75.6 | 21.9 ± 18.3 | 90.5 ± 84.1 | 4.9 ± 4.2 |
| | 1950–1980 (n = 7) | 109 ± 58 | 5.9 ± 2.7 | 0.9 ± 0.4 | 37.8 ± 11.7 | 127 ± 47 | 37.6 ± 14.1 | 138 ± 67 | 7.8 ± 3.5 |
| | After 1980 (n = 11) | 185 ± 70 | 11.4 ± 5.1 | 1.0 ± 0.4 | 42.5 ± 21.0 | 152 ± 81 | 42.5 ± 25.4 | 219 ± 90 | 14.1 ± 6.0 |
| | Average (n = 23) | 136 ± 77 | 7.9 ± 5.2 | 0.9 ± 0.4 | 37.6 ± 17.7 | 134 ± 68 | 39.1 ± 20.7 | 168 ± 90 | 10.4 ± 5.9 |
| ZJ | 1900–1930 (n = 1) | 14.6 | 0.9 | 0.2 | 5.10 | 19.3 | 21.2 | 21.7 | 2.0 |
| | 1930–1950 (n = 1) | 32.7 | 2.1 | 0.6 | 11.0 | 39.8 | 41.0 | 50.4 | 3.9 |
| | 1950–1980 (n = 4) | 126 ± 92 | 8.2 ± 6.0 | 2.0 ± 1.3 | 36.1 ± 21.6 | 149 ± 110 | 158 ± 104 | 172 ± 108 | 14.0 ± 9.8 |
| | After 1980 (n = 17) | 563 ± 298 | 30.6 ± 14.6 | 4.4 ± 1.7 | 48.5 ± 24.3 | 203 ± 85 | 233 ± 89 | 406 ± 182 | 22.8 ± 8.4 |
| | Average (n = 26) | 391 ± 342 | 21.5 ± 17.6 | 3.2 ± 2.2 | 38.5 ± 26.6 | 160 ± 105 | 182 ± 114 | 298 ± 218 | 17.6 ± 11.1 |

four wetlands. High Pb ARs also appear in AD 1900–1930 only in the ZBD wetland, and these values are higher than those that occur in AD 1950–1980.

## 4. Discussion

### *4.1. Impacts of reclamation on the accumulation of the nutrient elements and the carbon cycle*

To evaluate the effects of historical farmland reclamation on wetland ecosystems within the study region, the historical variations in sedimentation rates and the ARs of C, N, and P are used to evaluate the historical variations in the accumulation of the nutrient elements and their effects on the carbon accumulated in the wetlands (Fig. 2). Carbon decomposition processes are mainly influenced by water depth and microbial activities in wetlands, which are affected in turn by changes in the surrounding environment (Thormann et al., 2002; Knorr and Blodau, 2009). Thus, the C/N mass ratio is used as an indirect index and is used to evaluate the impacts of farmland reclamation on carbon decomposition in the surrounding wetlands (Fig. 2).

After the Qing government allowed people from central China to immigrate into northeastern China, immigrants migrated into the Sanjiang Plain and reclaimed farmland after AD 1895 (Zhang et al., 2006). The first reclamation period ended in approximately AD 1930. During this period, the area of farmland in the Sanjiang Plain increased from 133 $km^2$ before AD 1895 to 4400 $km^2$ in AD 1930, and the proportion of farmland increased from 0.2 to 8.5% (Group, 1998). In addition, the population of Heilongjiang province also increased from 1.3 million to 6.0 million (Li and Yuan, 1996). During this period, most of the landscape in the Sanjiang Plain was occupied by natural ecosystems that experienced no direct effects from human activities. During the first reclamation period, the sedimentation rates in most of the wetland profiles (i.e., the DFH, ZBD, HXZ and SJD wetlands) were somewhat higher than during the preceding or following periods; no obvious peaks are present in the other two wetland profiles. Because of the low proportion of reclaimed farmland, most of the wetlands reflect little influence from farmland reclamation on sedimentation rates during this period. As with the sedimentation rates, reclamation also had limited effects on the ARs of C and N, and no obvious increasing trends in the ARs of these two elements are seen during the first reclamation period. However, the ARs of P in the HXZ wetland begin to increase at the end of the 1920s, and the ARs of P in approximately AD 1930 are obviously higher than during the adjacent periods. The C/N mass ratios continued to decrease before the 1930s in all of the wetland profiles, which indicate an increase in the decomposition rates of C. The increase in farmland area may have reduced the amount of water available to the wetlands. In addition, the decrease in the amount of available water increased the contents of oxygen and enhanced the activity of microbes in the surface layers of the wetlands, which likely substantially increased the decomposition rate of C and led to the decomposition of additional C (Knorr and Blodau, 2009). Only the ARs of P increase in approximately AD 1930, and the C/N mass ratios clearly decrease during the first reclamation period; thus, the period of reclamation that extended from AD 1900–1930 influenced only the amount of water available to the wetlands and slightly increased the

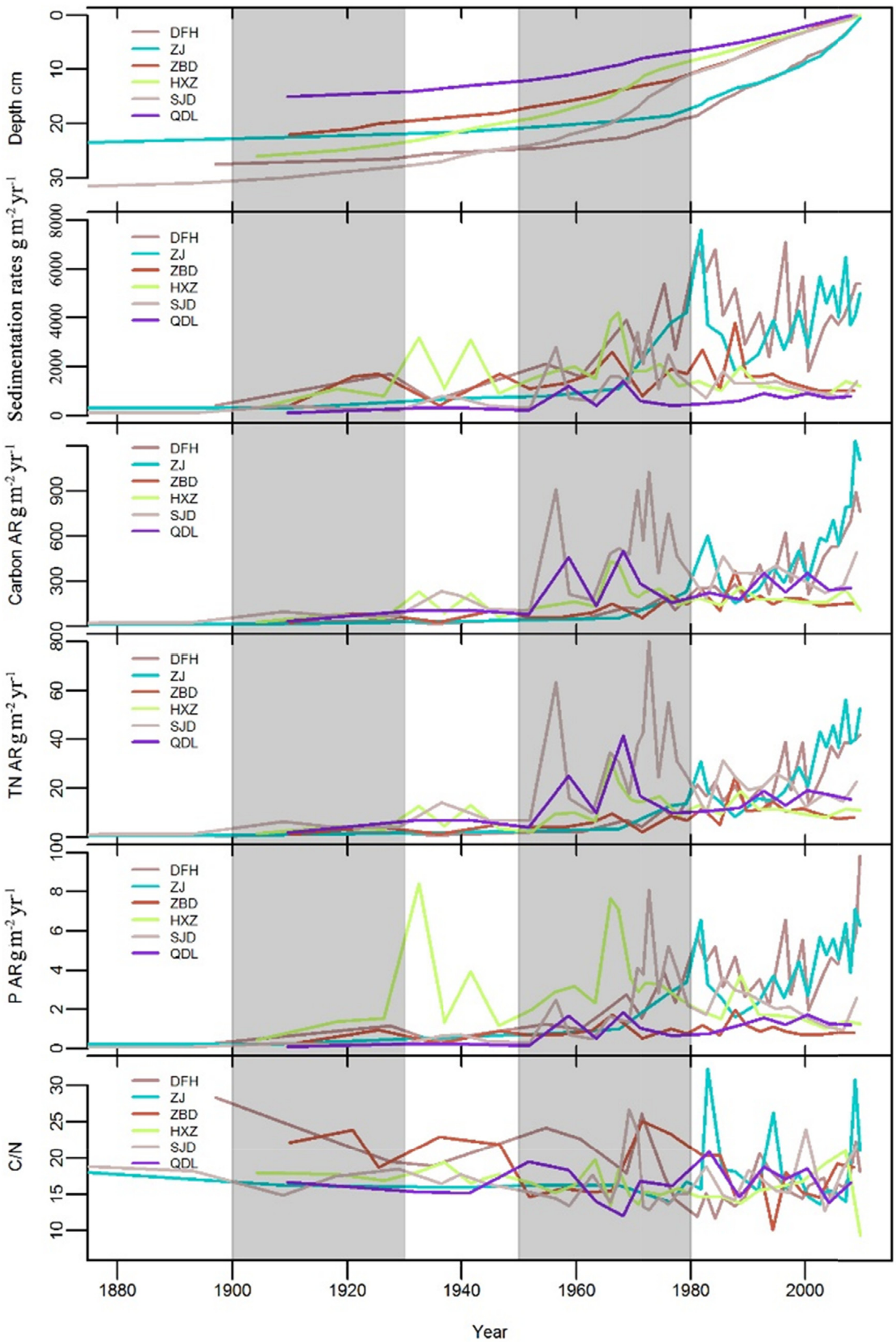


**Fig. 2.** Age-depth models of the wetland profiles in this study. Historical variations in sedimentation rates and the accumulation rates of carbon (C), total nitrogen (TN), and phosphorus (P) and the mass ratios of C/N in the six wetland profiles in the Sanjiang Plain.

accumulation of P in the surrounding wetlands.

From the 1930s to AD 1945, northeastern China was occupied by the Japanese army, and the wars caused part of the farmland to lie fallow (Zhang et al., 2006). Although the Japanese government encouraged reclamation in the Sanjiang Plain, the area of farmland in AD 1945 was similar to that in AD 1930. The second reclamation period extended from AD 1950 to the 1980s. In this period, the area of farmland increased from 7867 $km^2$ to 35,200 $km^2$, and the proportion of farmland in the Sanjiang Plain increased from 15.3% to 68.6% in AD 1989 (Group, 1998). More than 80% of the wetlands were reclaimed and changed to farmland during this period (Wang et al., 2011). The sedimentation rates and the ARs of C begin to increase in most of the wetland profiles in the Sanjiang Plain after AD 1930, and more obvious increasing trends appear in AD 1950–1980. Especially for the ARs of C, obvious peak values appear in AD 1950–1980 in the SJD, QDL and HXZ wetlands. Similar to the ARs of C, the ARs of N in these three wetlands also display clear increases during the second reclamation period and are clearly higher than before. The sedimentation rates in the DFH and ZJ wetlands also begin to increase in approximately AD 1960 and display intense fluctuations from AD 1960 to AD 1980. After AD 1950, the C/N mass ratios fluctuate, and weakly increasing trends can be seen in most of the wetland profiles. In approximately AD 1970, obvious peaks in the C/N mass ratios appear in the DFH, SJD, and ZBD wetlands. In addition, high C/N mass ratios also occur in approximately AD 1980 in the ZJ, QDL and SJD wetlands. The reclamation during the second period caused the sedimentation rates and the ARs of C in the wetlands to increase substantially, and they display considerable fluctuations after AD 1950.

After the second reclamation period, the local government devoted more attention to the environmental problems caused by farmland reclamation and began to implement several environmentally friendly policies and to reduce the rate of farmland reclamation. The major farming methods were gradually replaced by mechanization, and large amounts of fertilizers and pesticides were widely used to increase production (Group, 1998). In addition to the increasing influence of agriculture within the region, industrial development and the increase in the population of the Sanjiang Plain also increased the degree of anthropogenic effects on the surrounding environment after AD 1980. After AD 1980, the sedimentation rates in the DFH and ZJ wetlands clearly increase, and the ARs of C, N and P also display obvious increases. In the ZBD and HXZ wetlands, the ARs of N increase, similar to those in the SJD and QDL wetlands. Except for the DFH and ZJ wetlands, the ARs of P in the wetlands decrease and are approximately 1.5 $g\ m^{-2}\ yr^{-1}$ after AD 2000. The ARs of C in all of the wetlands display clear increases after AD 1980 and achieve their highest values after AD 2000 because the primary production of wetland plants in fens is substantially higher than that of bog plants, which receive limited amounts of nutrient elements (Bartsch and Moore, 1985; Turunen et al., 2004). The greater amounts of nutrient elements that were produced by human activities in the region and accumulated in wetland ecosystem during this period represent a factor that may have increased the primary production of plants and the ARs of C in wetlands. As the area of wetlands in the floodplain decreased, the amount of river clay and sand sedimentation in the remaining wetlands may have increased; an increase in the storage of carbon derived from river sediments in the wetlands is another potential factor that may have increased the ARs of C. Although the ARs of C clearly increased after AD 1950 and are 2–4 times higher than those before AD 1950, the loss of nearly 80% of the wetlands during this period caused the ecosystem service of carbon accumulation in the wetland ecosystem in the Sanjiang Plain as a whole to degrade and become weaker than before the farmland reclamation period. The C/N mass ratios in all six wetland profiles increase slightly and are slightly higher than those in the 1980s. In addition to the low degree of decomposition in the surface layers of wetland profiles, the greater C accumulation in wetlands is an important factor that led the sedimentation rates in these wetland profiles to obviously increase after the 1980s. In contrast to the second reclamation period, given the end of farmland reclamation in the region, the ARs of C, N and P in the different wetlands became similar after AD 2000. The similarities in the historical variations in the ARs of C, N and P indicate that anthropogenic effects, such as industrial development and population growth, have become the dominant factors in the region and have caused the ARs of the nutrient elements to display clear increases in the wetland ecosystems in the Sanjiang Plain after AD 2000.

Taken together, the farmland reclamation during the first reclamation period caused the ARs of P in the HXZ wetland to begin to increase only after AD 1920 and had few effects on the accumulation of the nutrient elements. During the second reclamation period, as the area of farmland and agricultural development increased, greater amounts of the nutrient elements accumulated in the surrounding wetlands, and the ARs of both N and P display clear increases. The high ARs of the nutrient elements increased the primary production of plants and caused the ARs of C to clearly increase during this period. After the rate of farmland reclamation decreased in the 1980s, the industry and population of the region became the major factors that influenced the ARs of C and the nutrient elements in the wetlands and caused the formerly different ARs of these elements in the different wetlands to become similar.

### *4.2. Effects of reclamation on the accumulation of the potentially toxic elements*

In wetland ecosystems, potentially toxic elements adsorb onto rocks, inorganic sediment particles, organic sediment particles and their associated microorganism communities. Because of the low rates of diffusion of these elements within sediments, only the upper layer of sediment (primarily the diffusive boundary layer) can potentially accumulate large amounts of these elements (Schaller et al., 2013), and the migration of elements from higher layers to lower layers in wetlands can be ignored. However, the presence of large amounts of decomposed carbon can cause larger amounts of potentially toxic elements to accumulate in layers with high degrees of carbon decomposition (i.e., low C/N mass ratios) (Biester et al., 2003). The C/N mass ratio is selected as an indirect index to evaluate the impacts of the historical degree of carbon decomposition on the potentially toxic elements that have accumulated in the wetlands. Pb, Cu, Zn, Hg and As are selected as typical potentially toxic elements to reflect the historical deposition of potentially toxic elements in the six wetlands in the Sanjiang Plain examined here. The historical ARs of these elements and the enrichment factors calculated using the Al concentrations are shown in Fig. 3.

During the first reclamation period (AD 1900–1930), three of the potentially toxic elements begin to increase in the DFH, ZJ, ZBD and HXZ wetlands. On the other hand, no obvious variations in the ARs of these potentially toxic elements are seen in the other two wetlands. Clear peak values of the potentially toxic elements appear in approximately AD 1920 in the DFH and ZJ wetlands. Unlike the ARs of the elements, the values of $Pb_{EF}$ and $Hg_{EF}$ are stable and similar in most of the wetlands during this period, with the exception of the QDL wetland. However, the values of $As_{EF}$ in the HXZ wetland are obviously higher than those in the other wetlands and are approximately 17. Except for the SJD wetland, for which the $As_{EF}$ values are approximately 3, the $As_{EF}$ values in the other four wetlands are approximately 7. The increasing population

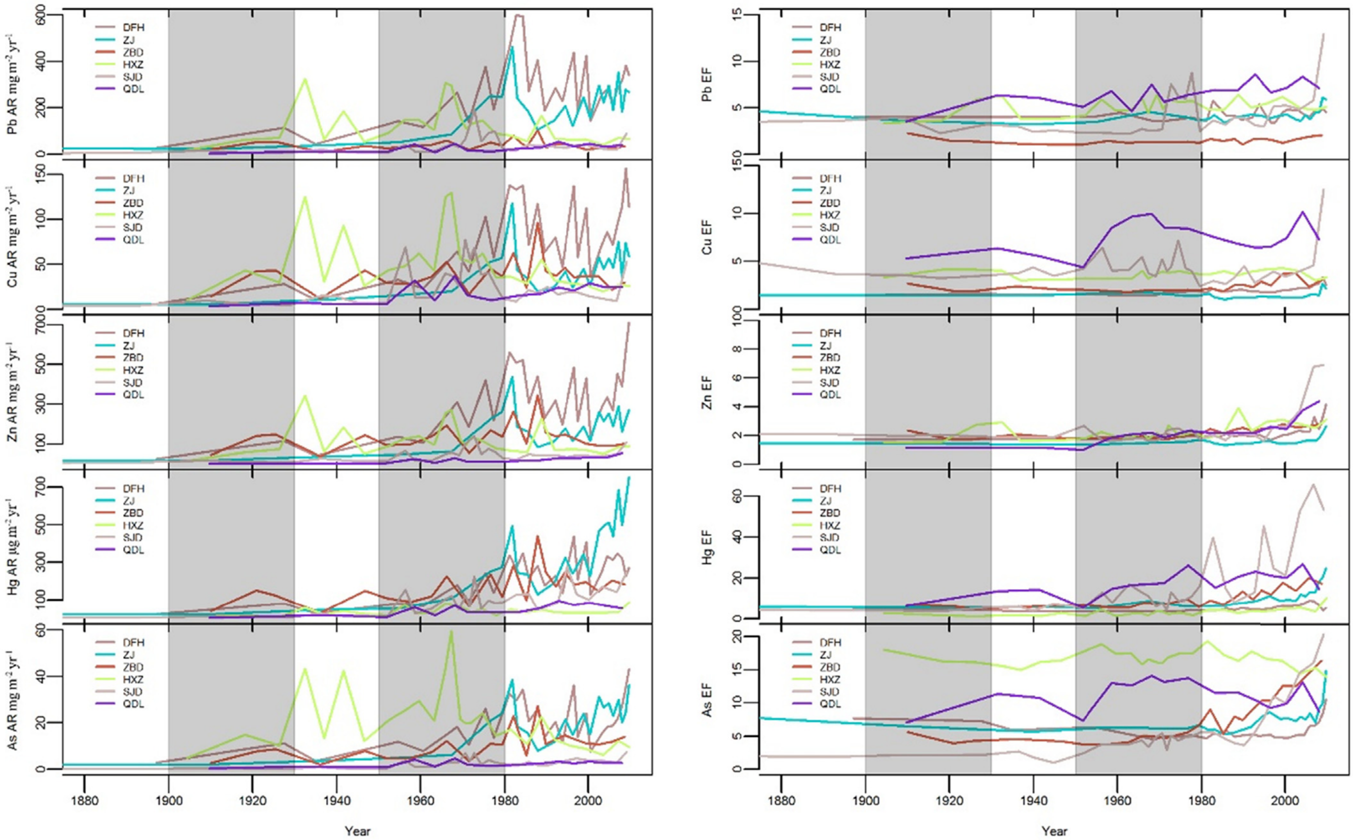


**Fig. 3.** Historical accumulation rates and enrichment factors of Pb, Cu, Zn, Hg and As in the six wetland profiles in the Sanjiang Plain. The accumulation rates of Cu, Pb, and Zn and the EFs in the DFH and ZJ wetlands were obtained from Gao et al. (2014b).

and area of farmland during the first reclamation period had already begun to influence the accumulation of the potentially toxic elements in the surrounding wetlands in the Sanjiang Plain, in contrast to C and the nutrient elements.

After the first reclamation period, the ARs of the potentially toxic elements in most of the wetlands decrease. Only in the HXZ wetland do the ARs of Pb and As increase and display considerable fluctuations during this period. Unlike the ARs of Pb and As, $Pb_{EF}$ and $As_{EF}$ both decrease in the HXZ wetland, which means that the major sources of these potentially toxic elements are natural. More dust enriched in Pb and As was deposited in the HXZ wetland and caused the ARs of these two potentially toxic elements to display obvious increases. After AD 1950, more people immigrated into the Sanjiang Plain, and the area of farmland increased at a much greater rate than during the first reclamation period (Zhang et al., 2006; Wang et al., 2011). During the second reclamation period, the ARs of the potentially toxic elements in the Sanjiang Plain increase substantially, and the enrichment factors of these potentially toxic elements also begin to increase. Especially in the DFH and ZJ wetlands, the highest ARs of Pb and $Pb_{EF}$ values both appear in approximately AD 1980. These high $Pb_{EF}$ values indicate that increases in the anthropogenic sources of Pb are the major factors that caused the ARs of Pb in the DFH and ZJ wetlands to increase in approximately AD 1980. In the other four wetlands, several clear peaks in the ARs of the potentially toxic elements are found in the second reclamation period. The increasing trends in the ARs of the potentially toxic elements and the high values of the enrichment factors seen in all of the wetlands indicate that the increases in population and the area of farmland are the major factors that caused serious environmental problems in the natural ecosystems during this period.

The production and emissions of potentially toxic elements (i.e., Hg, Cu, Pb, and Zn) have also obviously increased globally during the last 150 years, and the highest levels of emissions occurred in the 1970s (Nriagu, 1996; Hylander and Meili, 2003). After AD 1980, the emissions of these elements began to decrease globally due to the development of several environmental friendly policies and technologies, such as the use of unleaded gasoline in place of leaded gasoline. Due to the rapid industrial and economic development that has taken place in China in the last several decades, the emissions of most pollutants obviously increased after the 1980s (e.g., Bond et al., 2007). In addition, local sources of pollutants caused greater amounts of potentially toxic elements to accumulate in most natural ecosystems in China from AD 1980 to the 2000s (Bing et al., 2011; Bao et al., 2016). Similar trends in the of ARs of potentially toxic elements are also found in the Sanjiang Plain. Although the values of $Pb_{EF}$ and $Hg_{EF}$ in the DFH and ZJ wetlands do not obviously increase after AD 1980, the ARs of Pb and Hg do increase and are higher than those in the other wetlands. In particular, the values of $Hg_{EF}$ are relatively high in most of the wetlands in the Sanjiang Plain, particularly the SJD wetland (where they range from 10 to 70), which is located closest to the cities (e.g., this wetland lies 34 km from Jiamusi). In addition to $Hg_{EF}$, the values of $As_{EF}$ and $Pb_{EF}$ in the SJD wetland are also clearly higher after AD 1980 and particularly after approximately AD 2000. The local government began to pay attention to the environmental problems caused by reclamation, and the area of farmland in the Sanjiang Plain increased more slowly after AD 1980 than during the second reclamation period (Group, 1998). As agricultural development progressed, greater amounts of fertilizers and pesticides were applied to increase agricultural production. The increases in population, fertilizer use and the emissions of pollutants in the region also increased the amounts of the potentially toxic elements released from anthropogenic sources and deposited into and accumulated by the surrounding ecosystems.

Briefly, the ARs of potentially toxic elements in wetlands are more easily influenced by farmland reclamation than the ARs of the nutrient elements. In addition, the ARs of the potentially toxic elements in the ZBD and HXZ wetlands begin to increase during the first reclamation period. As the degree of reclamation increased, more potentially toxic elements produced by anthropogenic sources caused the enrichment factors and the ARs of all of the potentially toxic elements to display clear increases from AD 1950 to AD 1980. Although the local government began to protect the wetlands and decreased the rates of wetland loss after AD 1980, the increases in population and the amounts of fertilizer used in agriculture still led to the release of greater amounts of potentially toxic elements. In addition, local anthropogenic sources led to increases in the enrichment factors and the ARs of these potentially toxic elements in the surrounding wetlands. The wetlands close to the cities are more easily influenced by pollutants produced by industrial facilities in the surrounding cities than the other wetlands, as reflected by the ARs and enrichment factors of the potentially toxic elements (e.g., Hg).

### *4.3. Spatial and temporal variations in the degree of anthropogenic influence on the wetlands*

To identify the degree to which historical controlling factors (e.g., farmland reclamation and human activities) affected the wetlands in total, PCA is used to quantify these factors using the ARs of C, the nutrient elements (i.e., N and P) and the potentially toxic elements (i.e., Cu, Pb, As, Hg and Zn). The first component accounts for 71.2% of the total variance of the ARs of these elements and is characterized by negative loadings on the ARs of all of the elements (Table 3). The first principal component (PC1) is negatively correlated with the ARs of the potentially toxic and the nutrient elements, and PC1 can be regarded as a quantitative indicator of historical variations in the combined effects of anthropogenic factors on the wetlands in the Sanjiang Plain; low PC1 scores indicate a high degree of anthropogenic influence on the wetlands. Historical variations in the PC1 scores in the six wetland profiles are calculated and shown in Fig. 4b. Based on the decadal PC1 scores in each wetland profile, the heatmap analysis is used to divide the two factors (i.e., site and time) into several groups, and the results are shown in Fig. 4c.

Before AD 1900, the PC1 scores in all of the wetland profiles are high, indicating a low degree of human influence on these wetlands during this period. During the first reclamation period (which extended from AD 1900 to AD 1930), the PC1 scores in the DFH, HXZ and ZBD wetland profiles decrease, indicating that human activities began to influence these wetlands during the first reclamation period. However, no obvious decreasing trend in the PC1 scores can be seen in the other three wetland profiles, indicating that reclamation had a weak influence on environmental quality in other three wetlands during this period. After the first reclamation period, the PC1 scores in most of the wetland profiles are approximately 2 and are similar to those before AD 1900. Low PC1 scores appear only in the HXZ wetland, indicating that the HXZ wetland experienced the effects of human activities before the other wetlands in the Sanjiang Plain. During the second reclamation period, the PC1 scores in all of the wetland profiles display clear decreases, and the lowest PC1 scores appear in the SJD and HXZ wetlands. The low PC1 scores indicate that human activities had serious effects on the wetlands in the Sanjiang Plain during this period. After the second reclamation period, the PC1 scores in some of the wetlands increase and are slightly lower than those in AD 1930–1950. However, low PC1 scores are noted in the DFH and ZJ wetlands, indicating that human activities still affected these two wetlands to a substantial degree. The average PC1 scores in the six wetland profiles in the Sanjiang Plain may reflect the combined effects of historical human activities on wetland ecosystems in the Sanjiang Plain. As noted above, the average PC1 scores also show that human activities began to influence wetland ecosystems in the Sanjiang Plain during the first reclamation period, and the degree of this influence clearly increased during the second reclamation period. After the second reclamation period, the degree of human influence on the regional wetland ecosystems stabilized at levels similar to those in the second reclamation period.

Based on the PC1 scores and the heatmap analysis, the ZBD and HXZ wetlands are divided into one group; the historical PC1 scores in these two wetland profiles clearly differ from those in the other wetland profiles. In the HXZ and ZBD wetlands, human activities began to influence the elemental ARs in approximately AD 1900, much earlier than in the other four wetland profiles, which display the first signs of human activities in approximately AD 1950. Of the other four wetland profiles, human activities began to seriously influence the DFH and ZJ wetlands slightly later than the SJD and QDL wetlands (i.e., in the 1970s). Compared to the history of human activities in the region (Fig. 4a and b), wetland ecosystems in the Sanjiang Plain began to be influenced by human activities in approximately AD 1900, when immigrants from central China began to reclaim farmland during the first reclamation period (Zhang et al., 2006). The limited population and area of farmland during the first reclamation period meant that only a few wetlands began to be influenced by human activities. As increasing numbers of people immigrated into the Sanjiang Plain during the second reclamation period, the area of farmland increased substantially, and human activities within the region increased the degree of impact on all of the surrounding wetlands. Although the second reclamation period ended in the 1980s (Group, 1998), the greater amounts of pollutants produced by residential and industrial sources still influenced the surrounding ecosystems and led to serious impacts on these wetlands; these impacts increased further after AD 2000.

Based on the PC1 scores and the heatmap analysis, the influence of human activities on the wetlands in the Sanjiang Plain during the last 150 years can be divided into three periods. The first period is before AD 1900 and displays low degrees of influence of human activities on all of the wetlands. After AD 1900, human activities in the region began to influence the wetland ecosystems in the Sanjiang Plain, and two sub-periods can be identified within this period (i.e., AD 1900–1970 and AD 1970–present). From AD 1900 to AD 1970, some of the wetlands were influenced by human activities

**Table 3**
Loadings of the investigated elements of the first two principal components obtained using PCA of the entire Sanjiang Plain dataset.

| | PC1 | PC2 |
|---|---|---|
| Standard deviation | 2.38 | 1.15 |
| % Variance | 71.2 | 16.6 |
| Cumulative variance | 71.2 | 87.8 |
| C | −0.310 | 0.557 |
| N | −0.306 | 0.558 |
| P | −0.392 | 0.065 |
| Pb | −0.378 | −0.266 |
| Cu | −0.378 | −0.259 |
| Zn | −0.372 | −0.308 |
| Hg | −0.328 | 0.176 |
| As | −0.353 | −0.332 |

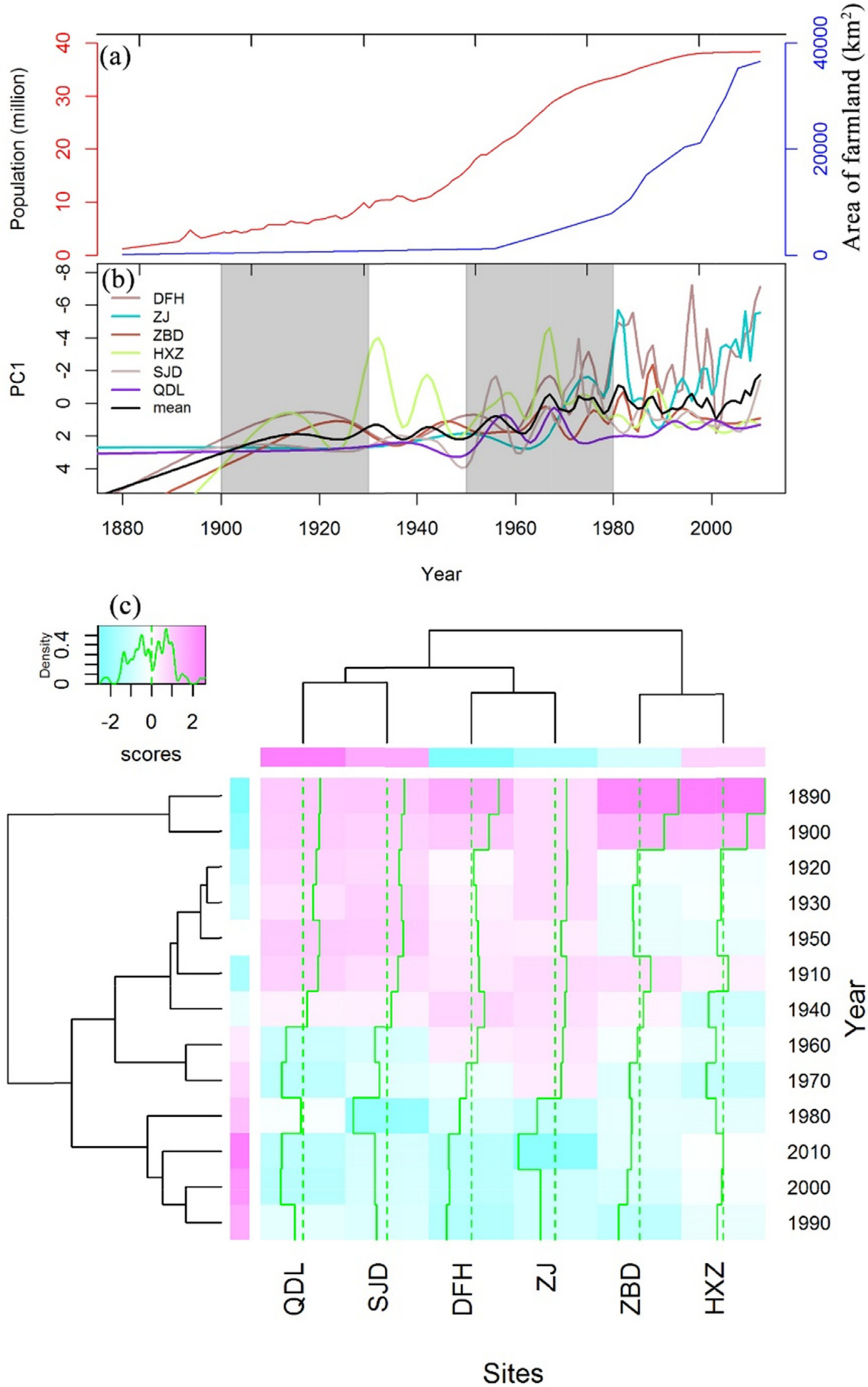


**Fig. 4.** a Population in Heilongjiang Province and the area of farmland in the Sanjiang Plain; b Historical variations in the PC1 scores in the six wetland profiles in the Sanjiang Plain. c Results of the heatmap analysis of PC1 scores (effects of human activities on wetlands) in the six wetland profiles in different periods (from AD 1880–2010).

in the region at particular times (i.e., ZBD and HXZ in AD 1900–1950 and SJD, QDL, ZBD and HXZ in AD 1950–1970). Few of the wetlands in the Sanjiang Plain were influenced by farmland reclamation during the first reclamation period, whereas more of the wetlands were influenced by farmland reclamation during the second reclamation period. Although the second reclamation period ended in approximately AD 1980, the greater amounts of pollution produced by the residents of the region and industrial development still influenced the environmental quality and increased the degree of anthropogenic influence on the surrounding wetland ecosystems in the Sanjiang Plain. All of the wetlands continued to be influenced by human activities in the region from AD 1970 to present, and these last 40 years are assigned to one group.

## 5. Conclusion

Based on the ARs of the key elements and statistical analyses, the historical effects of farmland reclamation on the surrounding wetland ecosystems in the Sanjiang Plain are evaluated in this study. The increases in the area of farmland and the development of agriculture resulted in increases in the accumulation of the nutrient and potentially toxic elements in the wetland ecosystems. The increases in the ARs of the nutrient elements is one of the important factors that led to the observed increases in the ARs of C in the wetlands. During the first reclamation period, in AD 1900–1930, the reclamation process influenced only a few of the wetland profiles (i.e., HXZ and ZBD). However, the second reclamation period caused the ARs of the potentially toxic elements in all of the wetland profiles to increase distinctly. All of the residual wetlands in the Sanjiang Plain began to be influenced by human activities after AD 1970. Although farmland reclamation decreased after AD 1980, the increases in the population and industrial development of the region continue to influence these wetlands and increase the potential environmental risks to all of the residual wetland ecosystems.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.02.075.

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