



Long-term variations in sediment heavy metals of a reservoir with changing trophic states: Implications for the impact of climate change



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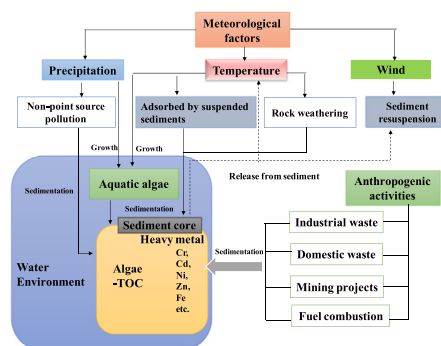
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HIGHLIGHTS

- Enrichment factors showed most of heavy metals were from anthropogenic activities.
- Cd, Zn, and P were mainly from the agricultural non-point source pollution.
- Algae-dominant TOC was negatively correlated with heavy metals in the sediments.
- Future climate change may have adverse impact on heavy metals in the aquatic environment.

GRAPHICAL ABSTRACT



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ABSTRACT

Two dated sediment cores from the Miyun Reservoir of Beijing in China were analyzed to reconstruct the pollution history of heavy metals including cadmium (Cd), chromium (Cr), iron (Fe), nickel (Ni), and zinc (Zn) as well as phosphorus (P). Enrichment factor (EF) and geoaccumulation index (I_{geo}) were applied to assess the enrichment status of heavy metals. Average EF and I_{geo} values indicated that the studied heavy metals in the sediments mainly originated from non-point source pollution and soil-water erosion, showing low ecological risks. In addition, correlation analysis and principal component analysis (PCA) identified that Cd, Zn, and P were mainly from agricultural diffusion pollution caused by utilization of the phosphate fertilizer; Zn, Ni, and Cr originated from soil erosion. PCA analysis was further conducted to investigate the relationships among meteorological factors, algae-dominant total organic carbon (TOC), and heavy metals. Results showed that algae-dominant TOC had strong positive correlation with temperature, which can be explained by that increased temperature accelerated the growth of algae. Meanwhile the opposite loadings between algae-dominant TOC and heavy metal suggested that primary production played an important role in migration and transformation of metals. Moreover, stepwise multiple regression models showed that Fe was sensitive to temperature, which accounted for approximately 39.0% and 40.1% of the variations in Fe of two sediment cores, respectively. Fe showed significant decreasing trends during the past 50 years. Reductive environment of water-sediment interface caused by increasing temperature probably contributed to the restoration of ferric iron, resulting in the release of soluble Fe to overlying waters. Future climate change with elevated temperature and extreme weather events will aggravate the ecological risk of heavy metals in water environment due to the enhanced leaching effect and non-point source pollution as well as the release of heavy metals from sediments to water environment.

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1. Introduction

The influence of climate change on water quality and aquatic environment has been widely documented recently (Arheimer et al., 2005; Schiedek et al., 2007; Hobæk et al., 2012; Ritson et al., 2014; Xia et al., 2015a, 2015b). It is often assumed that climate change can influence water environment directly and indirectly through affecting water flow, pollutant transformation and migration as well as their toxic effect (Glibert et al., 2014; Wu et al., 2014a, 2014b; Hua et al., 2015; Bussi et al., 2016; Ockenden et al., 2016; Yuan et al., 2016). Most of these studies focus on investigating nutrients and primary production such as phytoplankton and submerged macrophytes. For instance, through sediment core analysis, Xia et al. (2015a) found that global climate change contributed to the sediment nitrogen enrichment by accelerating the growth of phytoplankton; meanwhile the decreased contribution of submerged macrophytes to sediment nitrogen was observed. Bussi et al. (2016) also reported that both climate change and increase in agriculture would increase the average growth of phytoplankton in River Thames (England) in 2030.

The transformation and migration patterns of toxic pollutants such as heavy metals in the water environment are complex processes, and the influencing mechanism of climate change on heavy metals is still unknown. So far only a few studies demonstrating the impact of meteorological factors on heavy metals using the model simulation methods or short-term water quality data (Rozemeijer and Broers, 2007; Monteiro et al., 2012; Visser et al., 2012; Ye et al., 2012; Ma et al., 2013; Schroth et al., 2015; Chen et al., 2016; Lychagin et al., 2017). For instance, Rothwell et al. (2007) showed that strong rainfall could accelerate the transport of dissolved heavy metals such as lead (Pb) and vanadium (V) through the complexation process in streams of United Kingdom. Visser et al. (2012) reported that decreased precipitation and increased temperature caused by climate change would result in reduced leaching of heavy metals (cadmium (Cd) and zinc (Zn)) in summer of the Keersop catchment (Netherlands) during the period 2071–2100. Different from the results of Visser et al. (2012), our previous study (Wu and Xia, 2014) found that increasing temperature accelerated the concentrations of magnesium (Mg^{2+}) and calcium ions (Ca^{2+}) as well as ion strength in the Yellow River during the past decade. The different result could be explained by the different sources of the metals. Cd and Zn are mainly produced from human activities, and leached by surface runoff from soils to the receiving waters. Whereas, Mg and Ca are the major components of rocks; increasing temperature can enhance the weathering process, leading to the increase in major ions in the water. Thus identifying the sources of heavy metals and quantifying the impact degree of climate change would be beneficial for catchment water management.

Heavy metals in the lake/reservoir sediments are mainly from two sources i.e. natural sources such as rock weathering and anthropogenic activities such as mining and agriculture. For example, Cd was considered mainly from phosphoric fertilizers (Mar and Okazaki, 2012). Zn and Cd found in the water were from mining activities or industrial effluents as well as combustion of fossil fuels (García-Ordiales et al., 2016). In addition, some studies also indicated that fine sediments were the carriers of heavy metals in the surface land, flowing into the aquatic environment by surface runoff (García-Ordiales et al., 2016).

Meteorological factors and anthropogenic activities are considered to influence the sources and sedimentation process of heavy metals. For instance, heavy rainfall can enhance the leaching effect of heavy metals from the mineral zones and aggravate the agricultural diffusion pollution. Increased temperature can accelerate the adsorption process of heavy metals by suspended sediments in the aquatic environment, and enhance the weathering process (Weber et al., 2007). Also rising temperature is beneficial for the growth of aquatic plants, which would influence the transformation of heavy metals in the sediments through affecting binding capacity of heavy metals with TOC (Xu et al., 2013; Duan et al., 2014). Duan et al. (2014) reported that heavy metals

were strongly related with the eutrophication process in the water, mainly due to the diffusion pollution of phosphorus fertilizer containing several heavy metals. Meanwhile, by disturbing the water and sediment of the lakes/reservoirs, strong wind can release the heavy metal pollutants from surface sediments (Lin et al., 2008). In addition to meteorological factors, anthropogenic activities including industrial waste, domestic sewage, and mining projects play dominant roles in inputting of heavy metals into the aquatic environment; whereas, meteorological factors mainly affect heavy metals in the water by changing the distribution and migration processes.

Investigating the effect of climate change on temporal variations in heavy metals in the sediments of lakes/reservoirs, and identifying the relative contribution of different sources as well as their relationships with climate change and eutrophication would be of great significance for future water and soil pollution control and management under climate change. In this research, the chronological variations in contents of heavy metals including Cd, chromium (Cr), iron (Fe), nickel (Ni), and Zn were determined through analyzing two dated sediment cores from Miyun Reservoir. Parameters including enrichment factor (EF) and geoaccumulation index I_{geo} were conducted to assess the sources and the ecological risk of heavy metal in sediments. Source apportionment methods including Pearson correlation analysis and principle component analysis (PCA) were applied to identify the sources and influencing factors of heavy metals throughout the past years. Stepwise multiple regression analysis was utilized to quantify the contribution of meteorological factors to variations in heavy metals. Ecological risk of heavy metals under future climate change was predicted and discussed.

2. Materials and methods

2.1. Sediment sampling and analysis

Miyun Reservoir, as the largest and deepest reservoir in the Haihe River Watershed, was built in 1958. Two sediment cores named M1 and M2 were retrieved from two sites of the reservoir using a Glew-type gravity corer (8 cm in diameter) in Miyun Reservoir. Detailed maps of drainage network and satellite image around sampling sites are shown in Fig. 1. Sediment cores were subsampled in 1 cm-interval immediately, and transported to the laboratory for analysis within 24 h and freeze-dried soon after. The general parameters including pH, conductivity, and temperature were measured onsite using a multi-parameter analyzer (Mettler Toledo, SG23), and the general characteristics of the sampling sites are shown in Table S1.

Dry sediments were digested with HCl, HF, HNO_3 , and $HClO_4$. Then contents of the heavy metals (Cd, Cr, Fe, Ni, and Zn) and phosphorous (P) were analyzed using inductively coupled plasma atomic emission spectrometry (ICP/AES). The detailed analytical procedures were followed by the instruction of national test standard for soil in China, which has been discussed in study of Bing et al. (2016).

2.2. Ecological risk evaluation and statistical methods

EF and I_{geo} were introduced to assess the source and pollution level of heavy metals in sediment cores of Miyun Reservoir. Fe was selected as the normalizer to assess EF; the detailed calculations and evaluation criteria are shown in Supplementary materials and Table S2, respectively. To study the relationship of heavy metals with phosphorous fertilizer and primary production, Pearson correlation analysis and principle component analysis (PCA) were utilized. The primary production was represented by algae dominant total organic carbon (TOC). Then PCA analysis was conducted to investigate the relationship between heavy metals, P, TOC of multiple sources, and meteorological factors including temperature, precipitation, wind speed, and sunshine duration. In addition, stepwise multiple regression analysis was applied to quantify the contribution of meteorological factors to variations in heavy metals of Miyun Reservoir. Multiple statistical analysis was conducted by IBM

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