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Assessment of semi-volatile organic compounds in drinking water sources in Jiangsu, China

Yifeng Wu [a], Yongzhi Jia [b], Xiwu Lu [a,*]

[a] School of Energy and Environment, Southeast University, Nanjing 210096, China
[b] Water Conservancy Department of Jiangsu Province, Nanjing 210029, China

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ABSTRACT

Many xenobiotic compounds, especially organic pollutants in drinking water, can cause threats to human health and natural ecosystems. The ability to predict the level of pollutants and identify their source is crucial for the design of pollutant risk reduction plans. In this study, 25 semi-volatile organic compounds (SVOCs) were assessed at 16 monitoring sites of drinking water sources in Jiangsu, east China, to evaluate water quality conditions and source of pollutants. Four multivariate statistical techniques were used for this analysis. The correlation test indicated that 25 SVOCs parameters variables had a significant spatial variability ($P < 0.05$). The results of correlation analysis, principal component analysis (PCA) and cluster analysis (CA) suggested that at least four sources, i.e., agricultural residual pesticides, industrial sewage, water transportation vehicles and miscellaneous sources, were responsible for the presence of SVOCs in the drinking water sites examined, accounting for 89.6% of the total variance in the dataset. The analysis of site similarity showed that 16 sites could be divided into high, moderate, and low pollutant level groups at $(D_{\text{link}}/D_{\max}) \times 25 < 10$, and each group had primary typical SVOCs. These results provide useful information for developing appropriate strategies for contaminants control in drinking water sources.

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1. Introduction

The presence of xenobiotic compounds, especially organic pollutants in drinking water sources, can cause great threats to natural ecosystems and human health, owing to their chronic toxicity, persistence, and bioaccumulation (Wu and Lu, 2009; Zhou et al., 2006; Willett et al., 1998). The accumulation of these pollutants in various environmental compartments is becoming a growing environmental problem around the world. Some papers (Schriks et al., 2010; Loos et al., 2007, 2010) presented an EU-wide monitoring study on 35 organic compounds in European rivers with concentrations up to 4.0 μg/L. The majority of the compounds were semi-volatile organic compounds (SVOCs). In addition, the occurrence of emerging polar contaminants, such as benzotriazoles and metabolites of illicit drugs, in surface waters in The Netherlands was also reported (Hogenboom et al., 2009). The water quality in China has also deteriorated greatly during the past decades. Organic pollutants, including SVOCs, have a high detectable rate in many drinking water sources. There were 261 micro-organic pollutants found in 23 drinking water sources in Henan Province with the total concentrations of SVOCs ranging from 0.0078 μg/L to 0.59 μg/L (Wang et al., 2004). In the lower reach of Yangtze River, the longest river in China and the main drinking water source for many large cities, such as Nanjing, Zhenjiang, Wuxi, Suzhou and Shanghai, 62 micro-organic matters were detected with maximum total concentration of 2.879 μg/L. These organic chemicals could lead to $2.55 \times 10^{-6}\ \text{a}^{-1}$ health risk value (Jiu et al., 2009; Sun et al., 2002; Jiang et al., 2000). The worldwide increasing contamination of freshwater systems with thousands of industrial and natural chemical compounds is one of the key environmental problems facing humanity. Although most of these compounds are present at low concentrations, many of them raise considerable toxicological concerns (Schwarzenbach et al., 2006). Therefore, it is necessary to identify their source and their quantitative in drinking water. Identification and monitoring of these pollutants is of critical importance in an attempt to keep drinking water quality safe for the health of human beings.

A monitoring program for organic chemicals in water often results in complex datasets with a large number of parameters, which is often difficult to interpret and draw meaningful conclusions (Wu et al., 2009). Multivariate statistical techniques provide an alternative approach to study water quality and identify pollution source apportionments (Akbal et al., 2011). Recently, multivariate statistical techniques, such as cluster analysis (CA), principal component analysis (PCA), and correlation analysis, have been widely used in the interpretation of complex datasets to evaluate water quality and a variety of environmental issues, including inspecting spatial and temporal patterns of water

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* Corresponding author. Fax: +86 25 83795618.
E-mail addresses: xiwulu@seu.edu.cn, shinfun@163.com (X. Lu).

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quality, chemical species associated with hydrological conditions, assessment pollution sources, etc. (Bu et al., 2010; Kazi et al., 2009). One of the main advantages of these techniques is in the ability for analyzing large and complicated data, which have many variables and experimental units, and sometimes these techniques could create new variables in the comparison and interpretation of the data (Ragno et al., 2007). The CA could classify the objects into categories or clusters based on similarity within a class and dissimilarity between different classes. In hierarchical clustering, clusters are formed sequentially by starting with the most similar pair of objects and forming higher clusters step by step, the result of CA could help interpret the data and indicate patterns (Singh et al., 2005). The PCA is designed to transform the original variables into new, uncorrelated variables, and extracts the eigenvalues and eigenvectors, which describe the whole dataset rending data reduction with minimum loss of original information (Jameel et al., 2010). The result of PCA could allow for identification of major contamination sources or principal sources (Lu et al., 2010). Subsequently, correlation analysis and one-way analysis of variance (ANOVA) could further reduce the contribution of less significant variables to simplify even more of the data structure obtained from the PCA and CA. As a result, a small number of factors could usually account for approximately the same amount of information as do much larger set of original observations (Sofowote et al., 2008; Shrestha and Kazama 2007). Multivariate statistical techniques could offer valuable tools for developing appropriate strategies for effective management of water sources (Singh et al., 2008), and have been applied gradually to monitor program of water quality without requiring prior knowledge of the number and nature of sources (Mendiguchía et al., 2007; Pekey et al., 2004; kazi et al., 2009).

Jiangsu, one of the most advanced and the most open provinces in China, is located at the lower reach of Yangtze River. Jiangsu spans over 102,000 km^2 with a population of 77 million. A large number of rivers and reservoirs, such as Yangtze River, Huaihe River, Taihu Lake, Hongze Lake, Jing-Hang Canal, Subei Irrigation Canal, Yangcheng Lake, and Luoma Lake, are the dominate water sources for urban water supply in Jiangsu. This study was carried out as a survey and source apportionment of 25 SVOCs in 16 drinking water sources. The large water quality parameters dataset from this study was submitted to four multivariate statistical techniques to determine the SVOCs pollution status and to present their potential sources. The results of this study could provide some valuable information for regulatory agencies to refine pollution inventories and develop pollution control strategies to keep drinking water safe.

2. Materials and methods

2.1. Monitoring sites

The sampling network was designed to cover a wide range of determinates of key sites, which reasonably represented the dominate source water quality in Jiangsu. Fig. 1 shows the geographical distribution of the water quality monitoring sites. A total of 16 sampling sites (Site 1 to Site 16) for drinking water sources were selected to monitor organic pollutants. Site 1, Site 2, Site 3 and Site 4 were located in southern Jiangsu, and supplied raw water to Changzhou, Yixing, Suzhou, and Jiangyin regions, respectively. These regions were among the most developed areas of China. Site 5 was located in the lower reach of Tong-Yang Canal which assumed function for an important water-way and supplied raw water to Haian. Site 6 and Site 7 were selected to detect water quality for the Li Canal, which was the outlet of Gaoyou Lake and connected to Yangtze River and Huaihe River. Site 8, Site 9, and Site 10 were located at the downstream site of Hongze Lake, which belonged to Huaihe River Basin. Site 11 and Site 13 were designed for the lower reach of Huaihe River, which was also named as Subei Irrigate Canal, as a new output to Huanghai Sea of Huaihe River. Site 5 to Site 11, and Site 13 were all located in central Jiangsu. Site 12 was located at Luoma Lake as drinking water source for Suqian. Site 14 was selected to detect water quality for Sihong reach of Xuhong River, which supply raw water to Sihong. Site 15 was located at the lower reach of Xinshu River, which supplied raw water to Lianyungang. Site 16 represented Xuzhou's drinking water source and was located at Xuzhou reach of Jing-Hang Canal, downstream of Weishan Lake. Site 12, Site 14, Site15, and Site 16 were located in northern Jiangsu.

Water samples, taken within 1.0 m depth at each sampling site, were collected in 5.0 litre polyethylene bottles after rinsing several times with water from the point of collection. The samples were transferred to the laboratory in coolers containing ice to minimize the sample degradation before analysis. A solid phase extraction (SPE) method was used to extract the SVOCs from the water samples. In the laboratory, the collected aqueous sample of 1000 mL was flowed through a Supelco C_{18} SPE column at a rate of 1.0 mL/s; subsequently, the SPE column was eluted with 5.0 mL methanol. The combined extract was dehydrated by passing through anhydrous sodium sulfate and evaporated to 2–3 mL at 40 °C by a rotary evaporator. Then the samples were concentrated to 1.0 mL under a gentle nitrogen stream.

2.2. Sampling analytical methods

The water quality parameters under study were 25 SVOCs, i.e., aniline, trichloroacetaldehyde, nitrobenzene, 2,4-dichlorophenol, 2,4,6-trichlorophenol, tetrachlorobenzene, 4-nitrochlorobenzene, dimethyl-dichloro-vinyl-phosphate (DDVP), 2,4-dinitrotoluene, lindane, 2,4-dinitrochlorobenzene, dinitrobenzene, demeton/systox, 2,4,6-trinitrotoluene, hexachlorobenzene, dimethoate, parathion-methyl, pentachloropheno, malathion, parathion, heptachlor epoxide, dichloro-diphenyl-trichloroethane (DDT), polychlorinated biphenyl (PAE), benzo(a)pyrene (BAP) and aroclor. Working standards were prepared by combining the standard mixture with corresponding internal standard stock solution. Chemicals and reagents used for sample processing and analyses were chromatographically pure grade. All water samples were analyzed for SVOCs within 7 days. standard SVOCs mixture was purchased from Supelco (USA).

The SVOCs analyses were performed with a Thermo Scientific ITQ 1100 GC/MS (USA) equipped with an automatic injector and an electron capture detector. Organic compounds were separated with an Agilent DB-5 capillary column (30 m length × 0.25 mm i.d., 0.25 μm film thickness). A 1.0 μL sample was injected with a Triplus autosampler in the splitless mode. The GC/MS conditions for the sample analysis were detailed as following: the injection port temperature was maintained at 280 °C. The column temperature was programed at 60 °C (hold for 2 min) increasing at 10 °C/min to 300 °C (hold for 5 min). Helium was the carrier gas at a constant flow of 1.0 mL/min. The mass spectrometer was operated in the electron impact (EI) mode at 70 eV. The sources and interface line temperature was adjusted to 240 °C. Quantification of individual compounds was based on comparison of peak areas with those of the recovery standards. Here, the BAP and aroclor were separately analyzed using GC2010 (Shimadzu). The analytical methods were detailed in China National Standard GB3838-2002 (Ministry of Environmental Protection of China, 2002). All analysis was submitted to rigorous control procedures. Blank samples, standard samples, and duplicate samples were analyzed simultaneously in the experiment to provide quality control. The analytical precision, measured as relative standard deviation, was less than 8.0%. Accuracy of analysis was checked using standard and duplicate samples. The quality control gave good precision (S.D. $<$ 5%).

2.3. Statistical procedure

In order to evaluate SVOCs levels in different water sources and identify their distribution, the water quality dataset was further analyzed with different multivariate statistical techniques to explore their spatial trends and source apportionments. This study used one-way ANOVA, correlation analysis, PCA, and CA for SVOCs analysis with Statistical Package for Social Science (SPSS) version 19.0 for Windows 7.

3. Results and discussion

3.1. Contaminant status

The complete statistical summary of concentration parameters for SVOCs in water samples was given in Table 1. The concentrations of ΣSVOCs in different water sources ranged from 0.394 μg/L to 1.103 μg/L, with a mean value of 0.839 μg/L. In five sampling sites, which were Site 1, Site 2, Site 6, Site 10, and Site 13, the concentrations of ΣSVOCs were more than 1.0 μg/L. The lowest level of ΣSVOCs was determined at Site 16, with total concentration of 0.394 μg/L, followed by Site 7 and Site 11, with the total concentrations of 0.452 μg/L and 0.478 μg/L, respectively. The ΣSVOCs at other sites ranged from 0.5 μg/L to 1.0 μg/L.

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