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Pollution level, inhalation exposure and lung cancer risk of ambient atmospheric polycyclic aromatic hydrocarbons (PAHs) in Taiyuan, China

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ABSTRACT

Passive air samplers were deployed to collect both gas and particulate phase polycyclic aromatic hydrocarbons in Taiyuan between 2009 and 2010. Annual average concentrations of BaP equivalent concentration ($B[a]P_{eq}$) in background, rural and urban areas were 2.90 ± 0.29, 23.2 ± 30.8 and 27.4 ± 28.1 ng/m^3, respectively, with higher concentration in the winter than in other seasons. The median $B[a]P_{eq}$ concentrations of annual inhalation exposure were estimated to be in the range of 103–347 ng/d for all population groups in rural as well as in urban areas. The median values of incremental lifetime cancer risk (ILCR) induced by whole year inhalation exposure for all groups were basically larger than 10^{-6}, with higher values in winter than in other seasons and in urban than in rural area. In the same season and area, the ILCR of adults was larger than other age groups and that of females was a little higher than males.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of fusedring aromatic compounds that are formed during the incomplete combustion of organic material (McGrath et al., 2007). PAHs have generated considerable interest ever since they were recognized as a carcinogenic class of compounds in the late twenties (Okona-Mensah et al., 2005). Several individual PAHs such as benzo(a)pyrene (BaP), chrysene (CHR), indeno(1,2,3- c,d)pyrene (IcdP) and benzo(b)fluoranthene (BbF) have produced carcinogenic, mutagenic, and genotoxic effects in animal experiments (Deutsch-Wenzel et al., 1983).

In recent years, environmental PAH concentrations have increased in many industrialized and developing countries. In China, the PAHs emissions contributed over 20% of the total global PAH emissions in 2004 (Zhang et al., 2008) and the high emissions have resulted in heavy contamination of various environmental media, especially ambient air (Zhang et al., 2007). Lung cancer has been ranked as the fourth and fifth leading causes of cancer death in Chinese males and females, respectively (Jiang et al., 2008) due to serious air pollution. Causes of human lung cancer have always been associated with the inhalation exposure to PAHs (Chen and Liao, 2006). Therefore, exposure and health risk assessment of atmospheric PAHs is very essential for effective environmental management (Asante-Duah, 2002). So far, researches concerning respiratory exposure and lung cancer risk assessment of PAHs are quite limited in China (Bai et al., 2009; Zhang et al., 2009).

Taiyuan, the capital of Shanxi Province and an old industrial city, becomes one of the most polluted cities in China and PAHs may be a threat to the health of local residents (Zhang et al., 2009) mainly due to the high intensity of pollutant emission (Zhang et al., 2009). Thus, assessing the health risk level of citizens in Taiyuan associated with environmental exposure to PAHs is quite urgent. PAHs concentrations of foods consumed by local residents in Taiyuan were monitored in 2008 and the incremental lifetime cancer risks (ILCRs) were found to be larger than 10^{-4} at the 74.5th, 78.7th, 60.6th, 77.4th, 75.3th, 79.5th, 60.8th and 77.9th percentile for children, adolescents, adults and seniors of male as well as the above groups of female, respectively (Xia et al., 2010), implying significant dietary cancer risk. However, lung cancer risk level induced by inhalation exposure to atmospheric PAHs for citizens in Taiyuan is currently unavailable.

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The objective of this study is to determine the pollution level, quantify the inhalation exposure and further score the incremental lifetime cancer risk for citizens in Taiyuan, investigating the changes in both exposure and risk for different seasons, areas and population groups. To determine the overall uncertainty in predicted risks, the uncertainty resulting from the assessment of exposure was propagated through the risk characterization process using the Monte Carlo simulation. Moreover, a detailed sensitivity analysis was conducted to identify the input variables that were critical to the accuracy of the risk assessment.

2. Materials and methods

2.1. Sampling

Passive air samplers with PUF disk and glass fiber filter (GFF) were used to collect PAHs in the gas and particulate phases, respectively. Detailed calibration method and uptake rates of this sampler were described previously (Tao et al., 2009). Volume concentrations of PAHs were calculated based on calibration equations derived in Supporting Material (S.2, Equation S1 and S2). Passive air samplers were deployed at 25 background, rural and urban sites in Taiyuan, the capital of Shanxi Province and an old industrial city in China (S.1, Fig. S1 and Table S1), in the spring, summer-autumn and winter between 2009 and 2010. The sampling periods were 70, 155 and 140 days for spring, summer–autumn, and winter, respectively, and the sampling period in the winter included the residential heating time. Two identical samplers were deployed at each site on rooftops or open areas to avoid airflow obstruction (1.5–20 m heights). Detailed information on atmospheric sampling sites was presented in Supporting Material (S.1, Table S1). After sampling, all PUF disks were stored at −18 °C. GFFs were equilibrated in a desiccator (25 °C) for 24 h and weighed before and after the sampling, in accordance with USEPA Method (1980). Before sampling, the PUF disks were cleaned by extracting them in a Soxhlet with a 1:1 mixture of n-hexane and acetone for 8 h and the GFFs were cleaned by baking them in a furnace at 450 °C for 4 h.

2.2. Analytical procedure

PUF disks were Soxhlet extracted with150 ml 1:1 mixture of n-hexane and acetone at 52 °C for 8 h. GFFs were subjected to microwave extraction (MARS2X-press, CEM, USA) with 25 ml 1:1 mixture of n-hexane and acetone, being heated to 100 °C at 10 °C/min and then held for 10 min. After concentration with a vacuum rotary evaporator (R-201, Shanghai, China) at 37 °C, total extracts were transferred to the alumina silica gel column for purification. The eluted mixture from the column during cleanup was first concentrated to near dryness. The residue was then transferred and diluted with n-hexane and brought to exactly 1.0 ml by nitrogen blowdown (Eyela MG-1000) at room temperature (25 °C). The samples were sealed in vials and stored at −4 °C before analysis. Quantitative analysis of the air sample extracts was done by gas chromatography with mass spectrometer detector (Agilent 6890GC/5973MSD). A 30 m × 0.25 mm i.d. × 0.25 μm film thickness HP-5MS capillary column (Agilent Technology) was used. GC temperature was programmed from an initial 60 °C before commencing at 5 °C/min up to 280 °C, with a final holding time of 20 min. Helium was used as the carrier gas. A 1.0 μl aliquot of the extract was injected while the injector port was held at 280 °C and operated in splitless mode at a flow rate of 1.0 ml/min. The head column pressure was 30 kPa. The mass spectrometer was operated in scan mode with an electron impact ionization of 70 eV, an electron multiplier voltage of 1288 V, and an ion source of 230 °C. Concentrations were determined for 15 PAHs in all samples. They were 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).

2.3. Quality control

Quantification was performed by the internal standard method using 2-fluoro-1,1′-biphenyl and p-terphenyl-d_{14} (2.0 μg/ml; J&K Chemical, Beijing, China). All solvents used were analytical grade (Beijing Chemical Reagent, Beijing, China) and purified by distillation prior to use. Alumina and silica gel (80–200 mesh; Dikma, China) were heated at 650 °C in a muffle furnace (DLII-9, Beijing, China) for 10 h, kept in a sealed desiccator, and reactivated at 130 °C for 4 h immediately prior to use. All glassware was cleaned using an ultrasonic cleaner (KQ-500B, Kunshan, China) and heated to 400 °C for 6 h.

The field and laboratory blanks were analyzed, and the concentration of target PAHs in the field blanks were higher than laboratory blanks and more than one order of magnitude lower than real samples for both PUF disks and GFFs. All the results of air samples were field blank corrected. Recovery of individual PAHs ranged from 78% to 103% with a mean value of 89% for PUF disks, whereas that varied from 80% to 101% with a mean value of 88% for GFFs. Data analyzed in the article were not corrected for recoveries. The detection limits were in the range of 0.172–1.23 ng/ml.

2.4. Inhalation exposure estimates

The carcinogenic risk of a PAHs mixture is often expressed by its BaP equivalent concentration (B[a]P_{eq}). The B[a]P_{eq} of atmospheric PAHs (BEC) was calculated according to Equation (1):

$$\mathrm{BEC} = \sum_{i=1}^{n} Ci \times \mathrm{TEF}i \quad (1)$$

where C_i = concentration of PAH congener i; TEF_i = the toxicity equivalency factor (TEF) of PAH congener i (S.3, Table S2). For individual PAH concentration, when a result was below the limit of detection (LOD), the value was assumed to be half of the respective LOD. The carcinogenic potencies of 15 PAHs were estimated as the sum of each individual B[a]P_{eq}. We treated C, which followed lognormal distribution, in Equation (1) probabilistically.

Taiyuan consists of background, rural and urban areas. There are no citizens living in background area. In both rural and urban areas, the citizens were divided into eight population groups according to the age and gender: children (4–10 years), adolescents (11–17 years), adults (18–60 years), and seniors (61–70 years) of male as well as the above groups of female. Daily inhalation exposure level (E) for each population group was calculated as follows:

$$E = \sum_{i=1}^{n} \mathrm{BEC}i \times \mathrm{IR} \times Ti \quad (2)$$

where T_i = daily exposure time span in the ith area (S4); $\mathrm{BEC_i}$ = B[a]P_{eq} in the ith area (ng/m^3); IR = inhalation rate (m^3/day) (S4, Table S13). We treated BEC and IR, which followed lognormal and normal distribution, respectively, in Equation (2) probabilistically. Detailed information on inhalation exposure calculation was presented in Supporting Material (S4).

2.5. Cancer risk estimates

The incremental lifetime cancer risk (ILCR) of population groups in Taiyuan caused by PAHs inhalation exposure was calculated based on Equation (3).

$$\mathrm{ILCR} = \mathrm{SF} \times \mathrm{E} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{CF}/(\mathrm{BW} \times \mathrm{AT}) \quad (3)$$

where ILCR = the incremental lifetime cancer risk of the inhalation exposure (dimensionless); SF = the *cancer* slope factor for BaP inhalation exposure [a lognormal distribution with a geometric mean of 3.14 (mg kg^{-1} day^{-1})$^{-1}$ and a geometric standard deviation of 1.80](Chen and Liao, 2006); E = the daily inhalation exposure level (ng/d); EF = the exposure frequency (day/year) (S.5); CF = conversion factor (10^{-6} mg/ng); BW = body weight (kg) (S.5, Table S13); AT = average lifespan for carcinogens (25550 day). We treated SF E and BW, which obeyed lognormal, lognormal and normal distribution, respectively, in Equation (3) probabilistically. Detailed information on cancer risk calculation was presented in Supporting Material (S.5).

2.6. Uncertainty analysis

A Monte Carlo simulation using matlab6.5 software was implemented to deal with the uncertainties in the risk assessment in this study. We performed independent runs at 1000, 3000, 5000, and 10,000 iterations with each parameter sampled independently from the appropriate distribution at the start to test the convergence and the stability of the numerical output. The result showed that 5000 iterations are sufficient to ensure the stability of results. As the results manifested the effects of uncertainty in the input parameter statistics on the estimation, sensitivity analysis using matlab6.5 software could be done to find the variables that affect the risk most. Rank correlation coefficients between each input variable and the output (risk) were calculated, and then by squaring the output variance and normalizing it to 100%, the contribution of each input variable to the output (risk) variance was assessed, and the sensitivity of each input variable relative to one another was evaluated.

3. Results and discussion

3.1. Pollution level of atmospheric PAHs

Atmospheric concentrations (particulate + gas phase) of PAHs in Taiyuan are shown in Fig. 1 and the detailed results are presented in Supporting Material (S.3, Table S3–S11). In background area, the concentrations (particulate + gas phase) of total 15 PAHs and B[a]P_{eq} were in the range of 57.5 ± 5.32–73.6 ± 2.22 and