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Modeling the source contribution of heavy metals in surficial sediment and analysis of their historical changes in the vertical sediments of a drinking water reservoir

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SUMMARY

Increasing water pollution in developing countries poses a significant threat to environmental health and human welfare. Understanding the spatial distribution and apportioning the sources of pollution are important for the efficient management of water resources. In this study, ten types of heavy metals were detected during 2010–2013 for all ambient samples and point sources samples. A pollution assessment based on the surficial sediment dataset by Enrichment Factor (EF) showed the surficial sediment was moderately contaminated. A comparison of the multivariate approach (principle components analysis/absolute principle component score, PCA/APCS) and the chemical mass balance model (CMB) shows that the identification of sources and calculation of source contribution based on the CMB were more objective and acceptable when source profiles were known and source composition was complex. The results of source apportionment for surficial heavy metals, both from PCA/APCS and CMB model, showed that the natural background (30%) was the most dominant contributor to the surficial heavy metals, followed by mining activities (29%). The contribution percentage of the natural background was negatively related to the degree of contamination.

The peak concentrations of many heavy metals (Cu, Ba, Fe, As and Hg) were found in the middle layer of sediment, which is most likely due to the result of development of industry beginning in the 1970s. However, the highest concentration of Pb appeared in the surficial sediment layer, which was most likely due to the sharp increase in the traffic volume. The historical analysis of the sources based on the CMB showed that mining and the chemical industry are stable sources for all of the sections. The comparing of change rates of source contribution versus years indicated that the composition of the materials in estuary site (HF1) is sensitive to the input from the land, whereas center site (HF4) has a buffering effect on the materials from the land through a series of complex movements. These results provide information for the development of improved pollution control strategies for the lakes and reservoirs.

1. Introduction

As defined by the World Health Organization (WHO), safe drinking water should not represent any significant risk to human health over a lifetime of consumption (WHO, 2011). However, the development of human activities has led to serious water pollution due to an increased discharge of untreated wastewater to the environment (Jonnalagadda and Mhere, 2001; Gupta, 2008). The latest findings of the State Environmental Protection Department showed that the concentration of at least one type of heavy metal in 732 sediment monitoring sites was over the environmental background, accounting for 80% of monitoring sites (Zhou et al., 2011). The outbreak of heavy metal pollution in drinking water (China Economic Weekly, 2009,) and associated influences have attracted considerable attention due to high health risks for the kidney, liver, circulatory system and nerve tissue (Salem et al., 2000; Liu et al., 2011). The pollution could be due to the number of anthropogenic activities and natural processes that produce heavy metals with uncontrolled release into environment. Heavy metals can enter into the water environment primarily through

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human activities, although these heavy metals are natural constituents of the Earth's crust (Szefer et al., 1999). Generally, the factories and plants in the drainage area of the reservoirs, especially the metal smelting industry, the manufacturing process, the electroplating industry and the chemical industry, are the primary origins of the anthropogenic heavy metals in the water environment (He et al., 2008). In addition to the point sources, certain atmospheric heavy metals may have also been deposited in the soils around the reservoir and may have entered into the water environment with surface runoff.

By comparing the degree of enrichment and accumulation of heavy metals respectively in water and sediments, it can be concluded that the heavy metal pollution in sediments was more serious because the sediments act as a sink for all of the contaminants. Similar findings have been reported by other authors (Wu et al., 2014). Heavy metals could be absorbed from the water column onto surfaces of fine particles and typically deposited in sediments. However, sediments may also act as an internal source of heavy metal pollution because they may release back into the water environment through either human activities or natural phenomena (Bryan and Langston, 1992; Savvides et al., 1996). A range of regulations have been introduced to protect drinking water sources and to control heavy metal levels in the water. However, to design effective programs and strategies for the reduction of the heavy metal concentration in the ambient water environment, information about the sources and their respective contributions is required. Source apportionment describes the techniques used to quantify the contribution of the different sources of the heavy metal concentrations in the water environment.

Even though there are many qualitative research studies on the pollution of heavy metals in sediments from Hongfeng Reservoir, there is barely any quantitative research focusing on their sources. The most contaminated tributary by heavy metals was Maiweng River (Zeng et al., 2011). The distribution of heavy metals in surface sediments showed that the concentration of heavy metals in North Lake was significantly higher than that of South Lake (Huang et al., 2008). Semi-quantitative analysis revealed that the high concentration of heavy metals in North Lake sediments was mainly influenced by human activities (Tian et al., 2011b). Liu (Liu et al., 2010) used principal component analysis to study the sources of heavy metals in Hongfeng Lake sediments, indicating that industrial sewage and background were the dominant sources of heavy metal pollution. However, the quantitative contribution rate of various sources was not investigated in either of these previous studies.

The use of a combination of multivariate approaches, such as principal components analysis (PCA) and absolute principle component score (APCS), not only qualitatively identifies the possible factors/sources but also calculates their contribution to the specific pollution. The sources based on PCA analysis were identified on the basis of observations at the receptor site alone with entirely mathematical criteria and determined by subjective analysis (Viana et al., 2008). The identification and determination of sources based on chemical mass balance model (CMB) model were through a series of diagnostic parameters. For example the source apportionment of volatile organic compounds and carbon monoxide were well evaluated by the combined use of absolute principal component score and multiple linear regression (Guo et al., 2004a,b). However, some studies reported that the CMB model has more advantages in apportion of various sources with known source profiles and ambient profiles (Gupta et al., 2007; Xue et al., 2010; Gedik and Imamoglu, 2011).

The purpose of this study is to quantitatively apportion the major sources of the heavy metals in surficial sediments and to understand their historical change. The sub-objectives are organized as follows: (1) to review and identify the potential sources of the heavy metals in sediments, (2) to establish the source profiles of the heavy metals based on the collected data (sampling data and literature data), (3) to quantitatively determine the contributions of various sources to the heavy metals in the surficial sediments, and (4) to analyze the historical change of the major sources to the heavy metals based on the developed approaches. This study can quantitatively calculate the influence of the sources on the specific pollution and offers a valuable tool for targeted and specific management of water resources based on the quantitative results.

2. Study area

As shown in Fig. 1, the Hongfeng Drinking-water Reservoir, serving as a drinking water source for the Province's capital city, is the largest and deepest artificial karst reservoir on the Yunnan-Guizhou Plateau in the southwest of China (26°23′–26°41′N, 106°33′–106°47′E). The study area is located in the temperate monsoonal zone. Its dry season and wet season starts from May to October and November to April, respectively (Wang et al., 2010a,b). The Hongfeng Reservoir surface water area is approximately 32 km^2, and the watershed covers approximately 1596 km^2. The reservoir contains five primary inflows, namely, the Maibao, Maiweng, Yangchang, Maxian, and Houliu Rivers, and one outflow, the Maotiao River. Because of its low latitude and high altitude, the area is characterized by a humid subtropical monsoon climate with warm, dry winters and cool, moist summers. With the economic development and the increase in the population, the quantity and quality demand for drinking water has intensified. However, the water quality continues to deteriorate (Zhang et al., 2007a,b; He et al., 2008). A systematic study of the pollution sources is recommended with a goal to assist environmental managers and public health officials in the control of water pollution.

3. Materials and methods

3.1. Sediment sampling and chemical analysis methods

Fourteen ambient sampling sites, located in center reservoir and its main tributaries (as shown in Fig. 1 and Table 1), were chosen to analyze the spatial variation of the heavy metals. Each inflow river has two sites respectively located in its middle area and its estuaries to understand the trends of heavy metals during transportation process. Maibao River only has one site (H5) because of its shorter length compare to other tributaries. One site (H6) located in the outlet river Maotiao River. Four center sites (HF2, HF3, HF4 and HF8) respectively located at center of the northern and southern half reservoirs and their junction area to represent the spatial distribution in reservoir body. The location of sampling sites was convenient to analysis the distribution trend of heavy metals form inflows to outflow. Four field surveys were conducted in November 2010, December 2011, April 2012 and June 2013 covered rainy season (May to October) and dry season (November to April) according to the climate characteristic of this study area. Based on our purpose, the seasonal influence was ignored by using mean concentrations of from the four field surveys for source apportionment.

A core sampler equipped with Perspex tubes (8 cm inner diameter, 40 cm long) was used to collect the sediments from the 14 sites in the Hongfeng Reservoir and its tributaries. The sediment cores were carefully cut into layers every 10 cm and were subsequently placed in polyethylene bags and stored in a cooler on ice. The samples were then immediately transported to the laboratory and preserved under nitrogen at below 4 °C. Forty available sediment samples of the surface layer (0–10 cm) were collected from