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Contamination, ecological risk and source apportionment of heavy metals in sediments and water of a contaminated river in Taiwan

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ABSTRACT

Houjing River, located in Kaohsiung City, is one of the most seriously contaminated rivers in Taiwan. This study analyzed the concentrations of heavy metals (As, Cd, Cr, Cu, Pb, Ni, Zn and Hg) in sediments and water samples collected from this river. Analysis of contamination factor (C_f^i) and potential ecological risk factor (E_f^i) of heavy metals in water showed that there were low grades of contamination and potential ecological risk for all heavy metals, suggesting that heavy metals in water were less likely to pose risks to the ecosystem. However, sentiment samples were found to have severe contamination levels based on ranges found in sediment quality guidelines (SQG). The average Cu concentration was almost twice as high as the upper standard values in all the guidelines. Multi-element indices were used to evaluate the synergistic effects of different metals at the sampling sites. The calculated results of different indices, the modified degree of contamination (mC_d), Nemerow pollution index (P_N) and potential ecological risk index (RI), were in good accordance. 'Heavy' contamination and 'severe' ecological risk were found at three sites, Demin, Zhuwai and Renwu. Cadmium and copper contributed the highest to the ecological risk there. Results of positive matrix factorization modeling identified four sources of heavy metal pollution in both sediments and water. Heavy metal contamination in the Houjing River is attributed to companies carrying out various industrial processes along the riverbank, including traditional metal-plating, plastic manufacture and semi-conductor packaging. Therefore, future pollution control and management plans should emphasize the strict regulation of discharge from these industrial activities.

1. Introduction

Heavy metal pollution in aquatic environments has attracted widespread attention due to its persistence, accumulation in the food chain and negative effects on the ecological and human health (Lin et al., 2016; Zhang et al., 2016c). Industrialization creates a large amounts of residual anthropogenic metals (e.g. iron oxides, manganese, organic compounds, etc.), which are then rapidly deposited and strongly attached to different types of sediment fine grains (Tessier and Campbell, 1987; Simpson and Spadaro, 2016). The metals can also be detached and released into the water column, negatively affecting water quality (Simpson and Spadaro, 2016). This occurs more often for surface sediments because there are large environmental variations that can rapidly alter different factors such as pH, temperature, and bioturbation (or resuspension) (Simpson and Batley, 2007). Anthropogenic metals, different from those that are lithogenic, are highly mobile and bioavailable, thus are more likely to adversely affect aquatic species (Tessier and Campbell, 1987). Understanding the distribution, potential ecological risk, and sources of emission are crucial for the management of these heavy metals in the environment.

Although sequential extraction procedures can be used to determine chemical speciation of heavy metals and provide detailed information about their mobility, bioavailability and toxicity (Tessier and Campbell, 1987; Simpson and Spadaro, 2016), complications involved in laboratory work performing these procedures hinder their widespread application. Total metal concentration has been commonly used to assess pollution status as well as potential ecological risk (Duodu et al., 2016; Lin et al., 2016; Villanueva and Ibarra, 2016). Many approaches have been developed and optimized for the assessment of potential ecological risk (Brady et al., 2015). The most common method of evaluating heavy metal pollution and ecological risk in sediments is by calculating contamination factor (C_f) of single elements as well as their enrichment factor (E_f) and geo-accumulation (I_{geo}) (Islam et al., 2015b; El Nemr et al., 2016). Yet, because heavy metals are more likely to have synergistic effects in the environment, these single element indices could be insufficient in their assessment of contamination and risk (Duodu et al., 2016). Multi-element indices, e.g. modified degree of contamination (mC_d) and Nemerow pollution index (P_N), can take into

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account the synergistic effects of different heavy metals (Hakanson 1980; Yan et al., 2016). Similarly, synergistic ecological risk can also be quantitatively illustrated by the potential ecological risk index (*RI*).

Southern Taiwan, the most industrialized area of the whole island, has been negatively affected by the industries there (Wang et al., 2015). Kaohsiung City has been the center of a number of different heavy industries, which have ceaselessly grown for the last 30 years. Many industrial plants producing different metals have seriously polluted water bodies around the city. The Houjing River, located in the northern part of Kaohsiung City, receives a large amount of polluted water and sediment discharge from the industrial plants operating along its banks on a daily basis. Historically, there has been continual illegal discharge of huge amounts of different pollutants from the various manufacturing companies there (Lin et al., 2009, 2010; Jiang et al., 2015). The quality of the Houjing River has attracted wide public attention leading to the need for assessment of risk and frequent monitoring to guarantee the river's support of the health of the environment there as well as the health of its residents (Lin et al., 2009; Jiang et al., 2015). Despite this great need, no study has investigated the heavy metal contamination in the Houjing River in detail, assessed its impact or identified the possible sources of pollution there.

Principal component analysis (PCA) and cluster analysis (CA) are widely preferred when determining sources of contamination in sediments and water because they are easy to do and available in most statistical software packages (Comero et al., 2011; Duodu et al., 2017). However, since they are data-sensitive, their results may come with skewed dataset distributions, requiring subsequent statistical normalization (Praipipat et al., 2013). Furthermore, it is rare that much attention is paid to the ability of these analytical methods to handle outliers and below-detection-limit (BDL) and/or missing values (Pekey and Dogan 2013). However, positive matrix factorization (PMF), an advanced multivariate statistical method using least square approach developed by Paatero and colleagues (Anttila et al., 1995), can address those uncertainties by incorporating non-negative constraints into the configuration process and point-to-point estimating each data value's error (Brown et al., 2015). Another advantage of PMF is that its associated receptor-oriented model is effective even with analyzing limited datasets (Bhuiyan et al., 2015). While the application of PMF has been specifically preferred in the field of atmospheric chemistry for years (Wang et al., 2016), it is uncommon to find it applied to understanding contamination in sediment and water samples (Bhuiyan et al., 2015).

This study assessed the contamination and potential ecological risk posed by heavy metals to the Houjing River employing PMF in source apportionment of the contamination in sediments and water. The results of this study can help provide decision-makers with an insightful view of the current contamination status of that river, essential to impending changes in land-use there, the area's economic development and environmental protection strategies.

2. Methodology

2.1. Location

The Houjing River is situated in north-west Kaohsiung City in southern Taiwan (Latitude 22°69′21.63″N − 22°73′17.74″N and Longitude 120°25′74.36″E − 120°33′74.70″E). As can be seen in Fig. 1, the river has two upstream reaches − Dashe to the north-east in Ciaotou District and Renwu to the south-east direction in Renwu District. The two reaches then meet at Si-Chingpu Landfill before flowing directly into the Taiwan Strait. The length of the Houjing River is about 13 km and its basin has long winters and summers, and short springs and autumns. The main source of water for the river is discharge from industrial areas, wastewater treatment plants, municipal discharge from activities along the riverside and rainfall. In the past, the Houjing River was a reliable resource of water for irrigation and various agricultural activities in the area from where Kaohsiung City obtains much of its food. Urbanization and industrialization have led to serious pollution of the river and altered its nature and chemistry (Lin et al., 2010).

2.2. Sampling

In order to sample waters from all potential sources of discharge along the Houjing River, we established five sampling sites, Jingjian, Renwu, Demin, Zhuwai and Dehuei. Jingjian, located at the Dashe upstream reach, receives discharge from the Dashe Industrial Park, an industrial park mostly populated with mid-stream (resin) petrochemical factories. Renwu, situated at the Renwu reach, receives discharge from the Renwu and Zhuhou Industrial Parks. These parks are home to many large petrochemical (resin production for down-stream plastic manufacture) and traditional metal-surface-coating processing centers. Demin and Zhuwai are situated where they receive discharge from two points from the Nanzih Export Processing Zone (NEPZ), where many metal-processing activities (for computer chip packaging) operates daily along with some electroplating, surface-coating and plastic production activities. Companies in the NEPZ have been reported by the media to discharge illegally large amounts of heavy metal-polluted acidic wastewater into the Houjing River. Dehuei, located close to Si-Cing Bu Landfill, is encircled by a residential area with a number of markets and condominiums, which are sources of pollution introduced by leachate and illegal discharge. Waters from originating from all these upstream sources collect at and mix together at Dehuei, making this site the most prone to heavy metal contamination in the river.

This study used standard methods of water sampling (USEPA, 2013) and sediment sampling (USEPA, 2014) to collect samples in four durations, including November of 2015, and January, April and June of 2016. A total of twenty river sediment (10–15 cm from the surface) and twenty water samples were collected by Ekman Dredge (for sediments) and bucket (for water) from the middle of the river at an average distance of 5 m from the river banks. The samples were stored in polyethylene bottles and bags and kept in dark condition below 4 °C until analysis.

2.3. Instrumental analysis

Heavy metal analysis followed the instruction of US Environmental Protection Agency (USEPA, 2000). Water samples were first acidized with HNO_3 (Zhang et al., 2016a,b). Subsequently, water samples were filtered through Whatman glass filter paper (pore size 110 mm) prior to analysis. Sediment samples (approx. 0.2 g) were lyophilized in a vacuum freeze dryer (Eyela FDU-1200, Tokyo Rikakikai, Japan, − 50 °C, 10 Pa, 24 h) and then mixed with acids (3 mL HCl and 1 mL HNO3). The mixtures were next processed through a microwave digestion system (Topex, Preekem, Shanghai, China) and subsequently filtered through Whatman glass filter paper (pore size 90 mm) before being injected into an Inductively Coupled Plasma − Optical Emission Spectrometer or ICP-OES (Optima 2100 DV, PerkinElmer, USA) for the measurement of As, Cd, Cr, Cu, Pb, Ni and Zn (USEPA, 2000). Analysis for mercury, which only requires the sample be lyophilized first, was performed in a mercury analyzer (NIC MA-2, Systematic, Taiwan).

Glassware and plastic sample containers for ICP operations were soaked in 5% HNO3 and dried before use. Likewise, prior to mercury analysis, ceramic sample containers (ship-shaped) were soaked in 5% HNO3 and baked in an oven at 500 °C for 18 h. Mercury and ICP standard solutions were obtained from J.T.Baker (Avantor Performance Materials, USA) and High-Purity Standards (Charleston, USA), respectively. All reagents were of analytical grade, and deionized water was used in all analyses. Minimum detection limits (MDLs) were established using triple standard deviation from the analysis of seven samples with the same concentration. The MDLs for sediment samples (mg/kg dry wt.) were as follows: As (0.1), Cd (0.09), Cr (0.1), Cu (0.1), Pb (0.1), Ni (0.09), Zn (0.3), Hg (0.01). Similarly, the MDLs for water samples (mg/