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Characterizing ecological risk for polycyclic aromatic hydrocarbons in water from Lake Taihu, China

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Abstract Lake Taihu provides vital ecological services for humans in China; it receives a great deal of attention regarding its ecological and environmental conditions. In this study, the ecological risks of eight individual polycyclic aromatic hydrocarbons (PAHs) in water were assessed using probabilistic distributions of the hazard quotient based on Monte Carlo simulation. The results show that the 95th percentile of the hazard quotients ranged from 0.00074 to 2.831, and the ecological risk of Flua was highest, followed by, in descending order of risk, B[a]P>Pyr>Ant>Phe>Flu>Ace>Chr. The probabilities of hazard quotients exceeding a decision criteria of 0.3 were 18.09%, 6.51%, 3.76%, and 2.85% for Flua, B[a]P, Pyr, and Ant, respectively, indicating their potential ecological risks to aquatic organisms. The spatial distribution of hazard quotients for these four individual PAHs with potential ecological risk were obtained using Geographic Information System (GIS), and similar spatial distribution patterns were also observed in the lake. The highest ecological risks of these four individual PAHs to aquatic organisms were found in Meiliang Bay, followed by Gonghu Bay and Xukou Bay. The uncertainty within the ecological risk assessment was also discussed.

Keywords Polycyclic aromatic hydrocarbons (PAHs) · Probabilistic risk assessment · Lake Taihu · China

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of persistent organic pollutants and semi-volatile organic compounds that are widely distributed in the environment, especially in lakes, rivers, oceans, and estuaries (De Luca et al. 2004; Zhang et al. 2007; Zhou and Maskaoui 2003; Guo et al. 2011a, b). PAHs are produced by the incomplete combustion of fossil fuel, coal gasification and liquefaction, petroleum cracking, and the production of coke, carbon black, and asphalt; PAHs are also found in vehicle exhaust, waste incineration emissions, industrial wastewater, and municipal sewage (Countway et al. 2003). Moreover, spillage or leakage of crude oil or refined oil products is another important source of PAHs (Heemken et al. 2000; Srogi 2007).

Sixteen PAHs have been listed as priority pollutants by the US Environment Protection Agency (USEPA 1993). PAHs are of environmental concern due to their notorious toxic, mutagenic, carcinogenic, and teratogenic potentials. Four- to seven-ring PAHs are highly mutagenic and carcinogenic, while two- to three-ring PAHs are less mutagenic but can be highly toxic (Fernandes et al. 1997). It is well known that PAHs in water may have a wide range of adverse effects on aquatic organisms including reproductive toxicity, photo-induced toxicity, immune deficiency, mutagenicity, and carcinogenicity, whether through direct exposure or bioaccumulation processes. PAHs in water may alter population dynamics and disrupt trophic interaction and the structure of natural communities within the ecosystem (Patrolecco et al. 2010; Peterson et al. 2003). It has been reported that PAHs can decrease the reproduction capacity of oligochaetes (*Limnodrilus hoffmeisteri*) (Lotufo and Fleeger 1996) and impair the burrowing activity of amphipods (*Diporeia* spp.) (Landrum et al. 1994) and oligochaetes (*Lumbriculus variegatus*) (Kukkonen and Landrum 1994). Therefore, it is highly desirable to understand the full-scale environmental risks of PAHs to the ecosystem, especially for aquatic environments. Additionally, evaluating the ecological risk from PAHs is critical to protecting aquatic ecosystem security.

Ecological risk assessment is a process used for organizing and analyzing data, information, assumptions, and uncertainties in order to evaluate the likelihood of adverse ecological effects as a consequence of exposure to one or more stressors (Posthuma et al. 2002). In recent years, many environmental risk assessors prefer the probabilistic risk assessment to the traditional deterministic methods such as hazard quotient (Schuler et al. 2008; Zhong et al. 2010). A probabilistic distribution of hazard quotient is generated by multiple iterations using a Monte Carlo simulation based on the probabilistic distributions of exposure concentrations and toxicity data, which takes the uncertainty and stochastic property of the data into consideration. This approach can be designed for screening and ranking the risk to aquatic organisms for pollutants, as well as estimating the exposure of a population or community to potentially hazardous pollutants and their responses to the pollutants. For example, the probabilistic distribution of the hazard quotient generated by a Monte Carlo simulation was applied to assess the ecological risk of PAHs in rivers from Tianjin (Yang et al. 2006), and to analyze the risk of mercury to aquatic organisms such as the American alligator (*Alligator mississippiensis*), great egret (*Egretta alba*), and the raccoon (*Procyon lotor varius*) in the Florida Everglades (Duvall and Barron 2000). This method has also been widely employed in health risk assessment. Price et al. (2001) calculated the exposure concentrations of pesticides using a Monte Carlo simulation and evaluated the risk of pesticide exposure to humans based on the probabilistic distribution of the hazard quotient. Ma and Zhang (2000) also used the probabilistic distribution of the hazard quotient to analyze the risk of selenium to the residents in Jilin Province, China.

Lake Taihu, the third largest freshwater lake in China, is located in southern Jiangsu Province and has an area of 2.34×10^3 km^2. With a volume of 5.15×10^9 m^3, it provides ample freshwater resources for agricultural irrigation, industry, recreation, and transportation. However, the structure and function of this aquatic ecosystem may be threatened continuously as a consequence of anthropogenic activities. This has especially been true in the last few decades, during which time, urbanization, industry, agriculture, and tourism in Taihu Basin have developed rapidly, leading to the discharge of enormous quantities of pollutants into the lake. These pollutants may have had a drastic impact on the lake ecosystem. In recent years, many studies have focused on the concentrations of PAHs in the sediment of Lake Taihu, and some authors have applied sediment quality guidelines to assess the ecological risk of these pollutants (Qiao et al. 2006). However, there is little information available regarding the concentrations of PAHs in water and their potential ecological risks to the Lake Taihu ecosystem. In addition, it is important for environment managers and decision-makers to understand the ecological risks posed by PAHs to the ecosystem of Lake Taihu. Probabilistic risk assessment is an important tool for environment managers to identify and prioritize PAHs for aquatic environment protection.

Therefore, this study was conducted (1) to assess the ecological risk of eight individual PAHs in Meiliang Bay, Xukou Bay, and Gonghu Bay from Lake Taihu and rank the risk of these compounds to

aquatic organisms using a probabilistic distribution of hazard quotient, and (2) to assess the spatial variability of the risk of PAHs with potential risk to the aquatic ecosystem and identify their possible sources.

Methodology

Data sources

Exposure concentrations for PAHs were based on monitored values. A total of 33 surface water samples were collected from three bays in Lake Taihu. They included ten samples from Meiliang Bay, 11 from Gonghu Bay, and 12 from Xukou Bay. The detailed procedures for sample collection, preparation, and determination were presented elsewhere (Guo et al. 2011a). Briefly, the water samples were filtered through 0.45-μm glass microfiber membrane to remove sand and debris. The filtered water samples were extracted using a solid phase extraction (SPE) system from Supelco (Sigma-Aldrich Co, Bellefonte, PA, USA). The SPE cartridges were first activated with 10 ml of dichloromethane and then washed with 10 ml of methanol and 10 ml of ultra-pure water. Water samples (1.0 l) were passed through cartridges at a flow rate of 5 ml/min under vacuum pump. After extraction, the cartridges were eluted with 10 ml dichloromethane and then combined with dichloromethane rinse from the extraction glassware. The elution of water sample was added to a column filled with 2.0 g silica gel and 1.0 g anhydrous sodium sulfate for cleanup and fractionation. Elution was performed with 8 ml hexane first and then 10 ml hexane/dichloromethane (1:1, *v*/*v*). The second fraction containing the PAHs was reduced to 1–2 ml, subject to a solvent exchange to methanol, and concentrated to 1.0 ml by the rotary evaporator prior to chemical analysis.

PAHs were analyzed by a Water Column 1525 high-performance liquid chromatography (HPLC) system (Water column, USA) with Water column 1525 binary HPLC pump, a 474 scanning fluorescence detector, and a 2347 double beam UV detector. Separation was carried out using a ChromSep guard column (10×4.6 mm, particle size 5 μm, variation, USA) followed by a ChromSep C18 column (250×4.6 mm, particle size 5 μm, varian, USA). The HPLC separation was performed at a flow rate of 1.0 ml/min using a gradient elution program in which methanol–water was used as mobile phase. The analytical procedures were conducted under the strict quality control guidelines. None of the target compounds was detected in the procedural blanks. The recovery was in the range of 60–94%, with relative standard deviation (RSD) ranging from 5.6% to 12.8%. Under analytical condition, detection limits for PAHs ranged from 0.97 to 7.02 ng/L. In this study, eight PAHs, acenaphthene (Ace), phenanthrene (Phe), fluorene (Flu), anthracene (Ant), fluranthene (Flua), pyrene (Pyr), chrysene (Chr), and benzo[a]pyrene (B[a]P), were selected to assess their ecological risk to the aquatic organisms based on the available toxicity data.

To provide appropriate protection for aquatic ecosystems in Lake Taihu, the predominant aquatic species that are widespread in the lake (hydrophytes, algae, fish, crustaceans, mollusks, and noncrustacean invertebrates) were selected to develop the species sensitivity distributions (SSDs). For persistent organic pollutants, probabilistic risk assessment was performed based on chronic toxicity data. No observed effected concentrations (NOECs) were collected as chronic toxicity data from the ECOTOX database (http://www.epa.gov/ecotox/). If NOECs were not available, the median lethal concentrations or median effective concentrations (LC_{50}/EC_{50}) divided by the acute-to-chronic ratio (ACR=100) (Läng et al. 1998; Swartjes 1999) were used as a substitute for the chromic toxicity data. The toxicity data were only selected when their endpoints could be clearly related to the changes in the population structure such as growth, reproduction, and survival. For LC_{50} or EC_{50}, only 24–96 h durations were included in the SSDs for fish, crustaceans, mollusks, and noncrustacean invertebrates, whereas durations up to 168 h were included in SSDs for hydrophytes and algae. At least four species should be included in the SSDs to represent species distribution of the ecosystem (Aldenberg 1993; Carriger and Rand 2008). When multiple toxicity concentrations were available for a given species, the geometric mean was taken (Solomon et al. 2000). If the life-stage information was available for a species, the most sensitive life stage was selected as an endpoint for effects analysis.

Risk analysis

The exposure concentrations and toxicity data distributions were integrated using a Monte Carlo simulation to randomly sample values from distributions of exposure and toxicity data and to generate a probabilistic distribution of quotients. As Vose (1996) reported, point estimates in a model equation were replaced by probabilistic distributions; the samples were randomly selected from each distribution of input parameters, and the results of output parameters in the form of a probabilistic distribution were combined. In the present study, the exposure concentrations and toxicity data were derived stochastically from the corresponding lognormal distributions to generate the distributions of hazard quotients for the 10,000 iterations, which incorporated both uncertainty and variability. This method would be useful to identify and prioritize chemicals, especially when the hazard quotients were close to the decision criterion.

The hazard quotient distribution (HQD) was calculated as a ratio of exposure concentration distributions (ECDs) to species sensitivity distributions (SSDs) according to Eq. 1:

$$\mathrm{HQD} = \mathrm{ECDs}/\mathrm{SSDs} \quad (1)$$

The hazard quotients of PAHs with potential ecological risk were calculated by exposure concentrations for each spatial point divided by the toxicological benchmark concentration. Then, toxicological benchmarks applied in this study were 5% percentile values from probability distribution of toxicity data (Hall and Anderson 1999; Solomon et al. 1996). Finally, geostatistic analysis of the estimated risk quotients was conducted to obtain a visual representation of risk using spatial distribution maps.

Statistical analysis

All descriptive statistics were conducted to acquire the features of the datasets using the SPSS 11.5 software. The Shapiro–Wilk tests for normality were conducted using the Origin 8.0 programs, and a critical p value of 0.05 was taken to indicate significance. Monte Carlo simulation was conducted using Crystal Ball (version 11.1). The maps of the spatial distributions of hazard quotient were produced using the inverse distance to a power method with the Arc GIS 9.3 program.

Results and discussion

Statistical distributions of PAHs concentrations and toxicity data

Statistical distributions of PAH concentrations and toxicity data were tested prior to probabilistic risk assessment. The distributions of the raw and natural logarithm-transformed concentrations of Flua were illustrated in Fig. 1. The raw data were leptokurtic with a right skewed distribution and a portion of high exposure values that accounted for this phenomenon (Fig. 1a). Subsequently, the raw data was natural logarithm-transformed, and the log-normal distribution described the exposure concentrations of PAHs well (Fig. 1b). Similarly, the distribution of chronic toxicity data was right skewed (Fig. 1c), and chronic toxicity data followed a log-normal distribution curve as a consequence of natural logarithm transformation (Fig. 1d).

The Shapiro–Wilk test was applied to further test the normal distribution of both raw and ln-transformed data from the exposure concentrations and acute toxicity data (Table 1). The results indicated exposure concentrations and toxicity data were individually fitted to a log-normal distribution. The log-normal distribution types for the concentrations of PAHs in aquatic environments and chronic toxicity data have been reported in previous studies (Liu et al. 2009; Wang et al. 2002; Yang et al. 2006).

Ecological risk assessment

Probabilistic distributions of hazard quotients

The probabilistic distributions of the hazard quotients for the eight individual PAHs generated by Monte Carlo simulation were presented in Fig. 2. The calculated hazard quotients were natural logarithm-transformed due to their distribution being right skewed. As shown in Fig. 2, the distributions of hazard quotients represented both hazardous level of PAHs and the likelihood of the risk occurring over a large range. The 95th percentile values of the hazard quotient distribution calculated from Monte Carlo simulations were 0.0057, 0.131, 0.00778, 0.187, 2.831, 0.312, 0.00074, and 0.698 for Ace, Phe, Flu, Ant, Flua, Pyr, Chr, and B[a]P, respectively. Consequently, it was possible to rank the risk of these eight

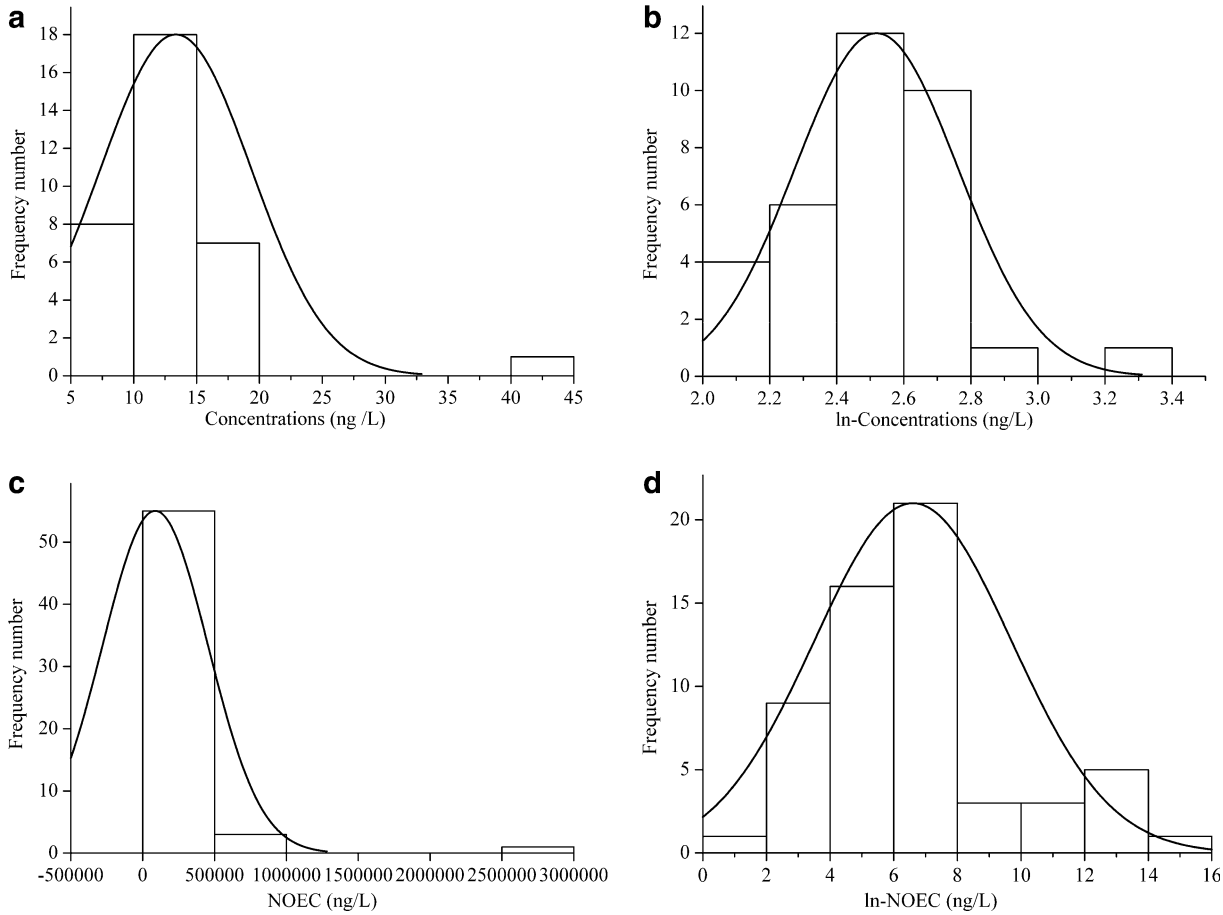

Fig. 1 Distributions of concentrations (**a**) and ln-transformed concentrations (**b**) for Flua in water and distributions of NOECs (**c**) and ln-transformed NOECs (**d**) for Flua to aquatic organisms

individual PAHs to the aquatic ecosystem as Flua>B[a]P>Pyr>Ant>Phe>Flu>Ace>Chr. According to other studies, Ant was the most hazardous chemical in the surface water from the Tianjin rivers, despite its moderate exposure level among the eight PAHs (Yang et al. 2006), while Pyr was the most toxic chemical among the 13 PAHs evaluated in the interstitial waters of San Diego Bay, Eagle Harbor, and Curtis Creek

Table 1 Distribution parameters for ln-transformed exposure and toxicity data of eight individual PAHs in water from Lake Taihu (nanograms per liter)

PAHs	*n*	Ln-transformed exposure concentrations			Ln-transformed chronic toxicity data		
		M	SD	$p_{s\text{-}w}$[a]	*M*	SD	$p_{s\text{-}w}$
Ace	33	0.634	0.764	0.000	8.43	1.79	0.679
Phe	33	3.384	0.306	0.006	8.10	1.61	0.226
Flu	33	2.443	0.316	0.036	11.37	2.45	0.292
Ant	33	0.501	0.469	0.018	5.48	1.93	0.821
Flua	33	2.491	0.367	0.014	6.60	2.09	0.478
Pyr	33	1.964	0.431	0.002	5.95	1.48	0.685
Chr	33	1.422	0.527	0.003	9.75	0.41	0.987
BaP	33	0.207	0.955	0.000	7.41	3.79	0.065

M arithmetic mean, *SD* standard deviation

[a] $p_{s\text{-}w}$: significant level with Shapiro–Wilk test for fit of normal distribution for ln-transformed toxicity data, a *p* value >0.05 suggested a normal distribution

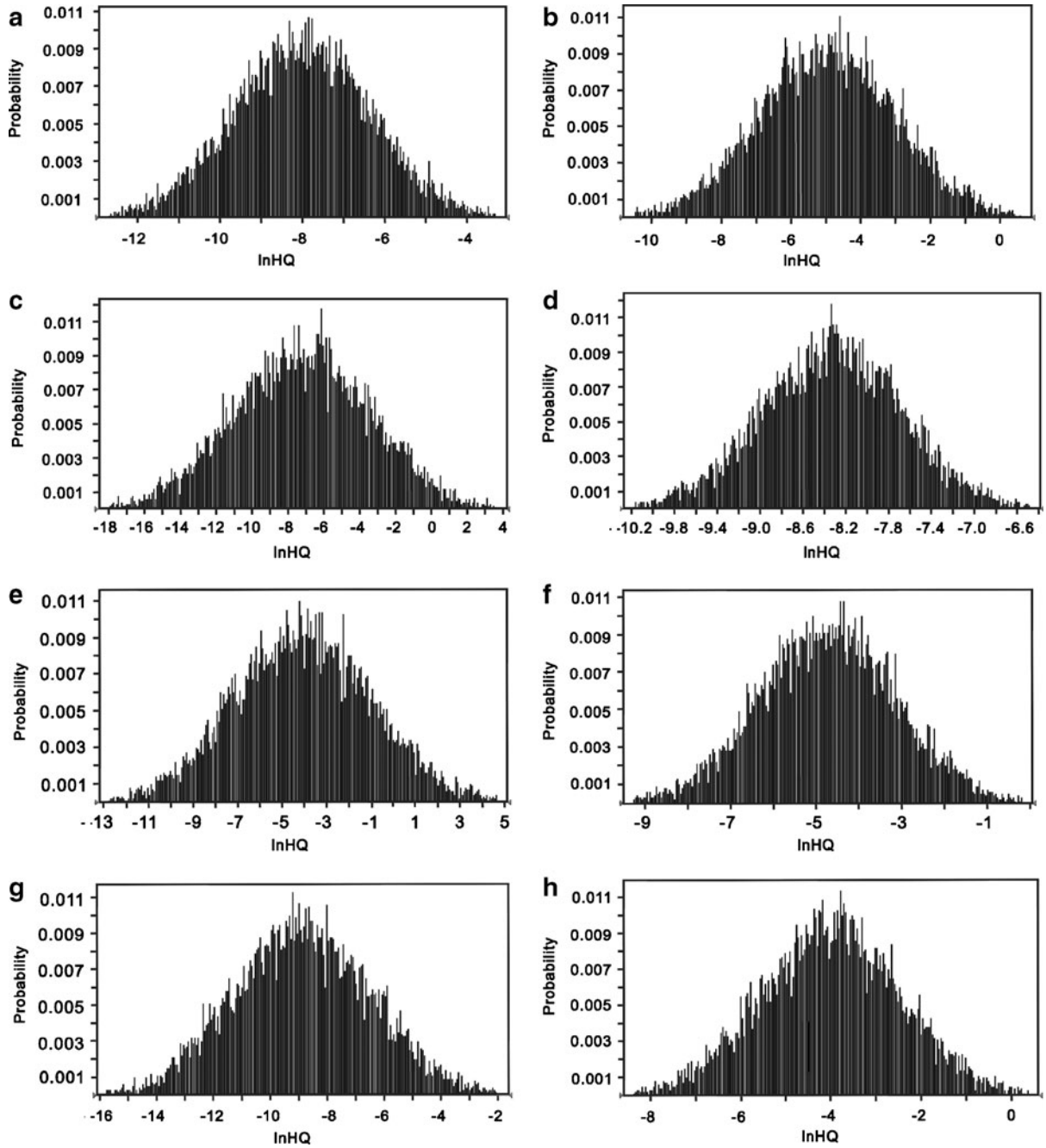

Fig. 2 Probabilistic distributions of hazard quotients of eight individual PAHs in the surface water of Lake Taihu. The calculated hazard quotients were natural-logarithm transformed: (**a**) Ace, (**b**) Ant, (**c**) B[a]P, (**d**) Chr, (**e**) Flua, (**f**) Phe, (**g**) Flu, and (**h**) Pyr

(Swartz et al. 1995). This may be attributed to subtle differences in exposure levels and aquatic ecosystem structure and functions in different study areas, as well as regional water quality parameters such as pH, dissolved organic matter (DOM), and hardness. For example, pH and ionic strength have an important impact on the distribution of PAHs between the aquatic phase and DOM; therefore, these factors affect the concentrations, transportation, and fate of PAHs in the aquatic environment (Wu 2009). According

to Landrum et al. (1992), DOM existing in the water may even decrease the bioavailability of organic pollutants to aquatic organisms.

The results of ecological risk assessment showed that although the exposure concentrations of Phe in water were considerably higher than those of other individual PAHs, its risk to the aquatic ecosystem was moderate. In addition, similar exposure concentrations for Flu and Flua were observed in Lake Taihu, but the risk to aquatic ecosystem for Flu was much lower due to its lower toxicity effect. It has been reported that the NOECs of Flua were in the range of 0.012–2,670 μg/L, with an average value of 94.61 μg/L, whereas the NOECs ranged from 2.12 to 14,000 μg/L for Flu, with a mean of 1,170.91 μg/L (http://www.epa.gov/ecotox/). Similarly, although the exposure concentrations of B[a]P in the rivers from Tiaijin were comparable to those of Flua, Flua imposed a relatively higher risk than B[a]P (Wang et al. 2002). These finding indicated that the ecological risk of PAHs was associated with both their exposure concentrations in water and toxicity effect on the aquatic organisms.

According to Parkhurst et al. (1996), chemicals with hazard quotients exceeding 0.3 may pose a risk to aquatic ecosystems due to interaction and accumulation of chemicals in the aquatic environment. The probabilities of the hazard quotients being less than 0.3 were high (100%) for Ace, Flu, Phe, and Chr, suggesting negligible ecological risks to aquatic organisms. In contrast, there were 18.09%, 6.51%, 3.76%, and 2.85% instances in which the probabilities of the hazard quotient exceeded the criteria of 0.3 for Flua, B[a]P, Pyr, and Ant, respectively, indicating a possibility of adverse effects on the ecosystem in Lake Taihu. As an important source of freshwater for surrounding areas, the risk of Flua, B[a]P, Pyr, and Ant should receive immediate attention. In an earlier study on the ecological risk from PAHs in the sediment from Meiliang Bay (Qiao et al. 2007), the probabilities of hazard quotients greater than 0.3 were 40%, 90%, and 90% for B[a]P, Pyr, and Flua in sediment, respectively, which were considerably higher than those observed in the present study. This may mainly be attributed to the fact that the exposure concentrations of B[a]P, Pyr, and Flua in the sediment in the earlier study were relatively higher than those in water phase in this study.

In order to calculate hazard quotients of these four PAHs with potential ecological risk, the toxicological benchmarks were derived from SSDs using Monte Carlo simulation, with the values of 9.21, 1.76, 90.02, and 21.24 ng/L for Flua, B[a]P, Pyr, and Ant, respectively. Subsequently, spatial distribution maps of the risk quotients were showed in Fig. 3 for Flua, Pyr, B[a]P, and Ant with potential ecological risks. As shown in Fig. 3, similar distribution patterns of hazard quotient for B[a]P, Pyr, Ant, and Flua were observed in the maps. Based on the spatial distribution patterns, the highest ecological risk of these four PAHs was found in Meiliang Bay, followed by Gonghu Bay and Xukou Bay, which may be related to the discharge of the vast majority of pollutants in Meiliang Bay. The industries located in the surrounding areas of the Meiliang Bay are primarily composed of machine shops, paper mills, textile mill, chemical plant, leather industries, power plants, and other manufacturers, all of which contributed to the levels of PAHs in Meiliang Bay. Weinstein et al. (2010) also found that there was a significant positive relationship between drainage area and the level of PAHs concentration. Overall, the presence of PAHs in Meiliang Bay of Lake Taihu indicated the primary contaminant sources were incompletely treated or untreated industrial and municipal effluents, as well as agricultural runoff from surrounding areas, together with riverine water containing industrial and municipal sewage. In contrast, few industries were located in Gonghu Bay and Xukou Bay, leading to relatively lower ecological risk.

Uncertainty analysis

There are several sources of uncertainties within ecological risk assessment, including knowledge uncertainty and variability of the exposure concentrations and toxicity data. Knowledge uncertainty is associated with a lack of knowledge such as misrepresentation of risk assessment model and measurement errors, whereas variability is related to heterogeneity in time and space (Oughton et al. 2008). In this study, uncertainty associated with exposure analysis arose from systematic errors resulting from human or analytical error, even though chemical analysis of PAHs was conducted in one laboratory. Furthermore, the spatial factors such as the DOM concentrations in water, pH, and other water quality parameters that may influence the bioavailability of PAHs were not considered during exposure analysis to aquatic

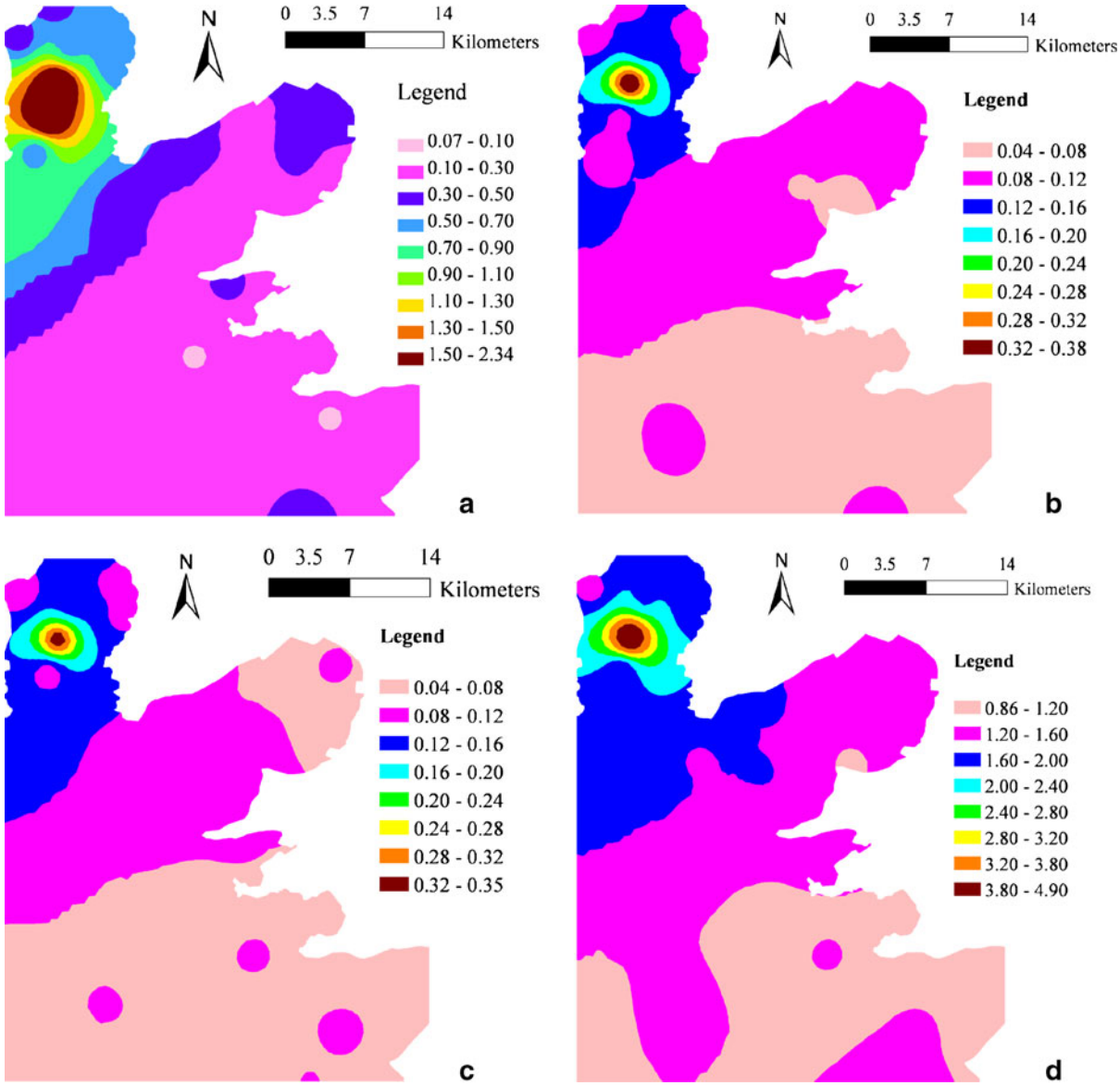

Fig. 3 Spatial distributions of hazard quotients for PAHs with potential risk to the aquatic organisms: (**a**) B[a]P, (**b**) Pyr, (**c**) Ant, and (**d**) Flua

organisms. During the effect analysis, there was a paucity of chronic toxicity data for PAHs; therefore, we used the acute–chronic ratio (ACR) to extrapolate the chronic toxicity data from the acute toxicity data, which may introduce uncertainty into the toxicity data analysis. In addition, the uncertainty of effect toxicity may be associated with the inherent biological variation in the responses of test organisms and inter-laboratory variability related to the conditions under which organisms were cultured, methods, etc.

Within the probabilistic risk assessment, both uncertainty and variability associated with exposure concentrations and toxicity data can be fully considered. In the present study, the impact of parameter uncertainty was quantified by probability distribution functions. The probability distributions of exposure and toxicity concentrations are commonly well fitted by log-normal or log-logistic distributions (Hall et al. 1998; Wagner and Løkke 1991; Meng and Wu 2010). However, an appropriate probabilistic distribution function should

be assigned to the exposure concentrations and toxicity data depending on the availability and quality of the data sets; accordingly, the probabilistic distribution types should be selected on a case-by-case basis. As stated before, the exposure and toxicity concentrations with a right skewed distribution were fitted with a log-normal distribution (Table 1 and Fig. 1), which can be used to describe their uncertainties during ecological risk assessment.

Additionally, sensitivity analysis can also provide quantitative information to identify the relative contributions of the parameters to the output uncertainty. In this study, the contributions of the toxicity data to the resulting distribution of the hazard quotient were 85.9%, 94.7%, 94.3%, 97.7%, 96.1%, 95.5%, and 94.7% for Ace, Ant, B[a]P, Flu, Flua, Phe, and Pyr, respectively, suggesting that the toxicity effect of chemicals was the predominant factor used to determine the resulting distribution of hazard quotients in comparison with the exposure concentration. This likely occurred because there was a limited availability of toxicity data and toxicological information, which resulted in the use of more general and uncertain toxicity data in the risk assessment model. In contrast, the contribution of exposure concentrations to the resulting distributions of the hazard quotient was 61.7% for Chr, suggesting that the exposure concentration affected the ecological risk of Chr to a great extent. Although Monte Carlo simulation was especially useful when there were multiple parameters involved in the complex models for risk assessment (Chiang et al. 2009), this method has several limitations. For example, the potential correlations among the parameters used in the risk model can lead to inaccurate results. Therefore, Spearmen rank correlation coefficient information was further calculated using a Monte Carlo simulation. In the present study, the correlation coefficients between exposure concentration and toxicity data were 0.0056, 0.015, −0.0044, 0.0046, 0.0007, 0.0042, −0.011, and −0.0159 for Ace, Phe, Flu, Ant, Flua, Pyr, Chr, and B[a]P, respectively, suggesting weak correlativity, which demonstrated the results of the sensitivity analysis of uncertainty were reliable.

Conclusions

The ecological risks of eight individual PAHs in Lake Taihu were assessed using probabilistic risk assessment. The rank of the risks was as follows: Flua>B[a]P>Pyr>Ant>Phe>Flu>Ace>Chr. The results suggest that there was potential risk to the aquatic organisms associated with chronic exposures to Flua, B[a]P, Pyr, and Ant, whereas the ecological risks associated with Phe, Flu, Ace, and Chr were negligible. Based on the spatial distributions of risk quotients for Flua, B[a]P, Pyr, and Ant, the highest ecological risk of PAHs was observed in Meiliang Bay, followed by Gonghu Bay and Xukou Bay. In addition, the uncertainty within the ecological risk assessment was quantified using probability distribution functions, risk assessment models, and sensitivity analysis. Sensitivity analysis suggests that toxicity data had a relatively more important impact on the probabilistic distribution of the hazard quotients than exposure concentrations did for all of the evaluated PAHs except for Chr. These observations should be taken into consideration for risk management and alleviation actions in the future.

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