Supplementary Materials

Table S1 Conditions for microwave digestion of sediment samples (MARS Xpress; CEM, Matthews, NC, USA)

Stage	Power		Ramp (min)	Temperature (°C)	Hold (min)
	Max (W)	Percentage			
1	1600	100%	6	120	2
2	1600	100%	5	150	5
3	1600	100%	5	180	20

Assessment methods

Pollution Load Index

The pollution load index (PLI) was proposed by Tomlinson et al (Tomlinson et al., 1980). For the entire sampling site, PLI has been determined as the nth root of the product of the *n* C^i_f:

$$\mathrm{PLI} = \sqrt[n]{C^i_{f1} \times C^i_{f2} \times C^i_{f3} \times \cdots \times C^i_{fn}}, \quad (3)$$

where C^i_f is the single element pollution factor and the values can be calculated by the equation as suggested by Hakanson (Håkanson, 1980).This empirical index provides a simple and comparative means for assessing the level of heavy metal pollution, and Table S2 shows the factor standard of different levels (Varol, 2011).

Potential ecological risk

The potential ecological risk (PER) method developed by Hakanson (Håkanson, 1980), involves assessing the pollution status and the associated toxicological effects to determine a representative index level. The equations for calculating the PER indexes are as follows:

$$E^i_r = T^i_r \times C^i_f = T^i_r \times (C^i_s / C^i_n), \quad (4)$$

$$RI = \sum_{i=1}^{n} E_r^i \ , \quad (5)$$

where C^i_s is the content of the element in samples, C^i_n is the background value of the element, C^i_f is the single element pollution factor, E^i_r is the PER index of an individual element, and T^i_r is the biological toxicity factor of an individual element, which are defined as Cd=30, Cr=2, Cu=Ni=Pb=5, and Zn=1 (Guo et al., 2010). *RI* is the comprehensive PER index, which is the sum of E^i_r. Table S2 shows the factor standard of different levels.

Risk assessment code

The risk assessment code (RAC) assesses the availability of metals in solution by applying a scale to the percentage of sediments that can reduce metals in the exchangeable fraction (B1). The classification is tabulated in Table S2 (Perin et al., 1985; Singh et al., 2005).

Consensus-based sediment quality guidelines

Consensus-based sediment quality guidelines (SQGs) method were developed by Macdonald et al. (MacDonald et al., 2000). There are two consensus-based values: threshold effect concentration (TEC), below which adverse effects are not expected to occur; and probable effect concentration (PEC), above which adverse effects are expected to occur more often than not (Niu et al., 2009; Ma et al., 2013). The mean PEC quotient ($Q_{m\text{-}PEC}$) was introduced to predict the toxicity of sediment samples, which can be calculated as the following equation:

$$Q_{m-PEC} = \overline{\sum C_n / PEC_n} \ , \quad (6)$$

where C_n is the measured concentration of heavy metals and PEC_n is the corresponding PEC. The PEC benchmark values for Cd, Cr, Cu, Ni, Pb, and Zn in sediments are 4.98, 111, 149, 48.6, 128 and 459 mg/kg, respectively. The evaluation criteria for $Q_{m\text{-}PEC}$ are listed in Table S2.

Toxic units (TUs)

Toxic units (TUs) are defined as the ratios of the detected contents to the probable effect level (PEL) values (Pedersen et al., 1998). The sum of the toxic units ($\sum$TU) can be

used to assess the potential toxicity of heavy metals in sediments, and calculated by the following equation:

$$\sum TU = \sum_{i=1}^{n} \frac{C_T^i}{C_{PEL}^i}, \quad (7)$$

where C^i_T and C^i_{PEL} are the total content and the PEL of metal i, respectively. PEL values represent the thresholds of chemical contents above which adverse effects are likely to occur. The PEL values for marine and estuarine ecosystems were referenced in this study (Long and MacDonald, 1998).

Table S2 Criteria of different assessment methods

Pollution load index (PLI)			
PLI	**Pollution level**		
≤ 1	No pollution		
> 1	Existing pollution		
Potential ecological risk (PER) index			
E_i^j	**PER of individual element**	**RI**	**Comprehensive PER**
<40	Low	≤150	Low
40–80	Moderate	150–300	Moderate
80–160	Considerable	300–600	High
160–320	High	>600	Serious
≥320	Very high		
Risk assessment code (RAC)			
Percentage of B1 (%)	**Risk**		
<1	No		
1-10	Low		
10-30	Medium		
30-50	High		
≥50	Very high		
Consensus-based SQGs			
$Q_{mi\text{-}PEC}$	**Sediment quality**		
≤ 0.5	Not toxic		
> 0.5	Toxic		

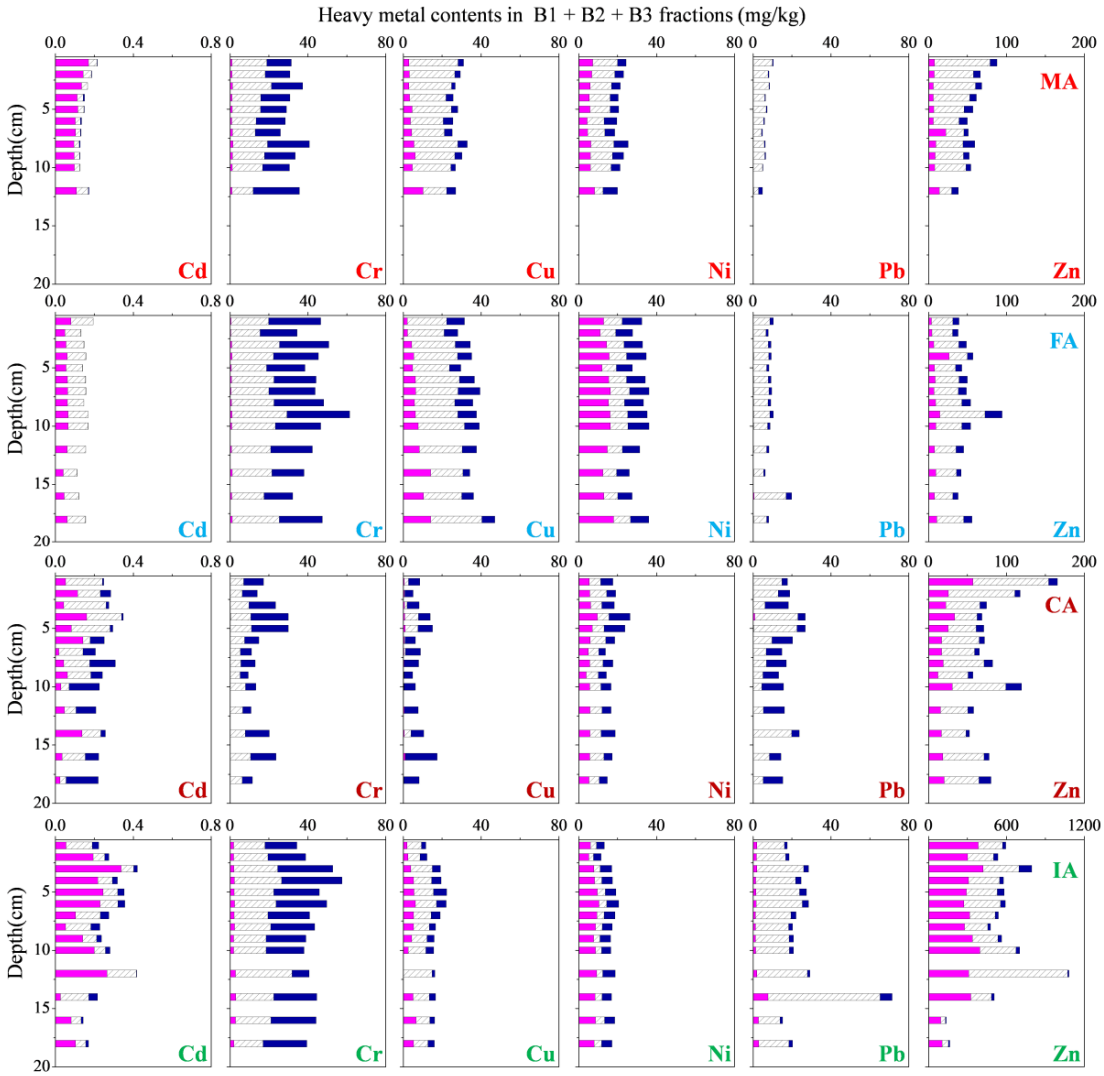

Fig. S1 Spatial-vertical distribution of heavy metals in B1+B2+B3 fractions in riverine sediment cores from different land-use areas (MA, mountain area; FA, farm area; CA, city area; IA, industrial area).

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