Environ Earth Sci (2012) 66:697–705
DOI 10.1007/s12665-011-1277-4

ORIGINAL ARTICLE

Temporal and spatial distribution of trace metals in sediments from the northern Yellow Sea coast, China: implications for regional anthropogenic processes

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Received: 2 September 2010 / Accepted: 24 July 2011

Abstract Surface sediment samples from 17 sites in the Yantai coastal area, the northern Yellow Sea, China, combined with a sediment core were employed for geochemical and chronological analyses for the purpose of characterizing the temporal and spatial distribution of trace metals in sediments and their implications for anthropogenic processes. The results indicated that the spatial distribution of trace metals (Cr, Ni, Ti, Pb, As, Zn, Mn and Cu) in surface sediments was significantly contributed by the sewage discharges along the Yantai coast, and the coastal currents played a major role for transporting the pollutants to offshore. The temporal concentrations of trace metals in the sediment core based on the chronology determined by a combination of radionuclide ^{137}Cs and ^{210}Pb activity demonstrated that trace metal concentrations increased step-wisely over the last ca. 100 years, corresponding to the intensity of anthropogenic processes in the Yantai area. The high levels of Cu and As before the late 1970s indicated the agricultural emission from the application of pesticides. While, all the high-trace metal concentrations since the early 1980s could be seen as diagnostic indictors of increasing industrialization, urbanization and sewage discharge in the Yantai area. Although the potential ecological risk evaluation of trace metals in the coastal area suggests low-potential ecological risk at present, some trace metals, such as As and Pb need particular attention due to their slight contamination.

Keywords Yellow Sea · Trace metals · Anthropogenic activities · Pollution

Introduction

The enrichment of trace metals in the environment is usually the result of both natural and anthropogenic processes. For the densely populated coastal areas, the potentially toxic trace metals from local and regional anthropogenic emissions have significantly threatened the environmental and ecological security (e.g., Förstner and Wittmann 1981; Nriagu 1989; Loring 1991). Documenting and better understanding of the basis of natural and human-induced trace metals emissions in environment is thus an important task for the environment scientists and policy-makers.

Trace metals in semi-closed coastal environments are difficult to dilute by water discharge or easily removed by self-purification (Loska et al. 2003; Chen et al. 2004). The concentrations of trace metals in sediments can thus act as ideal archives preserving valuable information of natural and anthropogenic processes in the environment, both temporally and spatially.

This work presents sediment records of trace metals from a semi-closed coastal area around Yantai city, northern Yellow Sea, China (Fig. 1). The aim was to

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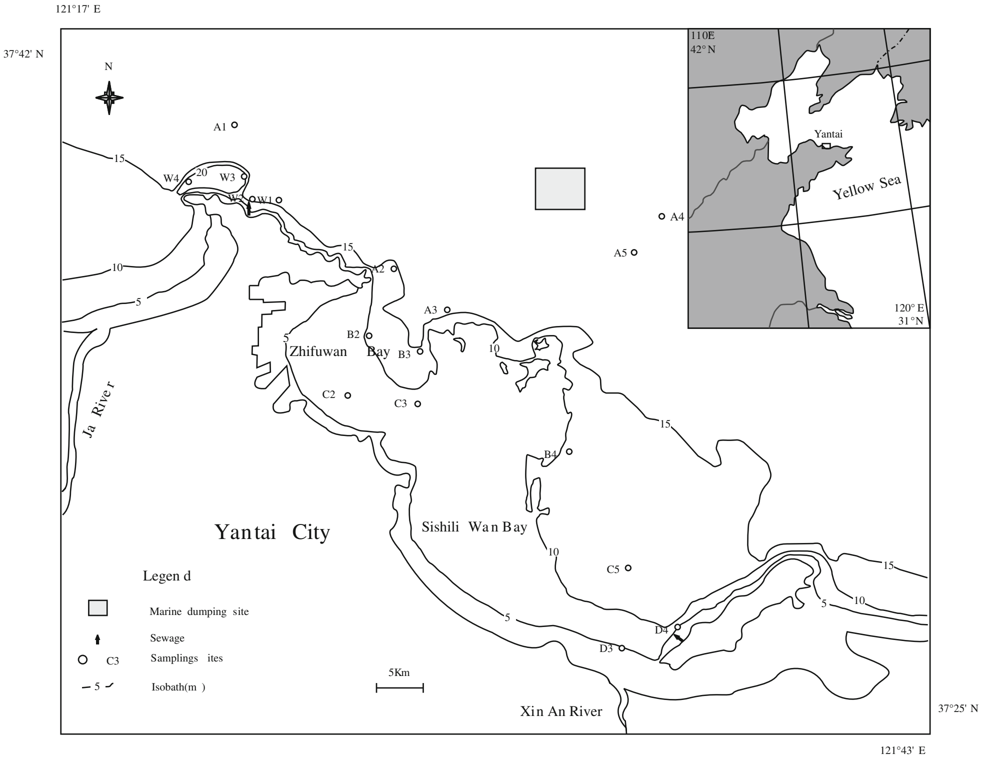

Fig. 1 Map of the study area showing the location of study area, sampling sites, city sewages, rivers and the coastal ocean bathymetry

characterize the temporal and spatial distribution of trace metals in the Yantai coastal area. Then, the historical data of the regional economy devolvement were incorporated into the time sequences reconstructed from the sediment core. This approach will help in evaluating the potential ecological risks of trace metals, and examine the historical intensity of anthropogenic forcing on the environment of coastal areas.

Materials and method

Study area and sampling methods

The study was carried out in the Sishili Bay (SB) and Zhifu Bay (ZB) along the Yantai coast which located in the northern Yellow Sea, China (Fig. 1). The semi-closed coastal area covers a northwest–southeast extent of 20 km and a width of 6–7 km with an area of 119 km^2, and the water depth of SB is generally <15 m. Two city sewage plants discharge waste water into the bay from the northern point (Zhifu B) and southern point (Xin An river), respectively (Fig. 1).

In November 2008, 17 surface sediment samples (0–5 cm) were collected in the coastal area using a Peterson Grab. Four sediment cores at site A5 were obtained using gravity sampling devices (Fig. 1). Site A5 is believed less affected by human activities, such as channel dredging, and has a relatively stable sedimentation environment which made it possible for high-resolution research (Ji et al. 2003). The sediment cores were sliced into subsamples at 2-cm intervals. All the sediment samples were then stored in refrigerator for multi-proxy analysis and dating. One of the sediment cores (130-cm in length) was used for radionuclide dating and elemental analysis.

Elements analysis

For elemental tests, sediment samples that were basically clay-silt were dried at 60°C, slightly disaggregated, and then sieved through a 200-mesh sieve to discard coarse materials. Sediment pellets of 3 cm diameter were prepared using a hydraulic press applying a pressure of 20 ton cm^{-2}. Major element oxides (SiO_2, Al_2O_3, TFe_2O_3, MgO, CaO, Na_2O, K_2O and P_2O_5 and trace metals Cr, Ni, Ti, Pb, As, Zn, Mn and Cu analyses were measured by Axios Advanced (model, PW4400) X-ray Fluorescence Spectrometry (XRF) at Institute of Earth Environment, Chinese Academy of Sciences(IEECAS). The precision and accuracy of the data were verified by parallel checks of two national reference samples (GSS-8 and GSD-12) which were measured as ‘unknowns’ with the samples and sample duplicates. The analytical uncertainties are estimated 1–2% for all major metals, and the relative standard deviation (RSD) is <5% for the trace metals. The concentrations of major and trace elements were given in oxide form in percentage (%) and form of mg/kg, respectively. Trace metals normalization to aluminum (Al) were also applied to core sediment to eliminate the grain size effect, since Al is thought as the most insoluble and common, terrestrially derived element (Brown et al. 2000).

Radionuclide ^{210}Pb and ^{137}Cs measurements

Samples from the sediment core were selected for radionuclide analysis (^{210}Pb and ^{137}Cs) to set up the chronology sequence. 4 g of pulverized sample was added to a tube with a cap. The activities of ^{210}Pb (peak energy 46.5 keV), ^{214}Pb (352 keV), ^{137}Cs (661.6 keV) and ^{40}K (1,461 keV) were then measured by gamma-ray spectrometry using well-type Ge detectors (Model Ortec HPGe GWL) at Institute of Geography and Limnology, Chinese Academy of Sciences. The ^{210}Pb and ^{137}Cs activities were given after correction with standard samples provided by Chinese institute of Atomic Energy Research and the University of Liverpool. The Constant Rate of Supply (CRS) model was applied for ^{210}Pb data processing in this study (Appleby and Oldfield 1983; Appleby et al. 1986; Benoit and Rozan 2001). Sedimentation rates were calculated for the core sediment on the basis of the ^{210}Pb and ^{137}Cs measurements.

Results

Trace metals in surface sediments of Yantai coast

For surface sediment samples collected from the 17 sites of Yantai coast, the concentrations of trace metals Cr, Ni, Ti, Pb, As, Zn and Cu show the highest levels in the sites close to the city sewage outlets in contrast to other sites (Fig. 2). For instance, site W4 records the highest levels of Ni (41.8 mg/kg), Cr (85.7 mg/kg), As (19.1 mg/kg), Mn (695.9 mg/kg) and Cu (55.3 mg/kg). Pb (56.5 mg/kg) and Zn (138.6 mg/kg) reaches maximum in site D4. Relatively higher levels of trace metals also appear at the sites (A4 and A5) near the marine dumping zone. Significant ($r > 0.8$) correlations are found between the trace metals (Table 1). For example, Pb is well correlated with Zn and Cu. As shows positive correlations with Cu, Ni and Zn. Correlations also exist between Cr and Ni ($r = 0.723$), Pb ($r = 0.697$), As ($r = 0.673$), Zn ($r = 0.804$) and Cu ($r = 0.794$).

Chronology of core sediment

The radionuclides ^{137}Cs and ^{210}Pb are commonly used for sediment dating over the last 100–200 years (Robbins and Edgington 1975; Delaune et al. 1978; Battiston et al. 1988; Benoit and Rozan 2001). Peak ^{137}Cs activity in sediment sequence is usually correlated upwardly to the world frequent atomic bomb testing at 1963 and 1975, as well as the fallout release from the accident at Chernobyl nuclear station in 1986. Excess ^{210} Pb (^{210}Pb$_{ex}$) in the sediment is also utilized to set up the sediment chronology when the ^{210}Pb$_{ex}$ activity versus depth follows an exponential distribution. The combination of ^{137}Cs and ^{210}Pb$_{ex}$ methods helps in reducing the uncertainty in sediment dating.

The ^{210}Pb$_{ex}$ of the sediment core A5 showed an approximately exponential recession, although there are some sub-fluctuations (Fig. 3). This still allows using constant rate of supply (CRS) model to estimate the sedimentation rate of the sediment core (Appleby and Oldfield 1983; Appleby et al. 1986; Benoit and Rozan 2001), despite some minor variances in dating resulting from the ^{210}Pb$_{ex}$ oscillations. The results indicate that the sediment core A5 was deposited during the time span of ca. 1880 to 2008.

The variation of ^{137}Cs activity versus depth of the sediment core is also shown in Fig. 3. The peaks of ^{137}Cs activity in the sediment core A5 are correlated to the highs of historical fallout emissions suggest that 40.5, 16.5 and 8.5-cm depth in core A5 deposits correspond to the years of 1963, 1975 and 1986. Comparison of peak ^{137}Cs activity with ^{210}Pb$_{ex}$ variations shows some discrepancies, possibly due to the post-depositional mobility of ^{137}Cs upwardly. In this study, the chronology of the sediment core was based primarily on the ^{210}Pb results.

Trace metals in the core sediment

Trace metal variations in A5 core sediment are shown in Fig. 4. As a whole, trace metals Ti, Ni, Zn, As, Pb, Cu and Mn show an increasing tendency upwardly. The concentrations of Ni, Zn, Pb, Mn remain relatively stable at the depth interval ca. 123–65 cm, exhibit a slight decline at the depth of ca.65–42 cm and show a rapid increase in the upper 42 cm section. Concentrations of As change little below a depth of ca. 65 cm, but it increase obviously at the depth of ca. 65–42 cm, slightly decline from ca. 42–30 cm and accumulate again in the upper section. Cu exhibits visible increases at ca. 95–65 cm, then declines at ca. 65–42 cm, and increases to the highest value in the upper core section.

Discussion

Spatial distribution and the impact factors

The distribution of trace metals, such as Ni, Cr, Pb, Zn, Mn, Cu and As in surface sediment of Yantai coast indicates that sewage discharge play an important role in controlling the concentrations of trace metals (Fig. 2). This is in agreement with the common practice that sewage systems collect city wastes and local industrial emissions, with trace metals frequently being drained into the coastal

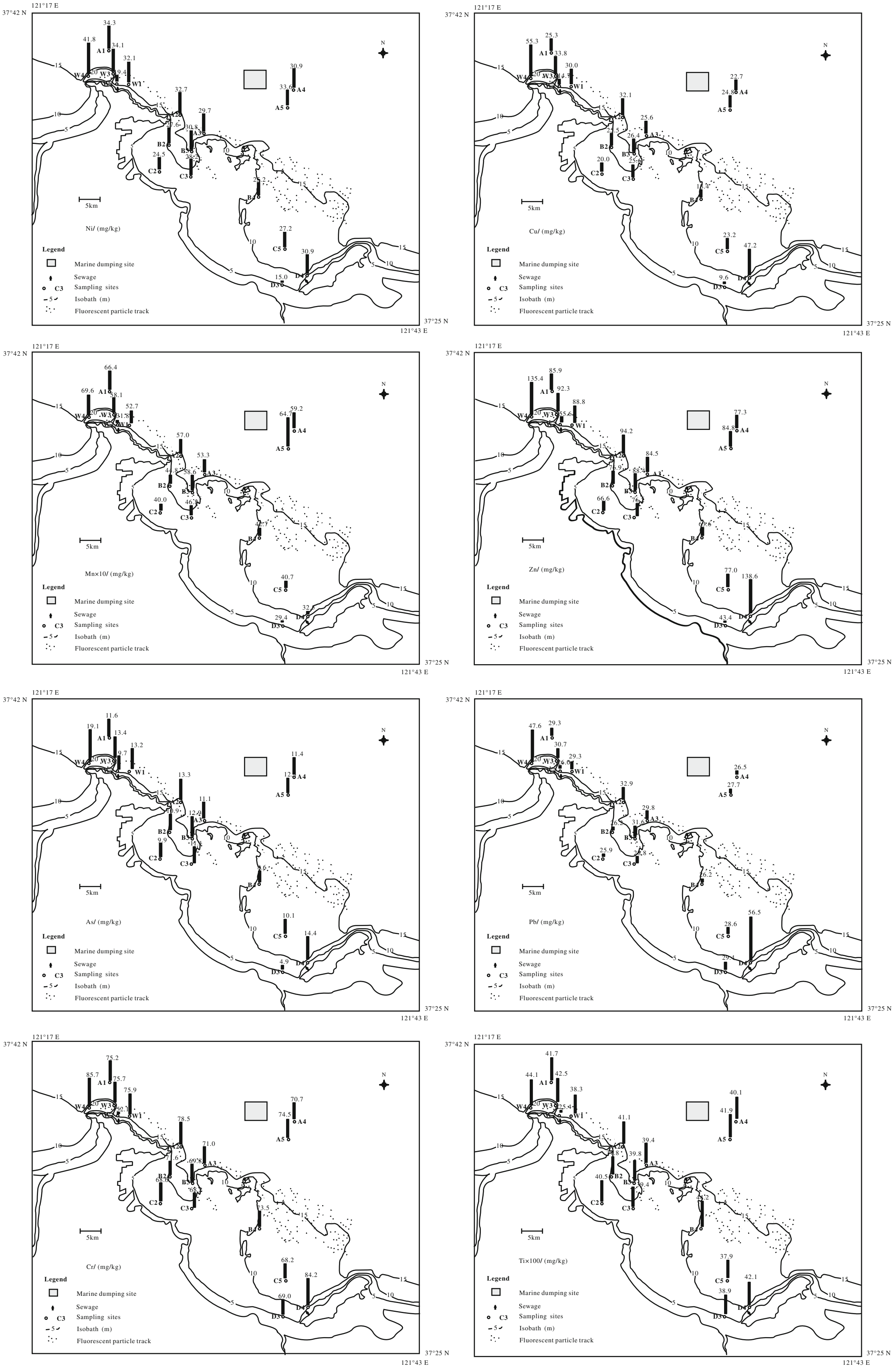

Fig. 2 Spatial distribution of trace metals and the tracks of the fluorescent particle tracer (*dots in black*)

Table 1 Correlation matrix of trace metals in surface sediment samples, Yantai coastal area

	Ni	Cu	Mn	Zn	As	Pb	Cr
Ni	1.000	0.811	0.854	0.803	0.909	0.437	0.723
Cu		1.000	0.436	0.977	0.928	0.838	0.794
Mn			1.000	0.421	0.646	0.021	0.477
Zn				1.000	0.904	0.871	0.804
As					1.000	0.637	0.673
Pb						1.000	0.697
Cr							1.000

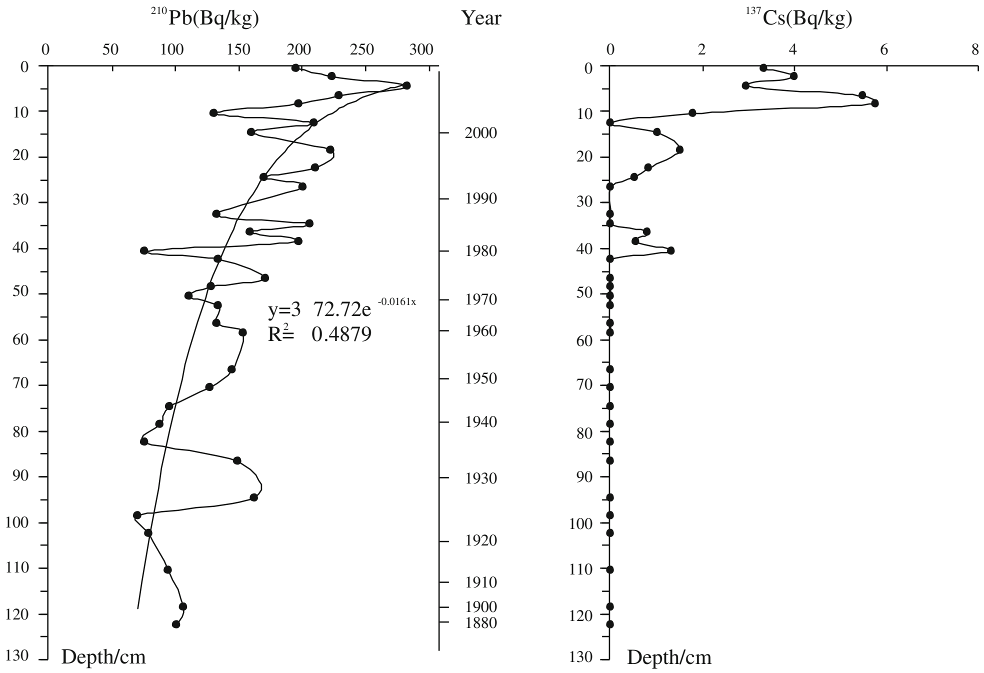

Fig. 3 Diagram of ^{137}Cs and ^{210}Pb activities versus depth of A5 sediment core

ocean. Runoff, tides and coastal currents as hydrodynamic forces usually impact the dilution, dispersion and transportation pathway of trace metals released via city sewage. A previous study showed that particle tracer released from the northern sewage was dispersed along the direction from northwest to southeast (Fig. 2), which roughly coincided with the prevailing movement of coastal currents (Zhang and Dong 1990). The distribution of trace metals in the surface sediment demonstrated that the transportation and dispersion pathway of trace metals followed the coastal currents, and the level of trace metals decreased from the north sites to the south sites. Coastal currents here played a major role for transporting the pollutants to offshore. For sites A4 and A5, the relatively higher trace metal enrichments would reflect somewhat the recent diffusion effects from the marine dumping zone which was put into service in 1988 with a purpose to collect the wastes from the Yantai harbour dredging, although it was believed to be minor by a previous study carried out in 2001 (Ji et al. 2003).

The distribution of trace metals in the core showed a significant increase since the 1980s (<40 cm) (Fig. 4). This result corresponds with the time of domestic sewage treatment plants being built. In the late 1990s, the municipal wastewater release to sea project of Yantai city was put into effect (Editorial office of Yearbook Yantai 1999), most industrial and domestic sewages of the city since then were drained to sea from the northeast after a preliminary treatment. It thus can be inferred that anthropogenic emissions here act as the major impact factor accounting for the high-trace metal concentrations in the surface sediments of the Yantai sea area.

Contamination risk evaluation

The potential ecological risk index for the given trace metals was employed to evaluate the intensity of trace metal pollution in the surface sediments. This is defined as (Hakanson 1980):

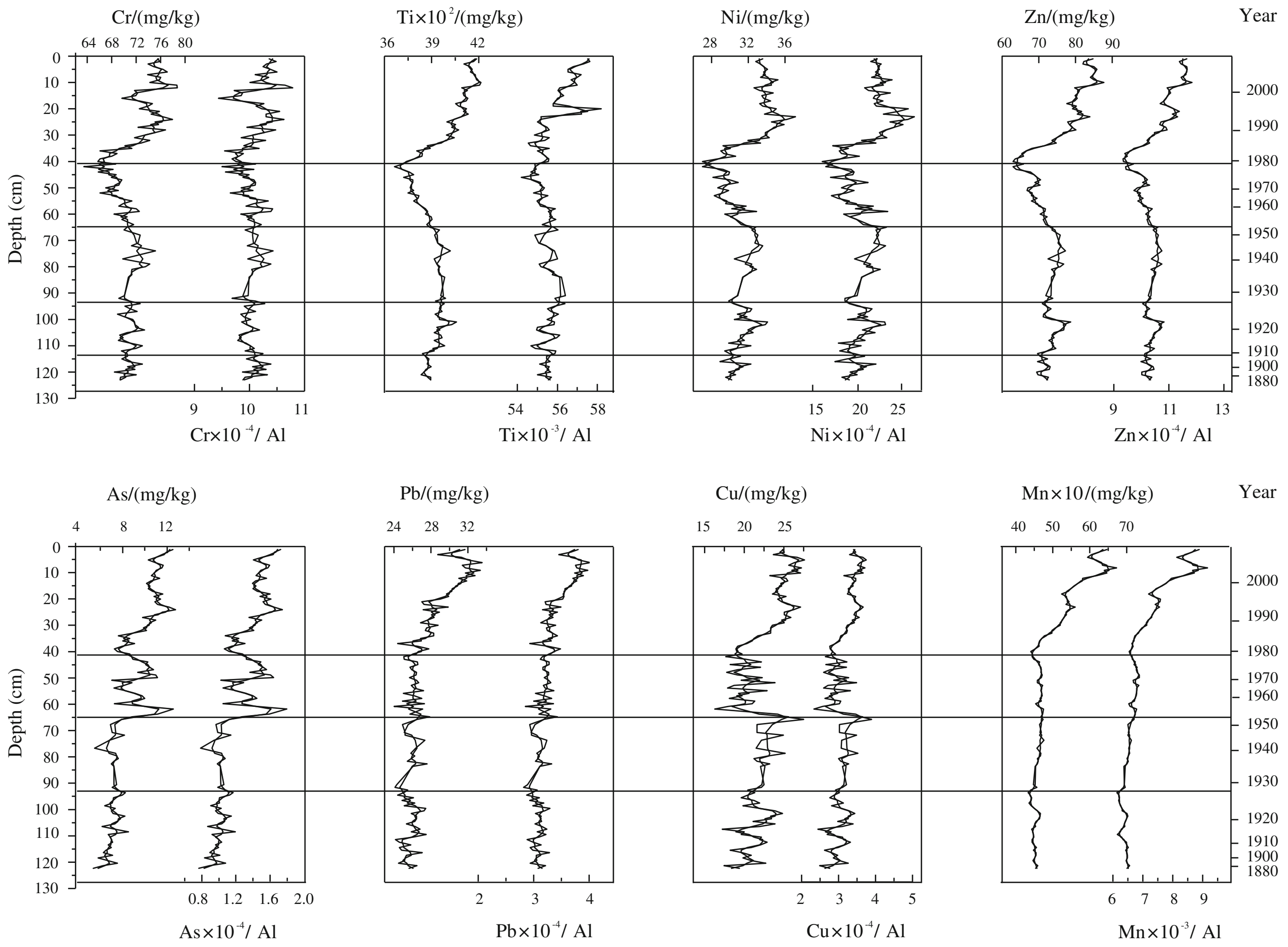

Fig. 4 Temporal variations of trace metals in the A5 sediment core covering the last ca. 100 years. The *thick solid line* is the trace metals data after 3-point average smoothing

$$E_r^i = T_r^i \times C_f^i$$

$$C_f^r = \frac{C_s^i}{C_n^i}$$

where E_r^i is the potential ecological risk index of an individual metal i, T_r^i is the toxic response factor provided by Hakanson (1980), and T_r^i is 5, 1, 10, 5 and 2 for trace metals Cu, Zn, As, Pb and Cr, respectively, C_f^i is the contamination factor of metal i, C_s^i is the concentration of metal i in superficial sediment, C_n^i is the background concentration of metal i. Here, C_n^i is deduced from the reports of coastline and integrated investigation of littoral zone resources in China and the given values are suggested 30, 80, 10, 20 and 80 mg/kg for metals Cu, Zn, As, Pb and Cr, respectively (Tang 1989).

The results are classified as $C_f^i < 1$, slight contamination, $1 < P_i < 3$ medium contamination, $3 < P_i < 6$ severe contamination, $E_r^i < 40$ low potential ecological risk, $40 < E_r^i < 80$ moderate potential ecological risk, $80 < E_r^i < 160$ considerable potential ecological risk, $160 < E_r^i < 320$ high potential ecological risk, $320 < E_r^i$ significantly high potential ecological risk (Hakanson 1980).

The contamination indices of the metals in the study area indicate that As (average $C_f^i = 1.17$) and Pb (average $C_f^i = 1.57$) show slight contamination currently, and need to be given particular attention in the future (Table 2). As a whole, Cu, Zn and Cr show less contamination except at sites close to the city sewage discharges. Presently, all the trace metals show low potential ecological risk in the study area (Table 2).

Temporal variations and the anthropogenic implications

Yantai city has been an industrial area since the late 1800s. As a sample of densely populated coastal cities, increasing anthropogenic emissions from industrialization and urbanization have become notable contributors of trace metals to the surrounding coastal ocean.

Table 2 Relative contamination (C_f^i) and potential ecological risk indexes index of trace metals in the surface sediments, Yantai coastal area

Sampling sites	Relative contamination indexes (C_f^i)					Potential ecological risk index (E_r^i)				
	Cu	Cr	Zn	As	Pb	Cu	Cr	Zn	As	Pb
A1	0.84	0.94	1.07	1.16	1.47	4.2	1.07	11.6	7.35	1.88
A2	1.07	0.98	1.18	1.33	1.65	5.35	1.18	13.3	8.25	1.96
A3	0.85	0.89	1.03	1.11	1.49	4.25	1.03	11.1	7.45	1.78
A4	0.76	0.88	0.97	1.14	1.32	3.8	0.97	11.4	6.6	1.76
A5	0.83	0.94	1.06	1.25	1.38	4.15	1.06	12.5	6.9	1.88
B2	0.85	0.84	0.96	1.09	1.31	4.25	0.96	10.9	6.55	1.68
B3	0.88	0.90	1.10	1.29	1.58	4.4	1.1	12.9	7.9	1.8
B4	0.61	0.87	0.80	0.86	1.31	3.05	0.8	8.6	6.55	1.74
C2	0.67	0.92	0.83	0.99	1.29	3.35	0.83	9.9	6.45	1.84
C3	0.85	0.86	0.95	1.11	1.44	4.25	0.95	11.1	7.2	1.72
C5	0.77	0.85	0.96	1.01	1.43	3.85	0.96	10.1	7.15	1.7
D3	0.32	0.86	0.54	0.49	1.47	1.6	0.54	4.9	7.35	1.72
D4	1.57	1.05	1.73	1.44	2.83	7.85	1.73	14.4	14.15	2.10
W1	1.00	0.95	1.11	1.32	1.46	5.00	1.11	13.2	7.30	1.90
W2	0.49	0.65	0.69	0.97	1.30	2.45	0.69	9.70	6.50	1.30
W3	1.13	0.95	1.15	1.34	1.53	5.65	1.15	13.4	7.65	1.90
W4	1.84	1.07	1.69	1.91	2.38	9.2	1.69	19.1	11.9	2.14
Average	0.90	0.91	1.05	1.17	1.57	4.51	1.05	11.7	7.84	1.81
Maximum	1.84	1.07	1.73	1.91	2.83	9.2	1.73	19.1	14.15	2.14
Minimum	0.32	0.65	0.54	0.49	1.30	1.6	0.54	4.90	6.50	1.30

To examine the anthropogenic impacts, temporal variations of trace metals and their ratios relative to Al were developed based on the Pb^{210} chronology of A5 core sediment (Fig. 4). The sediment core has presented the history of trace metal variations in the Yantai costal area since the late 1800s. Before the early 1950s, Cr, Ni, Zn, As, Pb, Mn and their ratios relative to Al showed slight increases and varied in narrow ranges except for Cu which exhibited an evident increment during late 1920s to the mid-1950s. As a whole, this suggested a relatively lower intensity of human activities in the Yantai area. Yantai has become the biggest wine-producing area in China since the 1900s; the increases of Cu in the sediment may reflect the wide application of Bordeaux mixture ($CuSO_4 + Ca(OH)_2$) as an insecticide in local grape planting during that time period.

The core sediment recorded a marked increase in As concentrations during the mid-1950s to the late 1970s, while other metals such as Cr, Ti, Ni, Zn and Cu exhibited a decline (Fig. 5). The low in trace metal concentrations roughly coincided with the economic stagnation of China during this period (Yantai Bureau of Statistics 2001) (Fig. 5), which resulted in the declines of industrial raw material consumption and the industrial waste discharges. Because agriculture was a major contributor to Yantai's economy before the late 1970s (Yantai Bureau of Statistics 2001) (Fig. 5), the coincidence of the first application of arsenic oxides, lead arsenate, and calcium arsenate in China as insecticides for the protection of crops at the early 1950s and the higher As concentrations occurrence in sediment suggested an inter-linkage. This causal connection can further be confirmed by high concentrations of As during the 1960s to 1970s and the mass usage of organic arsenic compounds, such as zinc methylarsenate, ammonium ferric methyl-arsenic and tuzet in agriculture throughout China (Wang 1981). Briefly, the trace metals of Cu and As in the coastal sediments served as diagnostic indicators of agriculture emissions intensity, especially the application of pesticides in crops before the late 1970s, and the alternative high Cu and As in the sediment implied a transition from application of inorganic copper sulfate insecticides to organic pesticides, e.g., the organic arsenical compounds.

Cr, Ni, Zn, Pb, Cu and Mn concentrations began to increase in the late 1970s and remained high before a slight decline occurred in the mid-1990s (except for As). The drop in As concentration in sediment here was probably the result of national policy which aimed at decreasing and banning the application of inorganic and some organic arsenic compounds in agriculture (Wang 1981). Meanwhile, economic statistics demonstrate that the industrial production began to hold the major position in local gross domestic production (GDP) after 1980 (Yantai Bureau of Statistics 2010) (Fig. 5). This would result in increased consumption of raw materials and discharge of industrial

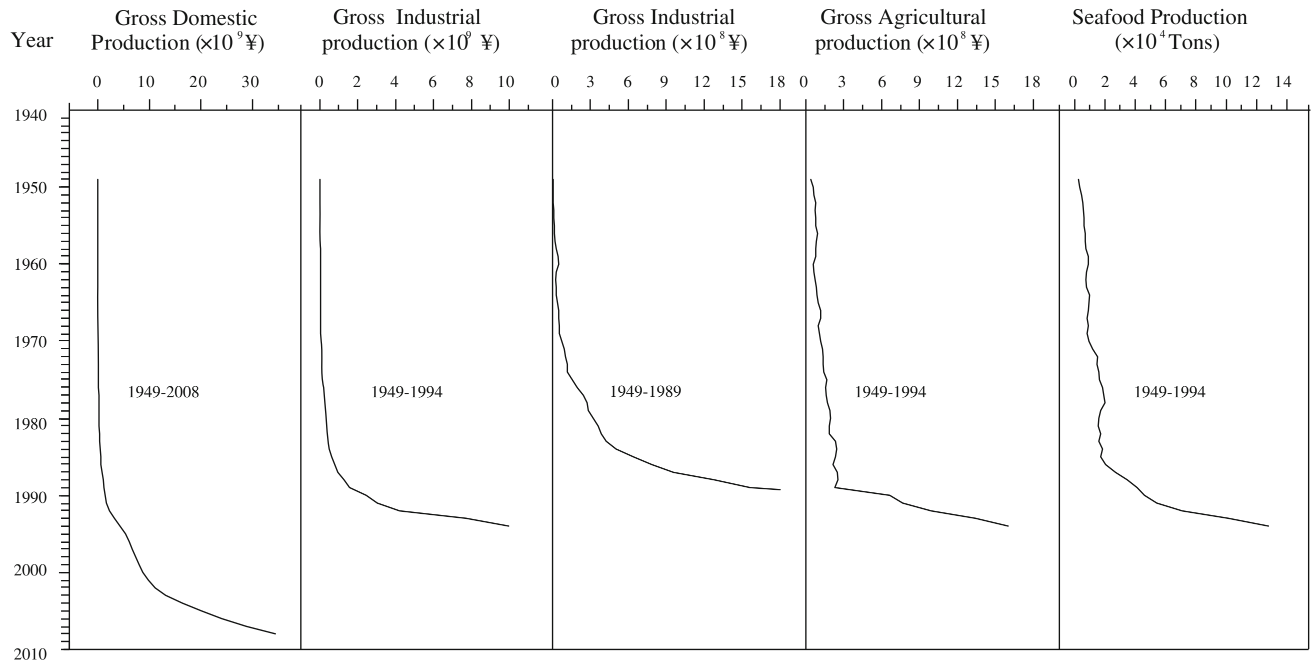

Fig. 5 The social development statistics data modified and intergraded from the statistical yearbooks of Yantai

wastes. It could be inferred that the high concentrations of trace metals in the sediment after 1980 indicate the increasing contribution of industry emissions to the coastal environment. Further the slight decline of trace metals in the sediment during the mid-1990s would reflect a slowing in the increases of economic activities in the Yantai area as revealed by the GDP data (Fig. 5). As a whole, all the trace metals showed evident upward accumulations from ca.1998 to 2008 corresponding to a boom in the economy of Yantai city. Although measures have been taken to treat the industrial and city wastes emissions since the late 1980s, the high concentrations of trace metals reflect the increasing impacts of anthropogenic processes related to the industrialization of Yantai city.

Conclusions

Trace metal characteristics in the sediment of Yantai coast demonstrate the impact of anthropogenic processes, spatially and temporally. Spatial distributions of trace metals in sediments suggest that city sewage discharges are major sources of contamination metals for the coastal area at present, and the prevailing orientation of coastal currents have visible impacts on the dispersion, transportation of trace metals from the city sewage discharges. Temporal variations of trace metals in the coastal sediments (as demonstrated by one sediment core) potentially reflect the changes in anthropogenic processes, especially the industrialization and urbanization of the city from a weak interference to an increasing intensity over the last ca. 100 years. The high concentrations of trace metals Cu and As in the sediment before the late 1970s largely indicated emissions from the application of pesticides, while high concentrations of several metals since the 1980s are primarily the results of the rapid industrialization of the study area. Although the potential ecological risk evaluation suggests that all the trace metals are in low potential ecological risk at present, the history variations of trace metals brings on some concerns on future environment protection and the establishment of harmony relations between human and the environment.

Acknowledgments The authors thank the anonymous reviewers for their constructive suggestions. This work was supported by CAS Innovative Programmer (No. KZCX2-YW-Q07-04), National Science Foundation of China (NSFC No. 40976097, 40872211) and the Ministry of Science and Technology of China (Grant No. SKLEC-2008KYYW02).

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