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Pollution characteristics of mercury (Hg) in surface sediments of major basins, China

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ABSTRACT

To investigate the pollution status and potential pollution risk of mercury (Hg) in China, surface sediment samples were collected from eight hundred and eighty-one sites, including ten major basins (Songhua River Basin (SRB), Liao River Basin (LRB), Hai River Basin (HRB), Yellow River Basin (YRB), Huai River Basin (HuRB), Yangtze River Basin (YtRB), Pearl River Basin (PRB), Southeastern River Basin (SeRB), Southwestern River Basin (SwRB) and Northwestern River Basin (NwRB)). Results showed that Hg concentrations in sediments of ten basins in China ranged from 0.001 to 8.800 mg/kg, with average ± S.D. value of 0.274 ± 0.675 mg/kg, which was obviously higher than Chinese soil background value (0.038 mg/kg) and Chinese sediment background value (0.040 mg/kg). The mean Hg concentration of ten basins decreased in the order of HRB > YtRB > SRB > PRB > HuRB > SwRB > YRB > SeRB > LRB > NwRB. Moreover, it was found that the Hg concentrations in the sediments of LRB, YtRB, PRB, SeRB and SwRB were partly driven by their total organic carbon (TOC) contents, while the effect of pH on the distribution of Hg was not obvious. The Hg concentration data were also compared with those got in other periods (1994–2015) to obtain the general variation tendency of Hg level. It was recorded that Hg concentrations in HRB have remained on high levels for a long history, while Hg contamination situation in YRB after 2004 has potentially turned to be better. The results of pollution assessment by sediment quality assessment guidelines (SQGs), contamination factor (CF), geoaccumulation index (I_{geo}) and potential ecological risk (E_i) suggested that YRB and HRB were the most seriously polluted river basins among the ten basins. It is urgent of constructing SQGs in China to scientifically evaluate the Hg pollution in the future.

1. Introduction

Mercury (Hg) has become of increasing concern because of its toxicity, non-biodegradable and persistent nature, and the bioenrichment ability in food chain (Shi et al., 2005). In our daily life, we are all exposed to some forms of Hg through the air we inhale, the water we drink and the food we eat (Syversen and Kaur, 2012). Additionally, Hg has been used in a vast variety of products ranging through seed treatment, consumer applications, dental fillings and preservatives in vaccines (Syversen and Kaur, 2012). The signal of Hg pollution has been found worldwide in various archives such as sediments, peat bogs and glacier ice. Among these archives, a great number of studies have indicated that Hg in sediments plays a critical role in the cycle of Hg in aquatic ecosystem (Zhang et al., 2014a). Hg discharged from anthropogenic sources may accumulate in bottom sediments as the suspended particles on which they are adsorbed settle out (Zhang et al., 2014a). On the other hand, coagulation, flocculation and co-precipitation can also cause removal of Hg from the water column to sediment due to changes in pH and salinity of waters during estuarine mixing (Boyle et al., 1977). As estimated by Ullrich et al. (2001), the total Hg concentrations in surface sediments fluctuate from 0.02 to 0.4 mg/kg in uncontaminated or less contaminated rivers, and can be as high as 100 mg/kg in urban, industrial or mining areas. As the major sink for Hg in aquatic systems, sediments have been suggested as more significant tools for the better understanding of Hg pollution status than the analysis of the overlying water column as a result of discontinuity and fluctuation in water flows (Castillo et al., 2013).

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Hg in sediments has correspondingly received more and more worldwide attention. A search on Web of Science using "mercury; sediment" as search phrases will give almost 6000 records in return since 1900. On the whole, most investigations in China mainly focus on regional rivers, lakes and reservoirs (Yu et al., 2012; Zhang et al., 2014a). For instance, Zheng et al. (2008a) characterized Hg concentrations in the sediments of Wuli River, Cishan River and Lianshan River. Yu et al. (2012) reported the distribution, speciation and bioavailability of Hg in sediments from the Pearl River Estuary. However, these studies are far from being sufficient to systematically reveal the Hg contamination on a national scale. Moreover, very limited attention has been given to exploring the Hg pollution of rivers in the Southeastern, Southwestern and Northwestern regions in China. Additionally, researchers tend to compare the obtained data with the past studies to figure out the variation tendency of metal concentrations. This reflects the significance of historical data of metal concentrations. Based on these, 881 sediment samples were collected from ten major basins (Songhua River Basin (SRB), Liao River Basin (LRB), Hai River Basin (HRB), Yellow River Basin (YRB), Huai River Basin (HuRB), Yangtze River Basin (YtRB), Pearl River Basin (PRB), Southeastern River Basin (SeRB), Southwestern River Basin (SwRB) and Northwestern River Basin (NwRB)) from 2003 to 2004, covering 30 provinces in China. The basins under investigation also cover most of water systems across the country. We thoroughly and systematically investigate the spatial distribution of Hg contamination on a national scale, then further compare our results with those got during 1994–2015 and perform the pollution assessment by several methods (i.e., sediment quality guidelines (SQGs), contamination factor (CF), geoaccumulation index (I_{geo}) and potential risk index (E_i)).

2. Materials and method

2.1. Samples collection

The 881 sampling sites were selected during 2003–2004, covering 30 provinces in China. Samples were collected from the seven major river basins, including the SRB, LRB, HRB, YRB, HuRB, YtRB and PRB. Sediment samples were also collected from rivers in Southeast, Northwest and Southwest internal rivers drainage areas because little data were available on the occurrence of Hg in the past several decades. The global positioning system (GPS) was used to locate the sampling positions. The distribution map of sampling sites is shown in Fig. S1.

All sediment samples were collected by hydrological bureau personnel using consistent sediment sampler and procedures designed to obtain representative samples. At each sampling site, three subsamples were collected and homogeneously mixed in order to gain a repressive sediment sample using a grab sampler and stored in acid-cleaned plastic containers. All sediment sampling equipment and containers were acid cleaned before use. The containers were double bagged, stored in an icebox during transport, and frozen at −20 °C in the laboratory until analysis. Large stones and detritus were firstly removed from the samples which were then dried under the natural condition. The sediments were ground in a mortar and then passed through a 180 μm mesh so that they would have consistent physical properties.

2.2. Analysis of mercury concentration

Hg concentrations in sediments were measured using an established method (Wang et al., 2003). Total Hg concentration was determined by atomic fluorescence spectrometry (AF-610, Beijing Ruili Analytical Instrument Company, China). The quality controls for the strong acid digestion method included reagent blanks, duplicate samples, and standard reference materials. The QA/QC results showed no sign of contamination in all the analyses. The accuracy of the analytical procedures employed for the analysis of the Hg concentrations in sediments was checked using the certified reference material of China (ESS-1 and GSD-9), obtaining recovery of 114.28% and 95.40%, respectively (Table S1).

2.3. pH measurement and total organic carbon analysis

The pH values of the samples were determined in the deionized water with a ratio of 1:5 (sediment to water) by using a pH meter. Total organic carbon (TOC) content in sediments was analyzed using national standard method of China (GB7857-1987).

2.4. Assessment Hg contamination in sediment

2.4.1. Sediment quality assessment guidelines (SQGs)

Sediment quality assessment guidelines (SQGs) are useful in terms of revealing sediment contamination by comparing the sediment concentration of metals with the corresponding quality guideline (Long et al., 1995; MacDonald et al., 2000; Smith et al., 1996). According to MacDonald et al. (2000), the reliability of the probable effect level (PEL), lowest effect level (LEL), threshold effect level (TEL), minimal effect threshold (MET), effect range low (ERL), effect range median (ERM), threshold effect concentration (TEC) and probable effect concentration (PEC) for assessing sediment quality conditions is determined based on their predictive ability.

2.4.2. Contaminant factor (CF)

CF is the ratio obtained by dividing the concentration of metal (C) in the sediment by the background value (C_0) (Hakanson, 1980).

$$\mathrm{CF} = \frac{C}{C_0} \tag{1}$$

According to Hakanson (1980), CF < 1 indicates low contamination; 1 < CF < 3 is moderate contamination; 3 < CF < 6 is considerable contamination; and CF > 6 is very high contamination.

2.4.3. Geoaccumulation index (I_{geo})

The geoaccumulation index (I_{geo}) was used to assess Hg contamination in sediments, and is expressed by Müller (1969) as follows:

$$I_{\mathrm{geo}} = \log_2\left(\frac{C_n}{1.5B_n}\right) \tag{2}$$

where C_n is the measured concentration of Hg (n) in the sediment, B_n is the Hg geochemical background value of each basin (CNEMC, 1990), and 1.5 is the background matrix correction factor due to lithogenic effects. Seven classes of the I_{geo} were adopted: uncontaminated (<0), uncontaminated to moderately contaminated (0–1), moderately contaminated (1–2), moderately to strongly contaminated (2–3), strongly contaminated (3–4), strongly to extremely contaminated (4–5), and extremely contaminated (>5).

2.4.4. Potential ecological risk

The potential ecological risk was proposed by Hakanson (1980). Risk index (RI) of a given contaminant (E_i) is defined as follows:

$$E_i = T \times \frac{C}{C_0} \tag{3}$$

where T is the toxic-response factor, and is 40 for Hg (Hakanson, 1980), C represents the Hg content of each sample, and C_0 is the Hg geochemical background value (CNEMC, 1990). The RI for one metal is typically classified as follows (Hakanson, 1980): low pollution risk ($E_i \leq 40$); moderate pollution risk ($40 < E_i \leq 80$); considerable