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Characterization and risk assessment of PAH-contaminated river sediment by using advanced multivariate methods

Yeuh-Bin Wang [a,b], Chen-Wuing Liu [a,*], Yu-Hsuan Kao [a], Cheng-Shin Jang [c]

[a] *Department of Bioenvironmental Systems Engineering, National Taiwan University, Taipei, Taiwan*
[b] *Department of Environmental Monitoring and Information Management, Environmental Protection Administration, Taipei, Taiwan*
[c] *Department of Leisure and Recreation Management, Kainan University, Taoyuan, Taiwan*

HIGHLIGHTS

- High-molecular-weight PAHs from pyrogenic sources dominate in the rainy season.
- Spatial similarity and dominating factors of PAHs in sediment identify hotspots.
- Receptor model combined with risk assessment can locate critical PAH sources.
- The toxicity from the biggest contributor to PAHs is not the most important.
- Non-point sources should be eliminated for improving sediment quality.

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ABSTRACT

This study applied advanced multivariate methods and risk assessment to evaluate the characteristics of polycyclic aromatic hydrocarbons (PAHs) in the sediment of the severely polluted Erjen River in Taiwan. High-molecular-weight PAHs (HPAHs) dominated in the rainy season. The ecological risk of PAHs in the sediment was low, whereas the total health risk through ingestion and dermal contact was considerably high. The SOM (self-organizing map) analysis clustered the datasets of PAH-contaminated sediment into five groups with similar concentration levels. Factor analysis identified major factors, namely coal combustion, traffic, petrogenic, and petrochemical industry factors, accounting for 88.67% of the variance in the original datasets. The major tributary and the downstream of the river were identified as PAH-contamination hotspots. The PMF (positive matrix factorization) was combined with toxicity assessment to estimate the possible apportionment of sources and the associated toxicity. Spills of petroleum-related products, vehicle exhaust, coal combustion, and exhaust from a petrochemical industry complex constituted respectively 12%, 6%, 74%, and 86% of PAHs in the sediment, but contributed respectively 7%, 15%, 22%, and 56% of toxicity posed by PAHs in the sediment. To improve the sediment quality, best management practices should be adopted to eliminate nonpoint sources of PAHs flushed by storm water into the major tributary and the downstream of the Erjen River. The proposed methodologies and results provide useful information on remediating river PAH-contaminated sediment and may be applicable to other basins with similar properties that are experiencing resembled river environmental issues.

1. Introduction

River sediment is the natural environment of benthic organisms and serves as a deposition area for contaminants discharged from anthropogenic and natural activities. Due to rapid industrialization and urbanization, large amounts of polycyclic aromatic hydrocarbons (PAHs) are discharged into rivers and accumulated in river sediment (Liu et al., 2009b; Li et al., 2015). PAHs can cause ecological and health risks because of their toxicity, persistence, and carcinogenicity (Ingersoll et al., 2001; Kim et al., 2013). The river sediment accumulates PAHs from two major sources including (1) "point" sources associated with definable acute or long-term activities, such as spills of petroleum-related products, and (2) "nonpoint" sources, such as the atmospheric fallout of combustion-derived particles, channelized storm water runoff, and general surface runoff from the surrounding urban or industrial community (Stout and Graan, 2010). Understanding the characteristics of spatial distribution, ecological and health risks, and source apportionment of PAHs in the river sediment is important for environmental management and protection.

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* Corresponding author.
E-mail address: cwliu@ntu.edu.tw (C.-W. Liu).

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In Taiwan, the Erjen River was the most seriously polluted river because the river was polluted by a huge amount of improperly treated domestic sewage, industrial effluent and livestock wastewater pollution in the past three decades. The river had even a notorious name "the black dragon river". But the environmental authorities launched various measures to remediate the river (Lee et al., 1996; Chen et al., 2004, 2014). According to the database of the river quality hosted by Taiwan EPA, the pollution status of the river improved gradually since then (Taiwan EPA, 2014). However, the characteristics of PAHs in the river sediment associated with the ecological and health risks have not been assessed systematically yet.

Multivariate analysis methods are efficient tools in extracting meaningful information from environmental monitoring datasets. They also can be used to explore comprehensively the characteristics of PAHs in the sediment (Song et al., 2014; Yuan et al., 2014). FA can demonstrate a relationship between monitored parameters by revealing multivariate patterns and major factors that can help to interpret original datasets. Researchers have used factor analysis (FA) to identify the dominant latent factors in the sources and processes of PAHs in the sediment (Christensen and Arora, 2007; Chiu et al., 2012). Furthermore, the factor scores can be applied to quantify the prevalence of each latent factor for every sampling site and obtain an insight into spatial characteristic of the sediment quality (Morales-Caselles et al., 2007). However, FA is based on linear relationships among the variables of monitoring datasets. On the other side, the self-organizing map (SOM) is an unsupervised artificial neural network method that can be used to extract similarity patterns in monitoring datasets for nonlinear systems. SOM can project temporal and spatial distributions of monitoring datasets on a two-dimensional plane to visualize the characteristics of contaminants in the sediment (Astel et al., 2007; Alvarvez-Guerra et al., 2008; Coz et al., 2008; Tsakovski et al., 2011). Jointly using FA and SOM can enable identifying highly contaminated areas, the control and remediation of which should be prioritized.

In addition, to take effective measures against pollution in these highly contaminated areas, the apportionment of major pollution sources and associated toxicity must be determined. Positive matrix factorization (PMF) is an advanced FA method that has been used to determine the source apportionment of PAHs in the sediment (Stout and Graan, 2010; Cao et al., 2011; Lin et al., 2013; Moyo et al., 2013; Comero et al., 2011; Comero et al., 2014). However, due to different compositions and different factions of carcinogenic PAHs for different sources, the sources might have different toxicity. Namely, the source apportionment to concentrations of PAHs might differ from the source apportionment to toxicity of PAHs. If the source apportionment to concentrations of PAHs is identified, then the resulting source apportionment to toxicity of PAHs from the corresponding major sources can be predicted by integrating PMF and toxicity assessment (Tian et al., 2013). But as far as we know, little research has involved combining the identification of PAH-contaminated hotspots with the apportionment of major pollution sources and associated toxicity for restoring PAH-contaminated sediment.

In our previous research, an assessment framework for identification of heavy-metal-contaminated hotspots in the sediment and apportionment of the major pollution sources and associated health risk was suggested (Wang et al., 2015). In this work, the similar framework was extrapolated to further study the PAH-contaminated sediment. This study assessed first the composition, ecological effects, and health risks of the PAHs in the sediment of the Erjen River. SOM and FA were then applied to evaluate the spatial characteristics of the PAH-contaminated sediment for indentifying contaminated hotspots. PMF and toxicity assessment were combined to estimate the possible apportionment of sources and toxicity posed by major pollution sources. Finally, an effective strategy was suggested for improving the sediment quality of the river.

2. Materials and methods

2.1. Study area

The Erjen River is located in southwestern Taiwan, and it originates from the Shanchuhu Lake at an altitude of 460 m. The mainstream is 61.2 km, the watershed area is about 339.2 km^2, and discharges into the Taiwan Strait. The regional climate is subtropical, with an average precipitation of about 1700 mm/year and average temperature of 24.3 °C; the dry and wet seasons are from November to April and May to October, respectively. The Sanyegong Creek is the major tributary. It flows through the Po-An Industrial Park and the metropolitan Tainan city. The location of the Erjen River is shown in Fig. 1.

In the 1970s, many polluting activities along the riverbank in the downstream, such as open burning and acid washing waste electrical circuit boards and cables recover valuable metals. These activities also discharged various toxic substances into the river. The river was ever classified as seriously polluted in Taiwan (Lee et al., 1996; Fu and Wu, 2006). To restore Erjen River, the environmental authority implemented many measures during the past three decades, including (1) prohibiting to import waste electrical circuit boards and cable; (2) banning waste open burning, (3) enforcing stricter factory effluent standard and permit management system, (4) demolishing waste aluminum melting industry factory, and (5) clearing out abandoned waste electrical circuit boards in the riverbank. Nowadays, the current pollution status of the river has been gradually improved (Chen et al., 2004; Taiwan EPA, 2014). But the characteristics of the PAHs in the sediment of the river and the associated risks have not been comprehensively assessed yet.

2.2. Data collection and treatment

Taiwan EPA set 26 sampling sites that located in the Erjen River to survey the sediment quality from 2010 to 2012. Sixteen sampling sites (E1 to E16) were located on the mainstream, and ten sites (S1 to S10) were on the major tributary. The location of these 26 sampling sites is shown in Fig. 1. Sediment samples were collected in dry and rainy seasons yearly. At each sampling location, three samples were collected by using a stainless steel grab sampler at 0–10 cm depth from the riverbed, mixed together and carefully preserved until laboratory processing. Sixteen USEPA priority PAH compounds, including Naphthalene (Np), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Fl), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flu), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)pyrene (BaP), Indeno(1,2,3-cd)pyrene (IcdP), Benzo(g,h,i)perylene (BghiP), and Dibenz(a,h)anthracene(DahA) were analyzed by the Taiwan EPA certified laboratory. Sediment samples were extracted, cleaned-up, concentrated, and quantitatively analyzed using gas chromatograph (GC) coupled with mass selective detector (MS) and a 30 m × 0.25 cm × 0.25 μm fused-silica capillary column. Method detection limits were 1–19 ng/g, and spiked recoveries of PAHs were 70–130%. All of the samples taken were analyzed in duplicate, and the relative standard deviation was less than 20%.

The undetectable values of raw monitoring data were replaced by half value of the corresponding method detection limit. Before SOM and FA analyses, the raw datasets also were standardized by the normalized method that scales the value of each variable between 0 and 1 to avoid error in analysis due to differences in the unit or range of the variables (Vesanto et al., 2000; Alvarvez-Guerra et al., 2008).

2.3. Analysis methods

2.3.1. Ecological risk analysis

Sediment Quality Guidelines (SQGs) have been empirically developed to provide a basis for evaluating the potential toxicological significance of