Accumulation and Potential Ecological Risk of Heavy Metals in the Sediments of Rivers System in Beijing-Tianjin Area

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Research Article

Keywords: Sediment, heavy metals, distribution, ecological risk, Beijing-Tianjin area

Posted Date: September 17th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-805182/v1

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Abstract

Human activities cause heavy metals to enter the water body and consequently deposit in sediment with slow flow velocity, however little studies have explored the spatial distribution and ecological risk of heavy metals in sediments. The risk, spatial distribution and toxicity of heavy metals in sediment were investigated along the North Canal in the Beijing-Tianjin area. The study revealed that the mean concentrations of heavy metals in sediments exhibited a descending order of Zn > Cr > As > Cu > Pb > Ni > Co > Cd. The average geoaccumulation index (Igeo) value of Cd was highest and ranged from 0.2 to 2.91. Moreover, the greatest contamination of Cd (the Igeo values > 2) was observed in three sampling sites around the Tianjin City. The pollution load index (PLI) of all sampling points were greater than 1, which indicated the inflow of heavy metals originated from anthropogenic sources. The risk index (RI) values of three sampling points were greater than 300, which demonstrated high potential ecological risk. With regards to the toxicity assessment of combined heavy metals, there were two probable effect concentration (PEC) quotient (Qm-PEC) values greater than 0.5 which suggested potential toxicity to certain sediment-dwelling organisms. Identification of the possible sources and factors contributing to the content and spatial distribution of heavy metals could assist in improvement of the water quality, as well as support efficient management strategies to restoration of the environment.

Introduction

The contamination of heavy metal in the urban riverine sediment have attracted much recent attention, which is the result of quick industrialization and urbanization across the globe (Yu et al., 2011; Islam et al., 2015; Martínez-Santos et al., 2015). Metals, such as Fe, Zn, and Cu, are crucial components in the metabolism of biota in the earth's crust. Moreover, heavy metals are widely distributed in the environment, and sediments and river water are usually important sink for heavy metals. However, due to their extensive detection in the food chains and associated toxicity to biota and the environment, heavy metals posed ecological risks and became an deep concern globally (Deng et al. 2014, Tagliaferro, et al., 2018, Zahra et al. 2014). Therefore, in the purpose of controlling and maintaining the water system, a comprehensive understanding of the levels, source, and spatial distribution of heavy metals in sediment is important. In order to achieve this, ecological risk analysis is a useful techniques for monitoring the potential problem of heavy metal pollution and the corresponding threats to the ecosystem, especially in wetlands (Wang et al. 2015).

Heavy metals in various chemical forms commonly have varying chemical stabilities and biological availabilities, thereby contributing to different ecological risks. One of the commonly used extraction procedure to assess chemical speciation of heavy metals is the European Community Bureau of Reference (BCR) standard method (Gao and Chen, 2012). The bioavailability of one specific metal is associated with the active fraction, which is its exchangeable, reducible, oxidizable portions, and the remainder is not bioavailable and maintains long-term stability (Yin et al., 2014; Rosado et al., 2016). Chemical components and statistical analysis have been extensively conducted to determine the pollution properties and bioavailabilities of heavy metals in sediment. In different reservoirs, the spatial
distribution and concentration of heavy metal are closely associated with the anthropogenic activities, depending on the hydrodynamic conditions, type of sediment and metal sources (Wang et al., 2014, Noronha-D’Mello and Nayak, 2016, Wang et al., 2018). The migration of metals to hydrophytic ecosystems could be observed from either natural elements such as lithologic structure or human disturbances. Moreover, the shape and morphology of the water system would significantly affect the deposition of heavy metals (Sjojka et al., 2018). Through investigation of heavy metal in river system, the natural and anthropogenic origin of heavy metals could be identified and the level of ecological risk could be determined accordingly. Ecological risk analyses could be employed in decision-making processes to detect environmental concerns and manage ecosystem health.

Beijing-Tianjin area is one of northern China’s most developed and populated areas. Water bodies in urban areas receive wastewater from various sources, such as sewage disposal plants, on-point source pollution, and unprocessed industrial effluent. This results in remarkable deterioration of the aquatic environment with rapid social and economic development. Besides, the urbanization process in this area also severely affects the deposition rates of organic matter (OM) and acid volatile sulfide, which further influences the heavy metal distributions and chemical patterns (Hong et al., 2010; Strom et al., 2011). Recently, increasing concerns on the potential adverse health effects of pollution are being reported, since river water is the main agricultural irrigation water resource. The pollution status of heavy metals in sediments around this area have been investigated, focusing on the measurement of the total amount of metals in sediments (Liu et al., 2009; Tang et al., 2013). However, the mineral contents and mobility of sediments varies in the river system through different geographic locations and seasons, which influence the bioavailabilities and toxicities of heavy metals in the sediments indirectly (Huang et al., 2012; Zhang et al., 2014). The extent of heavy metals in sediments could provide vital information for assessing the state of the environment. Therefore, it is necessary to monitor the components, bioavailability, and toxicity of heavy metal along with the principle river system and its tributaries from Beijing city to Tianjin City. The main objective of this research was to determine the spatial distribution of metals in sediments and evaluate the ecological risk with different models (SQGs, EF, and Igeo). By identifying the potential sources and factors determining the content and spatial distribution of heavy metals, the related environmental protection could be proposed to improve the water quality in Beijing-Tianjin area.

**Methods And Material**

**Sampling site description and sampling procedure**

The North Canal belongs to the Haihe River system. It flows southward from the southern foot of Yanshan Mountain in Changping District of Beijing to the Tongzhou District. At the upstream end of Beiguan gate in Tongzhou District, it is known as Wenyu River, and it flows through Xianghe County of Hebei Province, Wuqing District of Tianjin city, and joins Haihe River at dahongqiao of Tianjin city. It is the main sewage system in Beijing, accounting for 76% of the total sewage discharge in Beijing (Yang et al. 2021).
In April 2019, ten sediment samples were collected along Beisha River (BsR), Wenyu River (WR), Beiyun River (ByR), and Haihe River (HR) (see Fig. 1). Triplicate samples were collected at each site using handheld PVC corers (150 cm × id 80 mm) to fully represent the effects of land types on sediment quality. Individual sediment sample was packaged in a polyethylene plastic bag, which was previously rinsed by acid, and then air-dried to remove excess water. Finally, all the samples were freeze-dried, ground and sieved using a pre-cleaned 100-mesh sieve before analysis.

**Chemical analysis**

Samples were firstly digested with hydrofluoric and perchloric acids by microwave in Teflon vessels (Marsx press, CEM), and the obtained solution was stored at 4°C for the analysis of heavy metals. An Optima 2000DV inductively coupled plasma optical emission spectrometer (ICP-OES) (Perkin Elmer, Waltham, MA, USA), and an inductively coupled plasma mass spectrometer (ICP-MS) system (7500a Agilent Technologies, Santa Clara, CA, USA) were employed to measure the content of heavy metals in the solution, with detection limit of 0.003–0.050 mg/L and 0.025–0.200 µg/L, respectively. Certified reference material GBW07401, purchased from the Chinese Environmental Monitoring Center, was used to test the precision and accuracy of metals (i.e. As, Cr, Co, Cu, Ni, Pb, Cd, and Zn). Overall, the recoveries varied in the range of 91–102%, and the relative standard deviations (RSD) in all samples were lower than 3%.

**Relevant assessment and statistical analysis**

Geoaccumulation index ($I_{geo}$), shown as Eq. 1, have been extensively used to determine the metal contamination by indicating the accumulation of metals in sediment (Muller, 1969).

$$I_{geo} = \log_2 \left( \frac{C_n}{K \times B_n} \right)$$ (eq. 1)

where $C_n$ and $B_n$ are the the calculated concentrations and background concentrations of heavy metals, respectively, and $K$ is the background matrix correction factor ($K = 1.5$). By taking the non-polluted soil as the baseline, $I_{geo}$ could be used to quantitatively measure the degree of metal enrichment or contamination and classified into seven levels (Table 1).
| Criteria                              | Description                                      |
|--------------------------------------|--------------------------------------------------|
| **Geo-accumulation index ($I_{geo}$)** |                                                  |
| $I_{geo}$                            | Pollution level                                  |
| $\leq 0$                             | Unpolluted                                       |
| 0–1                                  | Unpolluted to moderately polluted                |
| 1–2                                  | Moderately polluted                              |
| 2–3                                  | Moderately to strongly polluted                  |
| 3–4                                  | Strongly polluted                                |
| 4–5                                  | Strongly to very strongly polluted               |
| $>5$                                 | Very strongly polluted                           |
| **Enrichment factor (EF)**           |                                                  |
| EF                                   | Heavy metal source                               |
| 0.5–1.5                              | Crustal materials or natural weathering processes|
| $>1.5$                               | Non-crustal or anthropogenic processes           |
| **Contamination factor (CF)**        |                                                  |
| CF                                   | Pollution Contamination                          |
| $<1$                                 | Low contamination                                |
| 1–3                                  | Moderate contamination                           |
| 3–6                                  | High contamination                               |
| $>6$                                 | Very high contamination                          |
| **Pollution load index (PLI)**       |                                                  |
| PLI                                  | Pollution level                                  |
| $\leq 1$                             | No pollution                                     |
| $>1$                                 | Existing pollution                               |
| **Consensus-based SQGs**             |                                                  |
| $Q_{m-PEC}$                          | Sediment quality                                 |
| $\leq 0.5$                           | Not toxic                                        |
| $>0.5$                               | Toxic                                            |
Geo-accumulation index ($I_{geo}$)

| Potential ecological risk (PER) index |
|--------------------------------------|
| $E_i$ | PER of individual elements | 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 |

Enrichment factor (EF) is another extensively employed index to evaluate the potential anthropogenic inputs of observed metals. The EF values of specific heavy metals could be estimated using the Eq. 2 to assess the sources and temporal variation of metals (Bhuiyan et al., 2010):

$$EF = \frac{C_n (\text{sample})}{C_{Al} (\text{sample})} \div \frac{B_n (\text{baseline})}{B_{Al} (\text{baseline})} \quad (\text{Eq. 2})$$

where $C_{Al}$ and $B_{Al}$ are the calculated concentration and background concentration of Al in the sediments, respectively. The soil background values were adopted as the baseline values in this study.

The potential ecological risk (PER) index is commonly used to determine the level of heavy metals in sediments, and shown as Eq. 3 (Håkanson 1980):

$$E_i^r = T_r^i \times C_f^i = T_r^i \times (C_s^i \div C_n^i) \quad (\text{Eq. 3})$$

where $E_i^r$ is the PER index of an individual metal, $C_f^i$ is the single metal pollution factor. $C_s^i$ and $C_n^i$ are the level of the heavy metal, and the reference value of the metal, respectively. $T_r^i$ is the biological toxic factor of an individual metal, where 30 was used for Cd, 2 was used for Cr, 5 was used for Cu, Co, Ni, Pb and 1 was used for Zn in this study (Guo et al., 2010).

$$RI = \sum_{i=1}^{n} E_i^r \quad (\text{eq. 4})$$

Risk index (RI), the sum of $E_i^r$, reflects the sensitivity of the biological community to the hazardous chemicals, aiming to illustrate the comprehensive PER caused by the overall contamination.

Single contamination factor (CF) was calculated by Eq. 5:

$$CF = \frac{\text{Metal concentration}}{\text{Metal background value}} \quad (\text{eq. 5})$$
The obtained data were used to test if there were any anthropogenic impacts. The pollution load index (PLI) was used to determine the quality of the sediments (Suresh et al., 2011):

\[
\text{PLI} = (\text{CF}_1 \times \text{CF}_2 \times \ldots \times \text{CF}_n)^{1/n} \quad (\text{Eq. 6})
\]

CF is the contamination factor and n is the number of elements used.

To assess the effects of multiple heavy metals, the mean probable effect concentration (PEC) quotient \((Q_{m-\text{PEC}})\) was proposed and be calculated using the following equation:

\[
Q_{(m-\text{PEC})} = \sqrt{\left( \sum_{i=1}^{n} C_n / \langle \text{PEC} \rangle_n \right)^n} \quad (\text{eq. 7})
\]

where \(C_n\) is the measured concentration of one heavy metal and \(\text{PEC}_n\) is the corresponding PEC value.

The PEC benchmark values for As, Co, Cd, Cr, Cu, Ni, Pb, and Zn are 30, 4.98, 111, 149, 48.6, 128, and 459 ug/g, respectively.

**Results And Discussion**

**Heavy metal concentrations and the property of sediment**

The characteristic concentrations of selected heavy metals (As, Cd, Co, Cr, Cu, Ni, Pb, and Zn) in the sediments from 10 sites were shown in Fig. 2. The distributions and mean concentrations of heavy metals in different locations varied greatly due to various factors, following a descending order of Zn > Cr > As > Cu > Pb > Ni > Co > Cd. Zn was detected in maximum concentration (199.2 ug/g) at the sampling points at S2, which was close to the intersection of WR and Ba River, and in minimum concentration (55.0 ug/g) at S9. This result indicated that Zn could probably be hazardous to aquatic biota in the study area, since it could be remobilised when environmental factors alter. Cr was found to peak at around 86–90 ug/g (S9 and S10) near the estuary of the HR and to plunged to 40.4 ug/g at the preceeding point S8. The highest and lowest concentration of arsenic were found at S10 and S9, 42.1 and 27.0 ug/g, respectively. Cu reached its maximum of 46.2 ug/g at S2 and its minimum of 15.3 ug/g at the sampling point S4. Pb concentration at the highest level was at S7 (average 57.5 ug/g), while the lowest values were observed at S3 and S4 (average 14.1 ug/g). A similar phenomenon, which was the sudden increment of Pd concentration at the sampling point S7, was also observed in the case of Cu, Zn, and Ni concentrations, probably resulted from the extra input by passing Tianjin City. The average concentrations of Ni were quite even, varied between 22.9 and 41.8 ug/g in all samples. However, the significant increase of Co concentration from the minimum of 7.4 ug/g at S7 to the maximum of 19.2 ug/g at S9 was observed, closely relating to the great variation of Co concentration at S8. Finally, the maximum and minimum of Cd were 1.0 ug/g at S6 and 0.1 ug/g at S4. Another significant tendency was that the concentrations of all detected heavy metals were remarkably raised at the last one or two sampling points, possibly due to the accumulation of all heavy metals in the estuaries of HR.
In terms of other metals (Fig. 3), Fe was found to be the highest level of concentration at all sampling points and almost maintained the same concentration around 30000 ug/g. Al was measured in maximum concentration (25800 ug/g) at the sampling points S6 near ByR and in minimum concentration (4900 ug/g) at S1. In comparison, Mn was found to vary between 180 ug/g and 865 ug/g in the whole study area. For Ca and Mg, the concentrations suddenly increased at the S5 position, which suggested that the external input from some location between S4 and S5.

**Correlation analysis between heavy metals and properties of sediments**

In this study, Pearson's correlation matrix analysis was performed to distinguish possible sources of metals and their transport mechanisms, and the strong positive correlations were recognized between a number of specific metals (Fig. 4). The mobility of the heavy metals was an effective indicator to evaluate the potential risk strongly connected to the environmental characteristics. Figure 4 revealed the relationship between the heavy metals and the sediment properties. The red and blue color represented the positive and negative correlation, respectively, and relatively increased as color intensity increased. Organic materials commonly play an important part in activating the transport of metals, therefore the correlations between organic materials and metals were clearly presented between SOM and most heavy metals. For the acidic property of the sediment, negative correlation was established between pH value and almost all heavy metals, except Co and Cr. Meanwhile, strong positive correlations were extensively found between Cd and Pd, Co and Cr, Cd and Zn, Cu and Ni, Cu and Pb, Cu and Zn, and Pb and Zn. According to the above result, it is easy to find that the most metals were positively correlated with each other, except As and Al.

**Heavy metal pollution in the sediments**

The enrichment factor (EF) was normally applied to assess the sources of heavy metals (Fig. 5). In most cases, the values less than 1.5 suggested that the heavy metals probably originated from natural weathering processes, and more than 1.5 suggested that the heavy metals were delivered by other sources, such as point and non-point pollution (Hanif et al., 2016). Depending on the lithologic background values, the order of EF values was as follows: Cd (5.33) > As (3.49) > Zn (1.15) > Cu (1.14) > Ni (0.96) > Pb (0.95) > Cr (0.87) > Co (0.68). In the whole testing area, enrichment was detected as significant for Cd and As, as moderate for Cu and Zn, and as low for Ni, Pb, Cr, and Co. The data of the EF values for Cr, Co, and Ni did not exceed a value of 1.5 at all sampling points. The enrichment of Ni was probably due to the mining quarry and Cr was thought to be derived from agriculture in the basin, household wastes, and plants using metal raw materials. For Pd, accumulators, batteries, vehicle fuels, and paints were usually considered to be the most vital sources, and its enrichment reached the highest value (1.90) at S7 point. This indicated that it was probably delivered by other sources, such as point and non-point pollution (Ma et al., 2013, Niu et al., 2019). The main anthropogenic sources of significantly enriched As and Cd almost in all sampling points were probably the chemical fertilizers, since both As and Cd were
the main component of phosphate fertilizers and could be removed by water during the agricultural activity and lithologic formations (Bolat and Kara, 2017)

In addition, the values of $I_{\text{geo}}$ also indicated the similar descending level of pollution (Fig. 6): Cd (1.35) > As (1.16) > Cu (-0.53) > Zn (-0.58) > Ni (-0.67) > Pb (-0.76) > Cr (-0.82) > Co (-1.20). The $I_{\text{geo}}$ values for Co, Cr, and Ni were below zero at all sampling points and could be classified as unpolluted in this study area. Meanwhile, the $I_{\text{geo}}$ values for Cu, Zn, and Pd were also lower than zero except for two or three sampling sites, which indicated that small local contamination of bottom sediments with Cu and Zn at S2 and S10, and with Pd and Zn at S7 and S10. This phenomenon suggested that these three sampling points should be focused on the external input of heavy metals, as well as the local pollution source of Cu, Zn, and Pd.

The $I_{\text{geo}}$ values for As were mostly concentrated between 1 and 2 in all samples, reflecting the weak contamination of As in the whole study area. The main anthropogenic sources of As were chemical fertilizers, geothermal sources, and fossil fuel use (Atabey, 2009). The average $I_{\text{geo}}$ value of Cd was the highest, which ranged from 0.2 to 2.91, and the largest contamination of Cd (the $I_{\text{geo}}$ values > 2) was noted in three sampling sites located around Tianjin City. The sediments in sampling sites S4 and S8 demonstrate unpolluted conditions with the value of $I_{\text{geo}}$ near zero.

### Heavy metal risk and toxicity in the sediment

The global index of PLI was extensively used to assess the bottom sediments’ pollution with heavy metals. It was surprising to notice that the PLI values of all sampling points were higher than 1, which indicated the inflow of heavy metals from anthropogenic sources (Fig. 7). The highest value of PLI was recorded at sampling S7, located in Tianjin City, and all values of PLI were close to the average value, which was the most concerning as the whole area faced the current situation of pollution. Similar to the PLI value, the risk index (RI) was also calculated and represented the sensitivity of the biocenosis to the toxic species. Clearly, the RI values of three sampling points (S5, S6, and S7) were higher than 300, exhibiting high potential ecological risk. Meanwhile, the RI values between 150 and 300 were observed at another three locations, where were facing moderate risk. For the toxicity assessment of combined heavy metals, a consensus-based sediment quality guideline (SQG) was employed, and the related $Q_{m-P\text{EC}}$ data were presented in Fig. 7, which ranged from 0.31 to 0.58. Although the difference in the values of $Q_{m-P\text{EC}}$ was not serious, there were two $Q_{m-P\text{EC}}$ values higher than 0.5 which were noticed at S7 and S10. This suggested that the heavy metal could be toxic to certain sediment-dwelling organisms (Ji et al., 2019).

### Conclusions

In this study, heavy metal pollution of sediments at 10 sites along Beisha River (BsR), Wenyu River (WR), Beiyun River (ByR), and Haihe River (HR) was evaluated, mainly focused on the extent of contamination of sediments by trace metals. The mean PEC quotients indicated a moderate to high toxicological risk in terms of bioavailability, with the potential toxicity reaching in the middle. The existent of Cd, Pb, and Zn in the present study area were the extensive use of phosphate fertilizer during agricultural intensification,
while Cu, Cr and Ni represented the natural source. Base on the background values, three sampling points exhibited high potential ecological risk, and another three faced moderate risk. The information from this study could be used in developing effective management strategies to control the heavy metal pollution in the Beijing-Tianjin area.

**Declarations**

**Ethics approval and consent to participate:** Not applicable

**Consent for publication:** Not applicable

**Competing interest:** The authors declare that they have no competing interests.

**Availability of data and materials**

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

**Contributions**

Conceptualization: SGY, WQZ; sample collection: WYL, MSW; measurement: SGY, WYL, ZHL; writing, original draft: SGY; reviewing and editing: SGY, WQZ, BQS. All authors read and approved the final manuscript.

**Acknowledgments**

This work was supported by the Youth Innovation Promotion Association CAS, Chinese Academy of Sciences (2018058); the National Major Science and Technology Program for Water Pollution Control and Treatment (2017ZX07107-004); the Key-Area Research and Development Program of Guangdong Province (2019B110205002).

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Figures

Figure 1

Location of samples in Beijing-Tianjin area.
Figure 2

Concentration of heavy metals of samples.
Figure 3

Concentration of heavy metals in all samples
Figure 4

Correlation analysis between heavy metals and properties of sediments
Figure 5

EF of heavy metal in the sediments
Figure 6

Igeo of heavy metal in the sediments
Figure 7

PLI, RI, and Qm-PEC of heavy metal