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Health risk analysis of atmospheric polycyclic aromatic hydrocarbons in big cities of China

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Abstract A probabilistic carcinogenic risk assessment of atmospheric polycyclic aromatic hydrocarbons (PAHs) in four big cities (Beijing, Shanghai, Guangzhou, Xiamen) of China was carried out. PAHs levels in these cities were collected from published literatures and converted into BaP equivalent (BaP_{eq}) concentrations. The health risk assessment models recommended by US EPA were applied to quantitatively characterize the health risk values of PAHs. Monte Carlo simulation and sensitivity analysis were applied to quantify uncertainties of risk assessment. The results showed that BaP_{eq} concentrations of four cities were all higher than the newest limited value (1 ng/m^3) of China. Health risk assessment indicated that atmospheric PAHs in Guangzhou and Xiamen posed no or little carcinogenic risk on local residents. However, the PAHs in Beijing and Shanghai posed potential carcinogenic risk for adults and lifetime exposure. Notwithstanding the uncertainties, this study provides the primary information on the carcinogenic risk of atmospheric PAHs in studied cities of China.

Keywords Polycyclic aromatic hydrocarbons · Health risk · Air · Monte Carlo simulation

Introduction

Polycyclic aromatic hydrocarbons (PAHs), as a group of persistent organic pollutants, have been found to be widely distributed in atmosphere because of their moderate vapor pressure, low solubility and low reactivity. Previous studies identified that PAHs in atmosphere were produced mainly through anthropogenic sources, such as residential heating (coal, oil and wood), vehicle exhausts, refuse burning, coke production, and industrial processes (Sun et al. 2011; Zhao et al. 2012). And the atmospheric PAHs in urban districts are higher than these in rural areas (Xu et al. 2006; Liu et al. 2007a). Therefore, there are many researches concerning the characteristics of distribution and sources of atmospheric PAHs in urban areas of China, especially in the big cities.

Due to potential carcinogenic effects of PAHs after long-term exposure (Kogevinas et al. 2003; Bosetti et al. 2007), it is necessary to determine carcinogenic risk of atmospheric PAHs in urban areas of China to protect local residents' health. However, little information is available from previous literatures. Human health risk assessment (HHRA) is a useful approach to quantify potential harmful index of environmental contaminants. In generally, human health risk assessment of chemicals includes four steps: data collection and analysis, exposure assessment, toxicity assessment and risk characterization. Exposure assessment is usually based on the site-specific chemical and resident parameters. Toxicity assessment refers to the toxicity data from animal experiment. Based on the exposure and toxicity assessment, health risk values of chemical can be characterized. Combined with probabilistic approaches, such as Monte Carlo simulation and sensitivity analysis, HHRA can provide the risk assessor with a flexible tool to conduct estimates of uncertainty as well as stochastic properties of both exposure and toxicity assessment. The probabilistic HHRA has been widely applied to characterize the carcinogenic risk of PAHs in the environment (Chowdhury et al. 2009; Wu et al. 2010; Shi et al. 2011).

In this study, we gathered the published monitoring data of atmospheric PAHs in four typical big cities of China:

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Beijing, Shanghai, Xiamen and Guangzhou. The concentration of multi-component PAHs in these cities were gathered and conserved into benzo(a)pyrene equivalent (BaP_{eq}) concentration to calculate their exposure level. A probabilistic HHRA was applied to evaluate and quantify carcinogenic risks of PAHs for different age groups (children, teens and adults). The uncertainties during risk assessment were analyzed by Monte Carlo simulation and sensitivity analysis.

Methodology

Exposure assessment

Atmospheric PAHs levels in Beijing, Shanghai, Guangzhou and Xiamen were obtained from published literatures. Their information is shown in Table 1. Although the sampling and digestion procedures in different literatures were not uniform, we hypothesized that the levels of atmospheric PAHs from different literatures represented the contaminant status of studied areas, and the differences in data quality were not taken into consideration.

The BaP_{eq} concentrations of multi-component PAHs in every city were calculated by multiplication of individual PAH concentration and its potency equivalency factor (PEF), which were applied to assess the exposure levels of PAHs for local residents. The PEF values used in this study were obtained from the previous reports (Levengood and Schaeffer 2011; Wu et al. 2011). For PAHs in air, inhalation has been approved to be the main exposure pathway. We applied the inhalation exposure assessment model recommended by US EPA to calculate the daily exposure dose (DED) of BaP_{eq}, which was shown as the following Eq. (1):

$$\mathrm{DED} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times cf \qquad (1)$$

where DED is daily exposure dose of BaP_{eq} through inhalation pathway (mg/(kg day)), C is the concentration of BaP_{eq} in air (ng/m^3), IR is the inhalation rate of air (m^3/day), EF is the exposure frequency (day/year), ED is the exposure duration (year), BW is the body weight (kg), and AT is the averaging time (day), cf is the conversion factor, in this study, $cf = 10^{-6}$.

Risk characterization

Carcinogenic risk (CR) associated with inhalation exposure was calculated using the following Eq. (2), which was adapted from USEPA (1989).

$$\mathrm{CR} = \mathrm{DED} \times \mathrm{CSF} \times \left(\frac{\mathrm{BW}}{70}\right)^{\frac{1}{3}} \qquad (2)$$

where CR is the probability of developing cancer over a lifetime as a result of exposure to atmospheric PAHs, DED is the daily exposure dose obtained from the Eq. (1), CSF is the cancer slope factor, which quantitatively defines the relationship between exposure doses of the carcinogen and increased carcinogenic risk. The CSFs used were derived assuming a body weight of 70 kg, which was different from actual condition of the exposure population in different age groups. Thus, the CSF values were extrapolated to the actual body weight of different age groups by multiplying the conversion factor, $(BW/70)^{1/3}$, which was recommended by USEPA (2004).

Uncertainty analysis

For probabilistic health risk assessment, the uncertainties of output values should be analyzed. In this study, Monte Carlo simulation was implemented to quantify the uncertainties and their impacts on the expected risk values. The probability distributions of input parameters employed for risk assessments were evaluated by fitting distribution functions (Liu et al. 2010; Zeng et al. 2013). Sensitivity analysis was used to determine the significance of input parameters on the basis of rank correlation coefficients between input and output values during Monte Carlo simulations. The Monte Carlo simulation and sensitivity analysis were all carried out by R language software (version 2.14).

Table 1 Basic information and BaP_{eq} concentration in air of four cities of China

City	Year	BaP_{eq} (ng/m^3)			References
		Mean	SD	90 %	
Beijing	2003–2006	43.67	312.27	77.33	(Hou et al. 2006; Okuda et al. 2006; Liu et al. 2007a, b; Zhang et al. 2009)
Shanghai	2003–2006	14.82	45.09	32.70	(Guo et al. 2004; Cheng et al. 2007; Chen et al. 2011)
Guangzhou	2001–2004	9.74	10.54	20.43	(Bi et al. 2003; Li et al. 2006; Tan et al. 2006; Duan et al. 2007)
Xiamen	2004–2009	2.11	3.09	4.69	(Hong et al. 2007; Wang et al. 2007; Zhao et al. 2010, 2011)

Results and discussion

Exposure and toxicity characterization

The concentrations of atmospheric PAHs in Beijing, Shanghai, Guangzhou and Xiamen were gathered from sixteen published literatures. The basic information on data sources was shown in the Table 1. Then the BaP_{eq} concentrations of every city were calculated according to the PEFs of individual PAHs. The probability distributions of obtained BaP_{eq} concentrations were evaluated by fitting distribution functions with the assistance of Kolmogorov–Smimov test, Anderson–Darling test and χ^2 test. The results showed that probability distributions of BaP_{eq} concentrations all fit to lognormal distribution. The data on BaP_{eq} concentrations during Monte Carlo simulation and health risk assessment is also shown in Table 1. In order to avoid the overestimation of Monte Carlo simulation, in this study, 90th values were used as high-end estimates instead of the maximal concentrations. As shown in Table 1, among the four cities, Beijing had the highest BaP_{eq} concentration, followed by Shanghai and Guangzhou. Xiamen had the lowest Bap_{eq} concentration. The limited value of atmospheric BaP has been improved from 10 to 1 ng/m^3 in the newest ambient air quality standard of China (GB3095-2012). All the 90th BaP_{eq} concentrations calculated in the four cities were higher than the limited value, which indicated that the atmospheric PAHs in studied four cities might pose potential adverse effects on local residents.

In order to effectively estimate the exposure levels of BaP_{eq} in different age ranges, three age groups, which were children, teens and adults, were considered in this study. The parameters for these age groups used in exposure assessment were gathered. Among them, body weight were obtained from statistical data of Ministry of Health of the People's Republic of China, other parameters were obtained from the recommended values of USEPA. Their probability distributions were also estimated by fitting distribution functions, which are shown in the Table 2. Based on above data and Eq. (1), the daily exposure doses and their distribution for different cities could be calculated.

For toxicity assessment of BaP, its CSF also needed to be estimated. We chose the values reported by Chen and Liao (2006). They referred the CSF for BaP inhalation exposure on hamster, and estimated the CSF values of inhalation exposure for human at a lognormal distribution with a geometric mean 3.14 [mg/(kg day)]$^{-1}$ and a geometric standard deviation 1.80.

Health risk values for different cities

Based on above exposure and toxicity assessment, the carcinogenic risks of BaP_{eq} in studied four cities were characterized for children, teens and adults. The lifetime carcinogenic risk (LCR) in different cities was calculated by summing the carcinogenic risk values of above three age groups. Table 3 showed that distribution of carcinogenic risk values in different age groups in terms of percentage. The 90th risk values were chosen as the high-end estimates. Among the four cities, Beijing had highest carcinogenic risk, followed by Shanghai. Lowest CR was found in Xiamen. Under most regulatory programs, a CR value over 1.00E−05 indicates potential carcinogenic risk (De Miguel et al. 2007). For Guangzhou and Xiamen, the 90th CR values for children, teens, adults and lifetime were all lower than 1.00E−05, which indicated that atmospheric PAHs of both cities could pose no or little potential carcinogenic risk. However, for Beijing, the 90th CR values for adults and lifetime were higher than 1.00E−05. The probabilities higher than 1.00E−05 were 15.2 and 22.1 % for adult and lifetime, respectively. For Shanghai, although the 90th CR values for children, teens and adults were lower than 1.00E−05, the probability higher than the value for lifetime exposure was 13.1 %. These results indicated that atmospheric PAHs of Beijing and Shanghai might pose potential carcinogenic risk on the local residents, which should be paid more attentions.

Table 2 Values and probability distributions of parameters used in exposure assessment

Units	Distribution[a]	Children	Teens	Adults
Age (year)	–	0–10	11–20	21–70
IR[b] (m^3/day)	Lognormal	LN (8.79, 1.45)	LN (13.61, 1.16)	LN (12.34, 1.21)
EF (day/year)	–	365	365	365
ED (year)	Uniform	0–10	0–10	0–50
BW[c] (kg)	Lognormal	LN (16.66, 1.48)	LN (46.35, 1.18)	LN (57.04, 1.1)
AT[b] (day)	–	25,550	25,550	25,550

[a] Geometric mean and geometric standard deviation were expressed as LN (gm, gsd) in lognormal distribution

[b] Adapted from USEPA

[c] Adapted from the statistical data of China

Table 3 Carcinogenic risk values in different cities

	10 %	30 %	50 %	70 %	90 %
Beijing					
Children	1.66E−08	1.02E−07	3.39E−07	1.12E−06	6.25E−06
Teens	1.30E−08	7.75E−08	2.61E−07	8.82E−07	4.67E−06
Adults	5.03E−08	3.10E−07	1.02E−06	3.27E−06	1.77E−05
Lifetime	1.30E−07	6.42E−07	1.96E−06	5.94E−06	3.04E−05
Shanghai					
Children	1.89E−08	9.72E−08	2.68E−07	7.18E−07	2.95E−06
Teens	1.58E−08	7.51E−08	2.06E−07	5.35E−07	2.13E−06
Adults	6.20E−08	2.99E−07	7.97E−07	2.08E−06	8.06E−06
Lifetime	1.55E−07	6.13E−07	1.50E−06	3.65E−06	1.30E−05
Guangzhou					
Children	5.16E−08	1.81E−07	3.92E−07	8.21E−07	2.20E−06
Teens	4.13E−08	1.47E−07	3.14E−07	6.20E−07	1.54E−06
Adults	1.69E−07	5.68E−07	1.21E−06	2.43E−06	6.01E−06
Lifetime	4.60E−07	1.17E−06	2.20E−06	4.06E−06	9.42E−06
Xiamen					
Children	7.83E−09	3.05E−08	7.22E−08	1.61E−07	4.81E−07
Teens	6.69E−09	2.41E−08	5.53E−08	1.20E−07	3.53E−07
Adults	2.62E−08	9.57E−08	2.13E−07	4.63E−07	1.33E−06
Lifetime	7.01E−08	1.95E−07	3.98E−07	7.92E−07	2.11E−06

Uncertainty and sensitivity analysis

In order to identify the most influential parameters contributing to the output risk values, sensitivity analysis was carried out during risk assessment. The results showed that BaP_{eq} concentration was the most influential parameter for all age groups, whose correlation coefficients were in the range of 0.58–0.92, followed by inhalation CSF of BaP, in the range of 0.26–0.48. Chen and Liao (2006) studied the health risk assessment on human exposed to atmospheric PAHs in Taiwan, and also found that inhalation CSF and BaP_{eq} concentration were the important influential parameters for risk estimation. These results indicated that the better definition of probability distribution for BaP_{eq} concentration and CSF could increase the accuracy of risk assessment.

Although Monte Carlo simulation and sensitivity analysis were applied to characterize the uncertainties and influences of input parameters on risk assessment, there were still other uncertainties that should be paid attention. Firstly, for exposure assessment, due to differences of sampling data and digestion procedures, PAHs levels determined in the different studies were different. In addition, some researches just determined the PAHs in gas phase or particle phase, which could underestimate the levels of atmospheric PAHs. These might cause the uncertainties of exposure assessment. Secondly, due to limited CSF values for multiple PAHs, the PAHs levels were converted into BaP_{eq} concentration by using PEFs. Then, the inhalation CSF value of BaP was applied to evaluate the health risk of PAHs. However, the PEFs and inhalation CSF of BaP were all obtained from animal experiments, which were influenced by experimental conditions and processes. The toxicity data of PAHs could be another source of uncertainties. Finally, although the body weight were obtained from statistical data of Chinese, due to limited data, inhalation rate of air was directly obtained from the recommended value of USEPA, which could be not specific for Chinese. The specific parameters might also raise the uncertainties of risk assessment.

Conclusions

The BaP_{eq} concentrations of Beijing, Shanghai, Guangzhou and Xiamen were all higher than the newest limited value (1 ng/m^3) of China, which indicated the potential adverse health risk for local residents. Then, probabilistic health risk assessment showed that atmospheric PAHs levels in Guangzhou and Xiamen posed no or little carcinogenic risk for local residents. However, for Beijing and Shanghai, the atmospheric PAHs might pose potential carcinogenic risk for adults and lifetime exposure. Notwithstanding the uncertainties, this study firstly calculated and compared the carcinogenic risks of atmospheric PAHs in four big cities of China. The results are useful for the control and management of atmospheric PAHs in these cities.

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Conflict of interest The authors declare that they have no conflict of interest.

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