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Heavy metal pollution in reservoirs in the hilly area of southern China: Distribution, source apportionment and health risk assessment

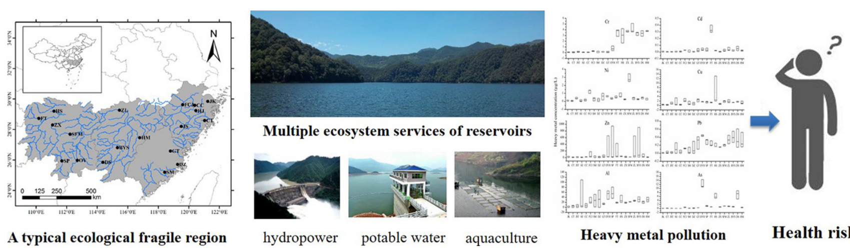
Xiaolong Wang, Lu Zhang, Zhonghua Zhao, Yongjiu Cai *

Key Laboratory of Watershed Geographic Sciences, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China

HIGHLIGHTS

- Dissolved heavy metals in 19 typical reservoirs in southern China were determined.
- Heavy metals showed great variations among these reservoirs.
- Aluminum presents great health risk to living beings in wet season.
- Arsenic is of greatest carcinogenic risk to residents.

GRAPHICAL ABSTRACT



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ABSTRACT

Reservoirs play more and more important role in providing potable water in many developing countries, including the China. In the present study, pollution characteristics, source and health risk of dissolved heavy metals in 19 representative reservoirs in the hilly area of southern China were investigated. The results presented great spatial heterogeneity in heavy metal concentrations among the studied reservoirs due to the disturbances by different anthropogenic activities. Several reservoirs had significantly higher concentrations of heavy metals compared with others, FengTan (FT) reservoir (0.34 μg/L in dry season), Cr in reservoirs of HuangShi (HS) and ZheLin (ZL) with values of 4.16 μg/L and 3.45 μg/L in dry season respectively, and Al in reservoirs of JiaoKou (JK), GuTian (GT) and DouShui (DS) with values of 1011 μg/L, 1036 μg/L and 1001 μg/L in wet season, respectively. Furthermore, there was a great difference in the seasonal variation of heavy metals, especially for Al and Pb in wet season characterized with relatively high values of 643 μg/L and 0.67 μg/L, respectively. Accordingly, Al was identified with a great health risk to living beings in view of its mean value in wet season, which greatly exceeded the criteria for drinking water of China, WHO and US EPA. Furthermore, As might be the greatest concern of health risk in this region considering its high carcinogenic risk to the local residents around the reservoirs of OuYangHai (OYH), DS, ZhiXi (ZX) and HS. Multivariate statistical analysis suggested that there was great heterogeneity in the sources of these heavy metals in the hilly area of southern China. Therefore, specific measures, such as controls on point source pollution control and tailings, should be taken for maintaining drinking water safety and aquatic ecosystem health.

1. Introduction

The surface water quality is a matter of serious concern today in developing countries, due to growing populations, rapid industrialization, urbanization and agricultural modernization. As the contaminants are primarily related to anthropogenic activities, pollution of heavy metals in surface water body has been a worldwide concern

* Corresponding author at: Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, 73 East Beijing Road, Nanjing 210008, China.
E-mail address: caiyj@niglas.ac.cn. (Y. Cai).

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because of its great threat to the health and well-being of aquatic organisms (Järup, 2003; Guo and Yang, 2016). Due to their persistence, irreversibility and biomagnifications, heavy metals pose potential long-term risks for human beings and aquatic systems. Generally, these metals are derived from different natural and anthropogenic sources, such as atmospheric deposition, erosion of bedrocks, agricultural activities, electroplating and mineral processing (Ikem et al., 2003; Li et al., 2014; Bing et al., 2016). Several studies have demonstrated the spatial and temporal variations of heavy metals and their sources, transportations, adsorption and desorption, as well as health risk assessment in aquatic ecosystems (Sedláček et al., 2017; Varol et al., 2017; Wu et al., 2017).

A river system, consisting of both the main course and the tributaries, carries a significant load of mater in dissolved and particulate phases from both natural and anthropogenic sources (Müller et al., 2008; Guo and Yang, 2016). In order to improve the local economy and social welfare, numerous dams have been built all around the world in the middle of the last century, substantially leading to more complicated river flow regime and water quality than ever before (Sedláček et al., 2017; Varol et al., 2017). As the result of damming, reservoirs play important roles in local services, such as potable water supply, irrigation, flood control and power generation. Accordingly, water quality of reservoir is crucial to health and well-being of residents around, as well as its sustainable utilization (Dummee et al., 2012; Gao et al., 2016). As a consequence of alteration on river flow, a greater amount of sediments are retained in reservoirs, which usually increases the accumulation of pollutants, including heavy metals. The transport and enrichment of heavy metals in reservoir systems have been intensively studied, including variation within surface water, distribution on hydro-fluctuation belt, sediment enrichment and organism toxicology (Ye et al., 2011; Sedláček et al., 2017). However, these studies usually focus on a certain reservoir that has great significance for local water quality conservation. Less information is available about the variation of heavy metals and their health risks in a group of reservoirs in ecologically sensitive regions.

As a typical ecologically fragile region in the world, the hilly area of southern China is a densely populated area characterized with wide undulating geomorphology of severely eroded land (Cao and Zhang, 1995; Zou et al., 2008). It is also one of the most important ecological function zones in China due to its desirable water and heat resource. In the middle of the last century, hundreds of reservoirs have been built at different scales in the hilly area of southern China for flood control, power generation and irrigation, which have greatly promoted the development of society and economy in this area (Luk et al., 1997). However, due to the wide application of chemical fertilizer and rapid urbanization, the water quality in these reservoirs is continuously deteriorating in the past decades. Substantial efforts have been performed to monitor the variations of water quality, phytoplankton biomass and structure in surface water of reservoirs in this area. Several intensive studies focusing on specific reservoirs have provided good insights into characters of pollutant input, as well as the further influence on aquatic ecosystems (Wang et al., 2004; Wei et al., 2016; Wang et al., 2017). To achieve sustainable management on the water quality at the regional level, it is still badly needed to assess variations of heavy metals within reservoir group in this area, and identify the effects on the variation of these pollutants based on comprehensive investigation.

In the present study, the characteristics of the heavy metal pollution in reservoirs in the hilly area of southern China were delineated, the potential health risks of these heavy metals were assessed, and anthropogenic influences on their seasonal and spatial variations were examined. Common water quality indicators and dissolved metals were determined in 19 typical reservoirs, which were uniformly distributed to represent the effects of different anthropogenic activities in this area. Moreover, multivariate statistical methods were applied to evaluate sources of heavy metals in studied reservoirs.

2. Materials and methods

2.1. Study area

The hilly area of southern China locates in the typical subtropical monsoon climate region with an annual mean air temperature of 18 °C and an annual frost-free period of 340 days. Concentrated during the period from May to August, the average annual rainfall is 1250 mm. Red soil of the area is formed from arenaceous shale with the thickness of 100 cm (Zhang et al., 2007). With the total area of 1.2 $\times 10^6$ km^2, this region has 3.8×10^8 inhabitants and includes hilly areas of 10 provinces in southern China. It is also the important production base for agricultural and animal husbandry, forestry and fishery in China. More than one thousands of large- and medium-sized reservoirs are densely distributed in this area, playing important roles in water supply, floodwater storage and pollution retention as well as biodiversity maintenance, especially in potable water supply (Wei et al., 2016). However, due to its strong weathering, intense leaching and human disturbance, the hilly area of southern China is characterized as a fragile and unstable ecosystem.

2.2. Sample collection and chemical analysis

A total of 19 representative reservoirs were selected in the hilly area of southern China according to spatial distribution (Fig. 1). The general descriptions and abbreviations of studied reservoirs were summarized in Table S1 in Supplementary materials. Two field surveys were conducted in wet season (August) in 2013 and dry season (April) in 2014 when the reservoirs were at high and low water levels, respectively. Three sampling sites were established as replicates in each reservoir. In each site, three replicates of surface water samples were collected at a depth of 0.5 m using previously acid-washed 5 L high density polyethylene (HDPE) containers and subsequently well mixed in situ. The mixed samples were filtered through 0.45-μm Millipore nitrocellulose filters for 8 h. About 200 mL filtrate was acidified to pH < 2 in precleaned HDPE bottles for metal analysis, and the other part was placed into a polypropylene sampling bottle for determination of other parameters. All samples were preserved in car refrigerators at 4 °C in the dark and analyzed within 48 h. The common parameters of water quality included pH, suspended solids (SS), transparency (Trans), dissolved oxygen (DO), permanganate index (COD_{Mn}), dissolved organic carbon (DOC), total nitrogen (TN) and total phosphates (TP). The pH and DO were measured at each sampling site in situ with a YSI V2–4 Multi-Parameter Water Quality Sonde (YSI Corporation, Ohio, USA). Trans was determined using the Secchi disk. SS concentrations were quantified with the filtration and gravimetric method. DOC concentrations were determined using total organic carbon analyzer (TOC VCPH analyzer, Shimadzu, Kyoto, Japan). COD_{Mn} concentrations were determined with titrimetric method by acid digestion with potassium permanganate oxidation. TN and TP concentrations were measured using a combined per-sulfate digestion (Ebina et al., 1983). The concentrations of dissolved metals (i.e. Cd, Cr, Cu, Pb, Zn, Ni, Al and As) in water samples were measured by inductively coupled plasma mass spectroscopy (ICP-MS) under standard operating conditions (the detection limits see Table S2 in Supplementary materials). Standard solution SPEX™ from the United States was used as the standard.

2.3. Risk assessment on human health

Heavy metals are identified to exert a significant challenge and risk to human health via three main pathways, including direct ingestion, inhalation through mouth and nose, and dermal absorption through exposures. Previous studies have well valued the received dose for exposure assessment through the individual pathway (US EPA, 2004; Dummee et al., 2012; Wang et al., 2018). Here, Eqs. (1) and (2) were