



## Ecological risk assessment of polycyclic aromatic hydrocarbons (PAHs) in the water from a large Chinese lake based on multiple indicators

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### ABSTRACT

In the surface water of Lake Chaohu, China, the concentrations of 16 priority polycyclic aromatic hydrocarbons (PAHs) were measured by gas chromatograph–mass spectrometer (GC–MS). Based on the species sensitivity distribution (SSD) model and the probabilistic risk assessment (PRA) model, the indicators were calculated to assess the potential ecological risk of the individual and of multiple congeners of PAHs and their probabilities. The results revealed that the average residual level of the total PAHs (PAH<sub>16</sub>) in the water ranged from 95.2 to 370.1 ng/L, with a mean value  $181.5 \pm 70.8$  ng/L. The PAH content in the water was dominated by the low-molecular-weight congeners. The multi-substance potentially affected fractions (msPAFs) of the studied PAHs obtained by the SSD model varied from 0.29% (site B3) to 1.58% (site B6), with an average of  $0.51 \pm 0.34\%$ . The average of the msPAFs (0.93%) for the inflow rivers was greater than that for the western (0.42%) and eastern (0.34%) parts of the lake. The greatest ecological risk probability calculated by the PRA model was found for Pyr (1.55%), followed by Ant ( $7.07 \times 10^{-2}\%$ ), Fla ( $2.21 \times 10^{-2}\%$ ), Phe ( $9.25 \times 10^{-6}\%$ ), Nap ( $1.01 \times 10^{-5}\%$ ), Flo ( $1.16 \times 10^{-14}\%$ ) and Ace ( $2.86 \times 10^{-16}\%$ ). The same order of ecological risks calculated by the two models was found for the studied PAH compounds. The toxicity data might be the primary source of the ecological risk uncertainties, as indicated by the greater values of coefficients of variation (CV) for the toxicity. This study concluded that the combinations of multiple indicators based on the SSD and PRA models for the ecological risk assessment are necessary to provide more general information on the spatial variations and the probabilities of potential ecological risks of the individual and multiple congeners of PAHs.

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### 1. Introduction

An ecological risk assessment has been defined as the process of estimating the likelihood that a particular event will occur with a given set of circumstances (Maltby et al., 2005; Domene et al., 2008). During recent decades, some indicators and methods of different complexities have been proposed for the ecological risk assessment of toxic chemicals in water. In the early stages of a risk assessment, the hazard quotient (HQ), which is the quotient of the measured or estimated environmental concentration divided by the toxicant reference value, was proposed for the individual-value estimate (Solomon et al., 2000). The species sensitivity distribution (SSD) approach is one frequently used method for ecological risk assessment (Solomon et al., 1996; Steen et al., 1999). A SSD model is a statistical distribution describing, among a set of species, the variation in toxicity of a certain compound or mixture (van Straalen, 2002). To assess the eco-risk of toxic pollutants using the

SSD model, some indicators, such as the maximum permissible concentration (MPC), negligible concentration (NC), potential affected fraction (PAF), hazardous concentration at which  $p\%$  of the selected species will be affected (HC<sub>p</sub>) and margin of safety (MOS<sub>10</sub>) can be calculated for both the ecological risk of an individual chemical and the combined ecological risk of multiple substances (Solomon et al., 1996; Steen et al., 1999). The SSD method has been proven as a useful site-to-site estimate both for the eco-risk of individual chemicals and for the joint eco-risk of multiple substances (Solomon et al., 1996; Steen et al., 1999). Although significant progress and improvements have been made for the SSD methods, there are still some flaws (e.g., the lack of uncertainty analysis) (Solomon et al., 2000; Forbes and Calow, 2002). To address this issue, a probabilistic risk assessment (PRA) was proposed (Solomon and Sibley, 2002). The PRA method considers the estimate of uncertainty and the stochastic properties of exposure and effects, and it allows the variability of exposure concentrations and the distributions of species sensitivity in the risk assessment process. It can better describe the likelihood exceeding the effect thresholds and the risk of adverse effects (Solomon and Sibley, 2002; Yang et al., 2006). The indicators, including the overlap area between the exposure and effect

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curves, and the joint probability are calculated by the PRA method to assess the ecological risks (Wang et al., 2002; Shi et al., 2004). The PRA method has proven very useful in estimating both the exposure of a population or community to potentially hazardous pollutants and their responses to the chemicals in the research area (Wang et al., 2002). However, the PRA method requires as many measured data as possible to construct the probabilistic distribution of the exposure levels. The combinations of multiple risk indicators based on the SSD and PRA models for the ecological risk assessment are necessary to provide the more general information on the spatial variations and on the probabilities of the potential ecological risks of the individual and multiple pollutants.

Polycyclic aromatic hydrocarbons (PAHs) are a group of ubiquitous persistent organic pollutants that are generally formed by the incomplete combustion of fossil fuels and biomass fuels (Rogge et al., 1993; Tao et al., 2005; Xu et al., 2006). PAHs are a major concern because of their potentially toxic, mutagenic, and carcinogenic properties (Khalili et al., 1995; Fernandes et al., 1997; Larsen and Baker, 2003; Li et al., 2009). The U.S. Environmental Protection Agency (USEPA) has established 16 PAHs as the priority control pollutants, and 7 of them are potentially carcinogenic to humans, according to the International Agency for Research on Cancer. Furthermore, PAHs enter a water body through wastewater discharge, surface runoff, atmospheric deposition and other means, such as crude oil leaks (Heemken et al., 2000). PAHs can adversely affect not only human health (through drinking water and skin contact) but also aquatic ecosystems. The ecological health risks of PAHs are being increasingly studied by environmental researchers. In China, PAHs emissions in excess of 27,000 tons/year have resulted in the contamination of various environmental media (Zhang et al., 2007). Lake Chaohu, the fifth-largest freshwater lake in China, is located near the Yangtze River delta region (Fig. 1), one of the most developed regions in China. With the rapid urbanization of the surrounding area, Lake Chaohu is becoming increasingly polluted by PAHs from human activities, such as the burning of fossil fuels and agricultural and industrial practices. This pollution will damage the lake ecosystem and compromise the safe use of the lake water as a water source for drinking, industrial production and agricultural irrigation. However, there is little information on the residual levels and ecological risks of PAHs in the water from Lake Chaohu.

There are three primary objectives of this study: (1) to investigate the residual levels and distributions of 16 priority PAHs in the water; (2) to estimate the potential ecological risk of the individual and multiple congeners of PAHs, based on both the SSD and PRA methods; and (3) to discuss the uncertainty of the ecological risks of the studied PAH components. A platform, named the Bayesian Matbugs Calculator (BMC), was developed to perform the best fittings of the distribution model, the ecological risk index calculations and the uncertainty analysis.

## 2. Methodology

### 2.1. Measurement of PAHs contents in the water

Water samples from 15 sites (Fig. 1) were collected in August 2009. An emphasis was placed on the eastern drinking-water source area with six sites (A6, B2, B3, B4, B5 and B7) and the inflow rivers with four sites (C1, C2, C4 and B6). Twenty liters of water was collected from each sampling site. After shaking and mixing, a 1-L aliquot of each collected water sample was filtered through a 0.45- $\mu\text{m}$  glass fiber filter (burned at 450 °C for 4 h) using a filtration device consisting of a peristaltic pump (80EL005, Millipore Co., USA) and a filter plate with a diameter of 142 mm. Surrogate standards of 2-fluoro-1,1'-biphenyl and p-terphenyl-d14 (J&K

Chemical, USA, 2.0 mg/mL) were added to the water samples to indicate the recovery before extraction.

The water samples were extracted using a solid phase extraction (SPE) system (Supelco). C18 cartridges (500 mg, 6 ml, Supelco) were prewashed with dichloromethane (DCM) and conditioned with methanol and de-ionized water. A 1-L water sample passed through the SPE system and was extracted. The cartridges were eluted with 10 ml of dichloromethane. The volume of the extracts was reduced by a vacuum rotary evaporator (R-201, Shanghai Shen Sheng Technology Co., Ltd., Shanghai, China) in a water bath and was adjusted to a volume of 1 ml with hexane. Internal standards (Nap-d8, Ace-d10, Ant-d10, Chr-d12 and Perylene-d12) were added for the GC analysis.

All samples were analyzed on a gas chromatograph with a mass spectrometer detector (Agilent 6890GC/5973MSD). A 30 m  $\times$  0.25 mm i.d. with a 0.25- $\mu\text{m}$  film thickness HP-5MS capillary column (Agilent Technology) was used. The column temperature was programmed to increase from 60 °C to 280 °C at 5 °C/min and then was held isothermal for 20 min. The MSD was operated in the electron impact mode at 70 eV, and the ion source temperature was 230 °C. The mass spectra were recorded using the selected ion monitoring mode. The concentrations 16 PAHs were determined: naphthalene (Nap), acenaphthene (Ace), acenaphthylene (Acy), fluorine (Flo), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h) anthracene (DahA), indeno(1,2,3-cd)pyrene (IcdP) and benzo(g,h,i) perylene (BghiP).

The quantification was performed by the internal standard method using Nap-d8, Ace-d10, Ant-d10, Chr-d12 and Perylene-d12 (J&K Chemical, Beijing, China). All of the solvents used were HPLC-grade pure (J&K Chemical, Beijing, China). All of the glassware was cleaned using an ultrasonic cleaner (KQ-500B, Kunshan, China) and heated to 400 °C for 6 h. In the sampling process, three parallel samples were been collected from each sample site. The laboratory blanks were analyzed with the true samples. The average recovery for Nap, Ace, Acy, Flo, Phe, Ant, Fla and Pyr ranged from 75% to 117%, and for BaA, Chr, BbF, BkF, BaP, DahA, IcdP and BghiP was 68%, 67%, 54%, 51%, 77%, 45%, 34% and 35%, respectively. The detection limits were in the range of 0.54–4.22 ng/L.

### 2.2. Ecological risk assessment

The multiple risk indicators based on both the SSD and PRA models were calculated to obtain a comprehensive picture of the potential ecological risks of the PAHs in the water from Lake Chaohu. The SSD method (Wheeler et al., 2002; Liu et al., 2009; Wang et al., 2009a,b) was used for the site-specific assessment of the ecological risk for both the individual and multiple congeners of PAHs, while the PRA method was used for the probabilistic assessment of the ecological risk of individual PAH congeners based on all of the sampling sites.

#### 2.2.1. General procedures of the SSD and PRA methods

The basic assumption of the SSD method is that the sensitivity of a group of organisms can be described by a distribution and that the available toxicological data are considered to be a sample of this distribution. Thus, the SSD is estimated from the sample of toxicity data and visualized as a cumulative distribution function. Both the acute (LC50, EC50) and chronic data (NOEC50) can be used to build the SSD; the acute data (LC50, EC50) was used in this study. To assess ecological risk using the SSD method, there are usually four steps: (1) obtain the toxicity data of the pollutants; (2) fit the SSD curves; (3) calculate the PAFs of the individual pollutants for the ecological risk assessment of an individual pollutant; and (4) calculate the accumulated multi-substance potentially

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