Comprehensive analysis of mercury pollution in the surface riverine sediments in the Haihe Basin, China

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Abstract The Haihe Basin extends over 318,000 km² or 3.3% of the total area of China, has a population of over 120 million, and is one of China's most polluted river basins. We investigated the pollution and risks from mercury (Hg) in surface riverine sediments of this basin and found that total Hg concentrations in surface sediments ranged from 0.001 to 1.05 mg/kg and averaged 0.054 mg/kg. Using the modified European Community Bureau of Reference standard method, we found that the exchangeable/acid-soluble, reducible, oxidizable, and residual fractions of Hg accounted for 9.86, 6.54, 11.0, and 72.6% of total Hg, respectively. The riverine sediments in the Da Qing He watershed had the highest anthropogenic enrichment of Hg (enrichment factor >1.5). There was a high potential ecological risk from Hg in the riverine sediments in the upper reaches of the Bei San He and Da Qing He watersheds, the middle reaches of the Luan He and Hei Long Gang watersheds, and the lower reaches of the Zi Ya He watershed. Comparison with risk assessment code showed that the riverine sediments in the Da Qing He, Luan He, and

Tu-Hai Ma-Xia He watersheds represented a medium risk, with 16.6, 11.9, and 11.8% of Hg in the exchangeable/acid-soluble fraction, respectively.

Keywords Mercury · The Haihe Basin · Contamination · Riverine sediments

Introduction

The contamination of sediments with trace metals that has accompanied rapid economic development in many countries has become a serious environmental issue worldwide (Ryu et al. 2016; Zhang et al. 2015). The concentrations, speciation, sources, and toxicity of trace metal pollutants in sediments have been studied extensively in recent years (Chakraborty et al. 2014; Sundaray et al. 2011). Of all the trace metals, mercury (Hg) has received particular attention because of the high bioaccumulation potential and toxicity of its methylated form (Leipe et al. 2013; Ma et al. 2013). Trace metal pollution in aquatic sediments has been studied intensively to facilitate effective management of aquatic ecosystems at a range of scales (Chen et al. 2015; Tang et al. 2013). Of these studies, many have considered the distribution of Hg in aquatic environments (Jackson 2013); to date, however, most previous studies have focused on individual catchments or rivers (Hodson et al. 2014; Wu and Cao 2010), and there have been few studies of Hg pollution in large river basins.

The Hg is ubiquitous worldwide and occurs in the environment in various forms including Hg(0)-elemental Hg, inorganic Hg(I), and Hg(II) species, and a series of organic forms (Leopold et al. 2010). Hg forms in sediments have been extensively studied (Horvat et al. 2003; Qiu et al. 2005), but there have been few studies of the chemical fractions of Hg (Sahuquillo et al. 2003; Yang et al. 2009), especially in large

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basins. It has been pointed out that chemical fractions should be used to study the mobility and bioavailability of Hg rather than total Hg (Islam et al. 2015), and environmental risk evaluations or remediation strategies should be based on individual species of Hg or their binding to different phases of the matrix rather than the total concentrations of Hg (Sahuquillo et al. 2003). The retention and accumulation of Hg in sediment are closely linked to the chemical form of the metal, and to the variable environmental conditions, such as the oxygen content or the redox conditions. Hg distribution in sediments is mainly governed by the high stability of its associations with sulfide and high molecular weight organic matter (Biester et al. 2000; Sahuquillo et al. 2003). Consequently, the mobility of Hg in sediments can be assessed from the chemical fractions determined in the sequential extraction procedure developed by the Bureau of Reference of the European Community.

The standard method of the European Community Bureau of Reference (BCR) is a sequential extraction that has been used widely to determine the chemical speciation of heavy metals (Arain et al. 2008; Gao and Chen 2012). The bioavailability of a metal is related to the concentrations of the exchangeable/acid soluble (B1), reducible (B2), and oxidizable fractions (B3); the residual fraction (B4) is not bioavailable and remains stable for long periods (Rosado et al. 2016; Yin et al. 2014; Zhang et al. 2014). Heavy metals in the B1 fraction of sediments are considered unstable because they can be released to the aqueous phase, where they become more bioavailable (Gao and Chen 2012). The contribution of the B1 fraction to the total concentration can be determined from the risk assessment code (RAC), which is widely used to estimate the bioavailability of and risks posed by heavy metals in sediments (Passos et al. 2010; Singh et al. 2005). A risk assessment code based on the B1 fraction of Hg^{2+} can be used to evaluate the mobility and bioavailability of Hg in sediments. For example, an increase in Hg mobility may result in uptake of monomethyl Hg (MeHg) into organisms via predation. On the other hand, the B4 fraction of heavy metals (including Hg) is generally chemically stable, only slightly toxic, and is not available for uptake by organisms.

The Haihe Basin is one of the most developed areas in China, with a large population and a highly developed economy (Chen et al. 2012). In recent years, the level of trace metal pollution in the Haihe Basin has increased, causing serious implications for aquatic ecosystems (Xiao et al. 2011). Previous studies have reported trace metal pollution in the Haihe Basin with, of the studied metals, most anthropogenic enrichment from Cd, Zn, and Cu, and a high potential ecological risk from Cd (Tang et al. 2013). This kind of information is not available for Hg, as it has not been studied as much as other sediment-associated metals (Chakraborty et al. 2014). Indeed, this study presents for the first time the distribution and speciation of Hg in riverine sediments described for a basin as large as the Haihe Basin in China. The aims of this

study were (1) to investigate Hg concentrations and speciation in surface riverine sediments, (2) to identify Hg sources and the degree of contamination from values of the enrichment factor (EF) and geo-accumulation index (I_{geo}), and (3) to evaluate the risks from Hg using the potential ecological risk (PER) index and RAC.

Materials and methods

Study area

The Haihe Basin (35°–43° N, 112°–120° E) spans the cities of Beijing and Tianjin; the provinces of Hebei, Shanxi, Shandong, Liaoning, and Henan; and the autonomous region of Inner Mongolia, in North China (Fig. 1). Of all the water resources in China, the Haihe Basin ranks among the scarcest. Along with the rapid development of the economy, the amount of domestic sewage produced and discharged into the rivers has increased, causing a sharp deterioration in the aquatic ecosystems of the Haihe Basin. As shown in Fig. 1, the basin comprises nine major watersheds, named the Luan He (LH), Bei San He (BSH), Yong Ding He (YDH), Da Qing He (DQH), Hai-He Gan-Liu (HG), Zi Ya He (ZYH), Hei Long Gang (HLG), Zhang Wei He (ZWH), and Tu-Hai Ma-Xia He (TMH).

Sample collection and analysis

A total of 410 sediment sampling sites were selected (Fig. 1). Surface sediments were collected to a depth of 10 cm from 220 stations between May and August, 2013, from the LH, BSH, YDH, DQH, HG, ZYH, HLG, ZWH, and TMH watersheds using a Peterson grab sampler. Surface sediment samples were only collected from 220 sites because some of the riverbed surfaces were hard and were composed almost completely of sand and gravel; other rivers had been dried up for many years, and some rivers had been dredged, and sediments had been removed to remediate contaminated sediment. Three cores of undisturbed sediment were collected at each station at randomly selected points that were between 50 and 100 m apart. The sediment samples were sealed in clean polyethylene bags and placed into a cool box at the sampling site. On arrival at the laboratory, the cooled samples were frozen for being freeze-dried at -50°C and then were ground and passed through a 100-mesh sieve before analysis.

A portion of each sediment sample (0.500 g) was digested in a 3:1 mixture of hydrochloric acid and nitric acid (aqua regia) before analysis for total Hg (Bao 2000; Bloom et al. 2003). Another portion (0.100 g) was digested in a 5:1 mixture of hydrofluoric acid and perchloric acid in a microwave oven (MARS Xpress; CEM, Matthews, NC, USA) before analysis for Fe (Tessier et al. 1979); the digestion conditions are

Fig. 1 Map of the sediment sampling stations in the rivers of the Haihe Basin, China

provided in Table S1. The precision and accuracy of the Hg results were assessed by comparing with the results from the analysis of a standard sediment reference material GBW07303a (National Institute of Metrology, Beijing, China). The Hg recoveries were between 90 and 110%.

We used a modified three-stage BCR sequential extraction procedure to determine the different chemical fractions of Hg in the sediments (Arain et al. 2008), namely the proportions of Hg in the B1 (acetic acid, 0.11 mol/L), B2 (hydroxylamine hydrochloride, 0.1 mol/L), B3 (hydrogen peroxide, 8.8 mol/L, then ammonium acetate, 1.0 mol/L), and B4 (aqua regia) fractions. All of the above solutions were stored at 4 °C until analysis. We measured the concentrations of Hg, with a detection limit of 0.005 µg/L, by atomic fluorescence spectrometry (AFS) (AF-610B, Beijing Ruili Analytical Instrument Company, China), and Fe by inductively coupled plasma optical emission spectrometry (ICP-OES, Optima 2000DV; PerkinElmer, Waltham, MA, USA). We assessed the accuracy of the optimized BCR procedure by comparing with the properties of a standard sediment reference material GBW07436 (National Institute of Metrology, Beijing, China). The recoveries of the sequential extraction were calculated using the following equation (Arain et al. 2008):

$$\text{Recovery (\%)} = \left[(C_{\text{step1}} + C_{\text{step2}} + C_{\text{step3}} + C_{\text{step4}}) / C_{\text{total digestion}} \right] \times 100, \quad (1)$$

The recoveries of the chemical fractions of the heavy metals ranged from 92 to 105%, and the relative standard deviation (used to indicate the precision of the method) was less than 5%. Before use, all plastic and glassware were soaked in 10% HNO₃ (v/v) for at least 2 days, and then were immersed in, and rinsed with, ultrapure water.

Assessment approaches and statistical analyses

The EF is derived by standardizing a measured element against a reference value. The reference element in this study was iron (Fe), the element most widely used for standardization. The EF values of heavy metals were calculated with the following equation (Bhuiyan et al. 2010):

$$\text{EF} = (C_n / C_{\text{Fe}}) \times (B_n / B_{\text{Fe}})^{-1}, \quad (2)$$

where C_n and C_{Fe} are the concentrations of the examined and reference metals in the sediments, respectively, and B_n and B_{Fe} are the background concentrations of the examined and reference metals in the reference sediments, respectively. In this study, a soil background value for the Haihe Basin of Hg of 0.065 mg/kg was adopted as the baseline value (CNEMC 1990). The different classes of this index are shown in Table S2.

The I_{geo} is another standard assessment model that facilitates evaluation of heavy metal pollution in sediments by comparing current concentrations with pre-industrial levels (Müller 1969). The I_{geo} is defined by the following equation:

$$I_{geo} = \log_2 \left(\frac{C_n}{K \times B_n} \right), \quad (3)$$

where C_n is the concentration of the heavy metal, B_n is the corresponding background heavy metal concentration, and K is the background matrix correction factor ($K = 1.5$). The classes of the I_{geo} that indicate the degree of sediment pollution caused by heavy metals are shown in Table S2.

The PER index was developed by Håkanson (1980) to assess the degree of contamination of trace metals in sediments. The equation for calculating the PER index of Hg is as follows:

$$E_r^i = T_r^i \times C_f^i = T_r^i \times (C_s^i / C_n^i), \quad (4)$$

where C_s^i is the concentration of the element in the samples, C_n^i is the background concentration of the element, C_f^i is the single element pollution factor, E_r^i is the PER index of an individual element, and T_r^i is the biological toxicity factor of an individual element, which is defined as $Hg = 40$ (Yang et al. 2009). The different levels of this index are shown in Table S2.

The RAC is based on the occurrence of different forms of heavy metals in the sediments. This index assesses the availability of metals in solution from the percentage of the B1 fraction in sediments (Singh et al. 2005). The classification for this index is provided in Table S2. Details of the abovementioned methods are provided in the Supplementary Materials (Table S2). We analyzed the Hg dataset with SPSS (version 18.0 for Windows) and processed the experimental data with ArcGIS 10.1.

Results and discussion

Total Hg concentrations and Hg speciation in surface sediments

Total Hg concentrations in surface riverine sediments of the Haihe Basin ranged from 0.001 to 1.05 mg/kg, and averaged

0.054 mg/kg (Fig. 2). The Hg concentrations exceeded the background values at 39 of the 220 sampling sites. The average total Hg concentrations in the BSH, DQH, HLG, LH, TMH, YDH, ZWH, and ZYH watersheds were 0.044, 0.058, 0.108, 0.052, 0.043, 0.030, 0.028, and 0.065 mg/kg, respectively (Table S3). The Hg concentrations were highest in the middle reaches of the LH watershed, in the middle reaches of the HLG watershed, and in the middle and lower reaches of the ZYH watershed.

There was considerable variation in the proportions of the four Hg fractions in the surface sediments (Figs. 3 and S1). The B1, B2, B3, and B4 fractions accounted for, on average, 9.86, 6.54, 11.0, and 72.6% of total Hg and ranged from 0 to 77.5%, 0 to 38.6%, 0 to 65.6%, and from 8.85 to 98.4%, respectively. The B4 fraction of Hg, the most stable fraction, dominated in the surface sediments of the Haihe Basin, which indicates that the risk to the aquatic ecosystem from Hg is extremely low, and the water can be considered safe (Yang et al. 2009). Total Hg concentrations ranged from 0.010 to 0.251, 0.011 to 0.311, 0.014 to 0.173, 0.001 to 0.093, 0.012 to 0.182, 0.003 to 0.136, 0.004 to 0.073, and from 0.006 to 0.256 mg/kg in the riverine sediments from the BSH, DQH, HLG, LH, TMH, YDH, ZWH, and ZYH watersheds, respectively (Fig. S2). The total Hg concentrations and the Hg speciation in the sediments of the Haihe Basin and other selected water bodies are compared in Table S4. The average Hg concentrations (0.045 mg/kg) in surface riverine sediments of the Haihe Basin were lower than those in other places, and the B4 fraction also dominated in other places (Table S4). Total Hg concentrations exceeded the background level at 17.7% of the sampling sites, and the threshold effect concentration (TEC) at 5.00% of the sampling sites (Table S5).

Sources of Hg contamination in surface sediments

Trace metals in sediments have two main sources: natural and anthropogenic (Feng et al. 2004). We calculated the EF value for each watershed to evaluate the influence of anthropogenic activities on Hg concentrations in the sediments of the Haihe Basin (Fig. 4). DQH, with a mean EF value of 2.69, was the most contaminated watershed, followed by the HLG, LH, BSH, ZYH, ZWH, TMH, and YDH watersheds, which had mean EF values of 1.43, 0.961, 0.865, 0.800, 0.701, 0.598, and 0.483, respectively. The distributions of the EF values and the Hg concentrations were similar. The average, minimum, and maximum EF values for Hg were 1.12, 0.011, and 41.6, respectively. As shown in Fig. 4, only 1.40% of the Hg EF values in the surface sediments were 1.5 or higher, and most were less than 1.5, which shows that Hg enrichment in the Haihe Basin was mainly from natural weathering and erosion (Table S2).

The Hg EF values were different for each river system in the Haihe Basin. The mean Hg EF in the sediments of the

Fig. 2 Spatial distribution of total Hg concentrations in surface riverine sediments of the Haihe Basin (mg/kg). The map was created and edited in ArcGIS (version 10.1, ESRI, USA) using the inverse distance weighted (IDW) interpolation method

DQH watershed was substantially more than 1.5 and exceeded 1.5 at 19.0% of the sampling points. The percentage of sampling sites with an Hg enrichment factor greater than 1.5 was highest in the BSH (20%), and this percentage decreased in the other watersheds in the following order: DQH > HLG > TMH > ZYH > ZWH > LH > YDH. While anthropogenic sources had some influence on surface sediments in the eight

major river systems, natural weathering and erosion and other natural factors were more important.

The mean I_{geo} value was highest in the HLG watershed (-0.668), which indicates that this was the most contaminated watershed. The mean I_{geo} values in the DQH, ZYH, TMH, BSH, ZWH, YDH, and LH watersheds were -1.38 , -1.40 , -1.51 , -1.75 , -2.12 , -2.29 , and -2.96 , respectively (Fig. 4).

Fig. 3 The proportion (%) of the B1 (exchangeable/acid soluble), B2 (reducible), B3 (oxidizable), and B4 (residual) fractions in surface riverine sediments of the Haihe Basin derived from the modified BCR

standard method. The maps were created and edited using ArcGIS (version 10.1, ESRI, USA) with the inverse distance weighted (IDW) interpolation method

When classified with this index (Table S4), 100% of the samples from the ZWH watersheds were clean; more than 90% of the samples from the LH, YDH, BSH, and TMH watersheds were clean, and 80% of the samples from the DQH, ZYH, and HLG watersheds were clean. Overall, the I_{geo} classification shows that the surface sediments of the Haihe Basin are only slightly polluted with Hg.

Hg risk assessment in surface sediments

The average values of the PER index in the surface riverine sediments of each watershed were ranked in the following order from highest to lowest: HLG (66.5) > ZYH (39.9) > DQH (35.5) > LH (31.9) > BSH (27.2) > TMH (26.7) > YDH (18.6) > ZWH (16.9) (Fig. 5). The $Hg E_r$ values of the HLG watershed surface

Fig. 4 Values of the enrichment factor (EF) and geo-accumulation index (I_{geo}) of Hg in surface riverine sediments of the Haihe Basin. The map was created and edited using ArcGIS (version 10.1, ESRI, USA) with the inverse distance weighted (IDW) interpolation method

sediments ranged from 8.48 to 418, with an average of 66.5, and indicate moderate risk (Table S2). In this watershed, 6.25% of the samples were classified as representing a serious risk, while 31.3% of the samples represented a moderate-risk level. The

mean E_r^i values for the other watersheds were less than 40, indicating a low risk of Hg pollution and low levels of Hg contamination in sediments in these rivers. However, the mean E_r^i value indicated a high level of risk in the upper reaches of the

Fig. 5 Risk assessment of Hg in surface riverine sediments of the Haihe Basin, potential ecological risk index (E_r^i), and risk assessment code (RAC, %). The map was created and edited using ArcGIS (version 10.1, ESRI, USA) with the inverse distance weighted (IDW) interpolation method

BSH and DQH watersheds, the middle reaches of the LH and HLG watersheds, and the lower reaches of the ZYH watershed (Fig. 5), which may reflect mining and industrial development in these areas. The pollution should be controlled in these areas so as to improve the river sediment quality and reduce the associated ecological risk.

We also used the RAC method to evaluate the degree of risk associated with Hg in surface sediments from the B1 fraction. The B1 fraction in the surface sediments represented 16.6, 11.9, 11.8, 9.84, 8.62, 7.63, 6.12, and 5.15% in the DQH, LH, TMH, BSH, ZWH, YDH, ZYH, and HLG watersheds, respectively (Fig. 5). The average percentages of the B1 fraction in the DQH, LH, and TMH watersheds indicated moderate-risk levels. The B1 fraction accounted for between 0.64 and 77.5% of total Hg and accounted for an average of 16.6%, in the DQH watershed, indicating a moderate-risk level; 2.78, 11.1, and 38.9% of the samples in this watershed presented very high, high, and moderate-risk levels, respectively. The B1 of metals is generally considered as the most mobile and bioavailable fraction of most metals, and while the proportion of the acid exchangeable Hg^{2+} was generally lower than the other fractions, it is still a useful indicator of the mobility and bioavailability of Hg^{2+} in sediments (Liang et al. 2012).

The PER method was developed based on the total concentration of Hg, while the RAC was determined based on the proportion of Hg occurring in the B1 fraction (Håkanson 1980; Singh et al. 2005). In this study, the risk assessment results were somewhat different from the PER and RAC values in some sediment samples, probably because of the different forms of Hg present in the sediments. For example, in HLG surface riverine sediments, the average value of $\text{Hg } E_r^i$ was 66.5 indicating moderate risk (PER method), but the Hg occurred mainly as the B4 fraction, the most stable fraction, with only a very small proportion in the B1 fraction, indicating a relatively low ecological risk (RAC method). Therefore, the results from PER and RAC were not contradictory, but should be combined to verify the risk of sediments. In the Haihe Basin, the riverine sediments posed a high PER from Hg based on its total concentrations in the upper reaches of the BSH and DQH watersheds, the middle reaches of the LH and HLG watersheds, and the lower reaches of the ZYH watershed, while only the riverine sediments in the DQH, LH, and TMH watersheds represented a medium risk based on the proportion of Hg occurring in the B1 fraction (16.6, 11.9, and 11.8%, respectively). Our results suggest that it is important to monitor and pay attention to Hg contamination in the surface riverine sediments in the DQH and LH watersheds of the Haihe Basin.

Conclusion

We assessed the Hg contamination in surface riverine sediments of the Haihe Basin. Concentrations of Hg ranged from

0.001 to 1.05 mg/kg, and Hg was mainly in the B4 fraction, followed by, in descending order, the B3, B2, and B1 fractions. Concentrations of Hg in surface sediments were higher in the LH, HLG, and ZYH watersheds. Most sediment-associated Hg appeared to be derived from natural sources, rather than anthropogenic pollution. The average PER values for surface sediments in each watershed decreased in the following order: HLG > ZYH > ZYH > LH > BSH > TMH > YDH > ZWH. Sediments in the HLG watershed were moderately polluted, while the PER values for the other seven watersheds were low. We found high PER values at many sampling sites throughout the Haihe Basin, mainly in the BSH, DQH, LH, and ZYH watersheds. The RAC values showed that there was a moderate risk from Hg in sediments in the DQH, LH, and TMH watersheds, but low risks in the other watersheds. This information will help support the development of effective management strategies to control Hg pollution in the Haihe Basin.

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