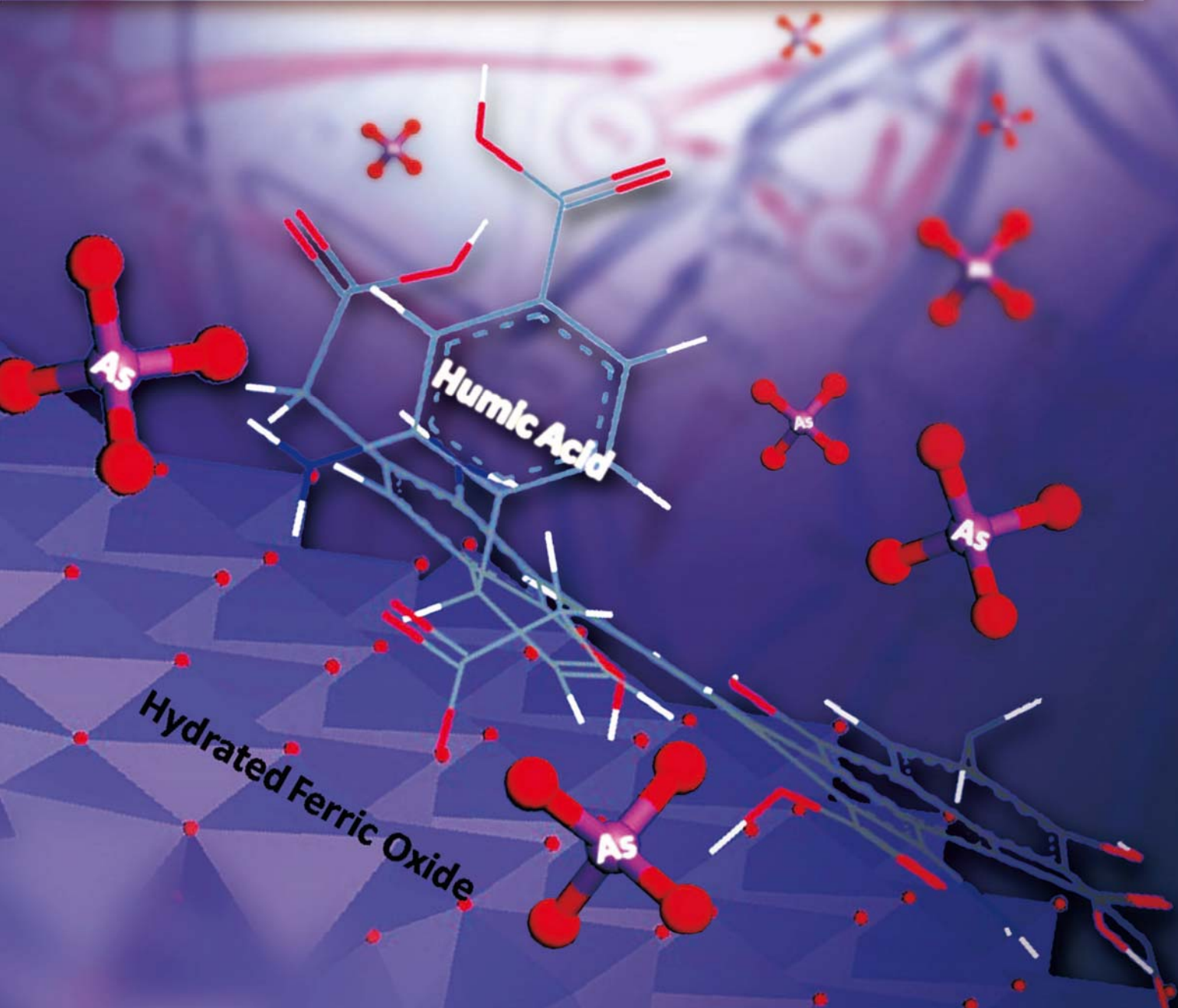


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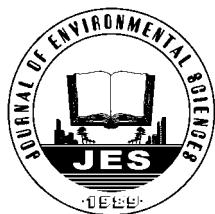
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## Exposure measurement, risk assessment and source identification for exposure of traffic assistants to particle-bound PAHs in Tianjin, China

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### ABSTRACT

To investigate the levels of exposure to particulate-bound polycyclic aromatic hydrocarbon (PAH) and to estimate the risk these levels pose to traffic assistants (TAs) in Tianjin (a megacity in North China), a measurement campaign (33 all-day exposure samples, 25 occupational-exposure samples and 10 indoor samples) was conducted to characterize the TAs' exposure to PAHs, assess the cancer risk and identify the potential sources of exposure. The average total exposure concentration of 14 PAHs was approximately  $2871 \pm 928 \text{ ng/m}^3$  (on-duty), and  $1622 \pm 457 \text{ ng/m}^3$  (all-day). The indoor PAHs level was  $1257 \pm 107 \text{ ng/m}^3$ . After 8000 Monte Carlo simulations, the cancer risk resulting from exposure to PAHs was found to be approximately  $1.05 \times 10^{-4}$ . A multivariate analysis was applied to identify the potential sources, and the results showed that, in addition to vehicle exhaust, coal combustion and cooking fumes were also another two important contributors to personal PAH exposure. The diagnostic ratios of PAH compounds agree with the source apportionment results derived from principal component analysis.

## Introduction

Polycyclic aromatic hydrocarbons (PAHs) are unique substances detected in various environments, that are also considered toxic, carcinogenic and mutagenic (Arfsten et al., 1996; Fent and Batscher, 2000). Most of the PAHs in the atmosphere arise from the emissions resulting from combustion, including natural emissions such as the forest fires (Gabos et al., 2001) and volcanoes (Zolotov and Shock, 2000), and anthropogenic emissions from fossil fuel (coal, petroleum and so on) combustion (Wild and Jones, 1995), tobacco smoke (Vanrooij et al., 1994), cooking operations (Mumford et al., 1995) and other domestic combustion processes (Oanh et al., 1999).

With the increasing numbers of vehicles, motor vehi-

cle exhaust has become a significant emission source of PAHs. Numerous studies have reported a high level of PAH exposure for traffic policemen. A total of 28 traffic policemen and 10 road builders were studied for PAH exposure in downtown Budapest, Hungary, during their working hours (Szaniszló and Ungváry, 2001). The results indicated that the PAH exposure level for traffic policemen was  $60.7 \text{ ng/m}^3$ , comparable to the exposure levels in most other European cities. Another personal exposure investigation performed among 44 traffic policemen and 45 office policemen in Bangkok, Thailand found that PAH exposure among traffic policemen ( $72.79 \text{ ng/m}^3$ ) was much higher than exposure levels for office staff ( $6.88 \text{ ng/m}^3$ ) (Ruchirawa et al., 2002). In China, Liu et al. (2007b, 2007c) reported the PAH exposure levels for policeman in Beijing, and the results showed that exposure levels to 16 gaseous and particulate PAHs for the traffic police were found to be  $(1525 \pm 759)$  and  $(148 \pm 118) \text{ ng/m}^3$ ,

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respectively, representing 2–2.5 times higher levels than at the control sites. The daily inhalation exposure of policeman was about 277 ng/(kg·day). In Tianjin City near Beijing, the traffic police have an exposure level (total for 16 PAHs of approximately 867.5 ng/m<sup>3</sup>) (Hu et al., 2007) close to the exposure level for Beijing police.

From November of 2004, in response to the worsening traffic situation resulting from the huge increment in automobiles, the first batch of traffic assistants (who assist the traffic police to manage traffic personnel, primarily responsible for the order of pedestrians and vehicles in heavy traffic road sections or junctions) (approximately 1100) was recruited to assist the traffic policemen in Tianjin. At present, the number of traffic assistants (TAs) in this city is approximately 3000. Although PAHs from automobile exhaust in their occupational environment may be the most important part of the exposure of TAs to PAHs, PAH exposure from cooking, tobacco smoke and other sources can play moderate roles. The TAs constitute individuals who will receive exceptional multi-way exposure.

From the short review above, some questions have been raised concerning the exposure of the TAs to PAHs. What is the PAH exposure level and what is the exposure profile of TAs in Tianjin? What type of risk does the inhalation of PAHs pose? Is the occupational environmental exposure the only or the major source for total inhalation exposure?

In this study, using survey questionnaires, personal sampling and chemical analysis, the PAH exposure level and profile were investigated and exposure risk was then assessed by Monte Carlo simulation (Hu et al., 2007; Liu et al., 2007b). Based on the characterization of the exposure profile and the personal exposure database, the data was rearranged by the principal component analysis method (Larsen and Baker, 2003; Manoli et al., 2004) and the contribution of major sources to the inhalation exposure could be derived. The risk assessment results can provide information on the risk for this special population. All of the above results can improve the reliability of the exposure-response relationship and provide the sources of the major contributors, which is important to control the exposure and promote the health of TAs for the purpose of environmental and occupational health management.

## 1 Methods

### 1.1 Sampling strategy

All samples were collected in Tianjin City from July to August in 2007. Tianjin is a typical metropolis of China, with a population of approximately 11.7 million. Tianjin is located in the north of China, at the lower reaches of the Haihe River and adjacent to the Bohai Sea. Tianjin has distinct seasons with an annual mean temperature of 13.1°C, and the average rainfall is 500–700 mm. Tianjin

is an important industrial center and a well-developed hub with a sea-land-air transportation network. At the end of 2008, there were approximately 909,600 vehicles in Tianjin (National Statistics Bureau, 2008).

Sixteen representative sites (T-shaped crossing centers and intersection centers, Fig. 1) in Tianjin were selected for sampling and 58 TAs were recruited as subjects in this study. Everyone was trained to record the time activity table and operate the sampling instruments. All of the samples can be organized into 3 classes: Class 1 (on-duty exposure samples), 25 samples collected by fixing the personal environmental monitor (PEM) in the center of the working road during the working time of the TAs (from 8:00 a.m. to 2:00 p.m.); Class 2 (all-day exposure samples), 33 samples collected by carrying the PEM with the subject for 24 hr; and Class 3 (indoor samples), 10 samples collected by fixing the PEM in the subject's home for 24 hr (from 8:00 a.m. to next day at 8:00 a.m.).

PM<sub>10</sub> samples were collected on quartz filters (37 mm, Pall Filter Co., USA) by Model 200 personal environmental monitors (PEM<sup>TM</sup>, SKC Co., USA) and the size-cut head was connected to the PEM by a plastic tube (75 cm). The sample head was fixed on the collar to collect personal samples in the breathing zone, and the samplers were operated at a constant flow rate of 10 L/min. Both the indoor and all-day samples were collected by fixing the sample head at 1.5 m above ground level. The sampler was set in the center of the room or road center. Before sampling, all quartz filters were pretreated by baking in an oven at 450°C for 4.5 hr to volatilize any organic contaminants. Samplers were calibrated by a Buck calibrator (M30, Buck Co., USA). During the whole sampling, the temperature ranged from 19.2 to 35.1°C and the atmospheric pressure ranged from 99,400 to 101,700 Pa. The lowest relative humidity was 27%. After sampling, the filters were packed with aluminum foil to avoid light and were conditioned at a constant temperature of 20°C with a constant relative humidity of 45% for 12 hr before weighing. The filters were stored in a refrigerator (below –15°C) until extraction. All filters were extracted within 2 weeks after sampling.

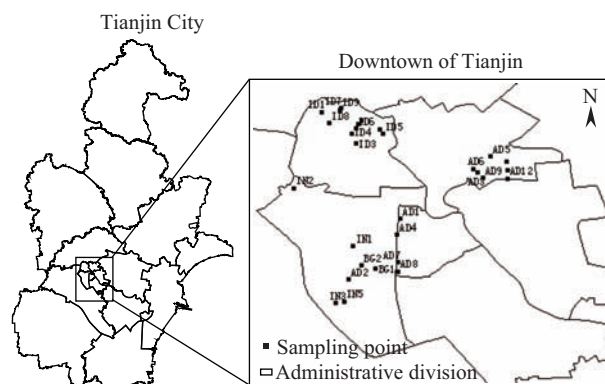


Fig. 1 Map of the sample site. IN1 to IN5: indoor samples, AD1 to AD12: all-day samples, ID1 to ID12: on-duty samples.

## 1.2 Chemical analysis

The PAHs on filters were extracted with 15 mL dichloromethane (MeCl<sub>2</sub>) in an ultrasonic extractor (AS3120, AutoScience Co., Tianjin, China) for 15 min and then the insoluble part was separated by centrifugation (0406-1, Shanghai Medical Instrument Co., Shanghai, China) at 3000 r/min for 10 min. The procedure was repeated 3 times. The solvent volume was reduced using a rotary evaporator (RE52CS, YaRong Biochemistry Instrument Co., Shanghai, China) under  $-20,000$  Pa and  $40^{\circ}\text{C}$  after extraction. A silica gel column previously activated at  $450^{\circ}\text{C}$  for 4 hr was used for separating the compound groups. The aliphatic compounds were eluted with 40 mL of hexane, the PAH fraction was collected with 120 mL of 1:1 (V/V) dichloromethane/hexane mixture, and the remaining polar components were eluted with 50 mL of dichloromethane. The PAH fraction was then evaporated ( $-80,000$  Pa and  $45^{\circ}\text{C}$ ) and reduced under N<sub>2</sub> to approximately 0.5 mL. All sample extracts were transferred to individual amber 2 mL glass HPLC vials and stored in the refrigerator ( $-4^{\circ}\text{C}$ ) until subsequent analysis.

The external standard solutions were prepared including 18 PAHs: naphthalene (Nap), acenaphthylene (Aci), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluo), pyrene (Pyr), benz(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbFlu), benzo(k)fluoranthene (BkFlu), benzo(e)pyrene (BeP), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (InP), dibenz(a,h)anthracene (DiBahA), benzo(g,h,i)perylene (BghiP) and coronene (Cor). The mixture of EPA610 (48743, 16 PAHs of EPA listed), BeP (36962) and Cor (36963) were purchased from Supelco Co. in USA. These standard PAHs were diluted with dichloromethane to 4 different concentration levels in the range of 0.08–40  $\mu\text{g}/\text{mL}$ .

The 18 PAHs were identified and quantitated using a GC-MSD system (GC6890-MSD5975, Agilent, USA). The GC used a DB5-MS (J&W Scientific Technologies, USA) fused silica capillary column (30 m  $\times$  0.25 mm i.d.  $\times$  0.25  $\mu\text{m}$  phase thickness), and helium was employed as carrier gas at 1.2 mL/min. Then 1  $\mu\text{L}$  of each extract was injected into the GC in splitless mode using an auto injector (7683B, Agilent, USA). The GC injector port was maintained at  $290^{\circ}\text{C}$ . The GC oven was programmed with an initial temperature of  $70^{\circ}\text{C}$ , held for 2 min, heated at a rate of  $8^{\circ}\text{C}/\text{min}$  to  $295^{\circ}\text{C}$ , and held for 15 min. The transfer line between the GC and MSD was held at  $280^{\circ}\text{C}$ . The MSD was operated under the following conditions: Electron ionization mode with energy of (nominal) 70 eV, heptacosane as tuning standard, the Qua-Detector temperature at  $230^{\circ}\text{C}$ , and mass range of 50–600 amu. Retention time comparisons to authentic standards and reference mass spectra (National Institute of Standards and Technology) were used to perform qualitative analysis. The chromatogram peak areas were used to perform quan-

titative analysis in selected ion mode (SIM), the quantified ions included the following: 128 amu (Nap), 152 amu (Aci), 153 amu (Ace), 166 amu (Flu), 178 amu (Phe, Ant), 202 amu (Fluo, Pyr), 228 amu (BaA, Chr), 252 amu (BbFlu, BkFlu, BeP, BaP), 276 amu (InP, BghiP), 278 amu (DiBahA), and 300 amu (Cor). The data from the GC/MS analysis were processed using a ChemStation data analysis system.

## 1.3 Statistical analysis of data

### 1.3.1 Calculating BaP equivalent concentrations

BaP is the most intensively studied PAH species in animal tests and is the only PAH that has been tested for carcinogenicity in inhalation (WHO, 2000). The toxic equivalency factors (TEFs) were defined to rank the relative carcinogenic potential of other PAHs compared to BaP. The TEF for BaP was 1 (Nisbet and LaGoy, 1992). The TEFs can quantitatively represent the health hazards of PAHs. The relative toxicities of other PAHs can be computed and normalized by an index list based on the toxicity of BaP, as described in Eq.(1).

$$\text{TEC}_{\text{BaP}} = \text{Con}_{\text{PAHs}} \times \text{TEF}_{\text{BaP}} \quad (1)$$

where,  $\text{TEC}_{\text{BaP}}$  ( $\text{ng}/\text{m}^3$ ) is the normalized equivalent concentration for toxicity of BaP over 24 hours (Zhang et al., 2009);  $\text{Con}_{\text{PAHs}}$  ( $\text{ng}/\text{m}^3$ ) is the concentration of each PAH compound over the period of 24 hours;  $\text{TEF}_{\text{BaP}}$  is the toxicity equivalent factor based on BaP. After the calculation of  $\text{TEC}_{\text{BaP}}$ , the total toxicity concentration was obtained by adding all of the  $\text{TEC}_{\text{BaP}}$  together.

### 1.3.2 Monte Carlo simulation of the risk based on an incremental lifetime cancer risk model

An incremental lifetime cancer risk (ILCR) model was applied in this study to assess the lifetime risk of the exposure of the TAs to PAHs. The ILCR model can be used in estimating the incremental probability of an individual developing a tumor over a lifetime and described as Eq. (2):

$$\text{ILCR} = \frac{\text{CSF} \times \text{TTEC}_{\text{BaP}} \times \text{IR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (2)$$

where, ILCR is the incremental lifetime cancer risk from the occupational exposure to PAHs; CSF ((kg-day)/mg) is the inhalation cancer slope factor of BaP;  $\text{TTEC}_{\text{BaP}}$  ( $\text{ng}/\text{m}^3$ ) is the total equivalent concentration of 14 PAHs compared to the toxicity of BaP over 24 hours; IR ( $\text{m}^3/\text{hr}$ ) is the inhalation rate; EF (250 days/yr) is the exposure frequency, adapted from the Human Health Evaluation Manual (1991) (USEPA, 1991); ED (25 years) is the exposure duration, adapted from Human Health Evaluation Manual (1991) (USEPA, 1991); BW is body weight (log-normal (59.78, 1.07)), adapted from Department of Health,

ROC (Chen and Liao, 2006); AT is the average life time (70 years for carcinogens).

The Crystal Ball 7.2 was used as a tool in the Monte Carlo random simulation in risk assessment.

### 1.3.3 Source identification

Principal component analysis (PCA) was performed using PASW Statistics 18.0 software.

### 1.4 QA and QC

No significant contamination of solvents, apparatus and background level was found. A filter spiked with 2 mL of the standard solution was extracted as a substitute for an actual sample to check the recovery. The PAH concentrations were corrected for the recovery efficiency of extraction. The mean field recovery was Nap (58.99%), Aci (39.92%), Ace (50.73%), Flu (58.70%), Phe (97.42%), Ant (71.93%), Fluo (82.39%), Pyr (76.15%), BaA (76.26%), Chr (74.76%), BbFlu (89.16%), BkFlu (72.18%), BeP (72.45%), BaP (75.47%), InP (111.21%), DiBahA (76.82%), BghiP (73.72%) and Cor (78.11%), and the limitation of machine detection (LMD) ranged from 0.01  $\mu\text{g}/\text{mL}$  (Aci) to 0.15  $\mu\text{g}/\text{mL}$  (Nap).

All of the questionnaires were collected by a professional staff and input using Epidata 3.0 software.

## 2 Results and discussion

### 2.1 Exposure level of PAHs

Nap, Aci, Ace and Flu had a poor recovery (less than 60%) or were not detected in most of the samples, so only 14 PAHs (Phe, Ant, Fluo, Pyr, BaA, Chr, BbFlu, BkFlu, BeP, BaP, InP, DiBahA, BghiP and Cor) were discussed in this study.

The PAH exposure levels for the TAs are shown in Fig. 2. The total exposure concentrations of 14 PAHs are approximately 2871  $\text{ng}/\text{m}^3$  (on-duty) and 1622  $\text{ng}/\text{m}^3$  (all-day). The total indoor PAH level is 1257  $\text{ng}/\text{m}^3$ . Among the 14 PAHs, Fluo (41.67  $\text{ng}/\text{m}^3$ ) is the least abundant compound, and Cor (449.72  $\text{ng}/\text{m}^3$ ) is the most abundant compound in the on-duty samples. Fluo (30.60  $\text{ng}/\text{m}^3$ ) is the least abundant compound, and InP (233.78  $\text{ng}/\text{m}^3$ ) is the most abundant compound in all-day samples. Fluo (11.76  $\text{ng}/\text{m}^3$ ) is the least abundant compound, and InP (211.66  $\text{ng}/\text{m}^3$ ) is the most abundant compound in indoor samples. Except for BaP, the concentrations of other compounds in exposure samples show the following sequence: on-duty > all-day > indoor. Traffic emission makes a huge contribution to air quality on the job site. Due to the diversity of traffic flow in different places, the PAH exposure level for TAs on duty varied from 90.91 to 886.36  $\text{ng}/\text{m}^3$ . There is no significant difference between the daily average exposure level and the indoor level. Most of the Cor comes from automobile exhaust emissions, so exposure on-duty shows the highest level (449.72  $\text{ng}/\text{m}^3$ ), more than the all-day exposure (222.00  $\text{ng}/\text{m}^3$ ). The indoor Cor (197.58  $\text{ng}/\text{m}^3$ ) shows the lowest concentration.

The concentration of BaP inhaled by TAs in Tianjin was 310.61  $\text{ng}/\text{m}^3$  (on-duty), 142.52  $\text{ng}/\text{m}^3$  (all-day) and 117.83  $\text{ng}/\text{m}^3$  (indoor). The daily average concentration limit promulgated by the State Environmental Protection

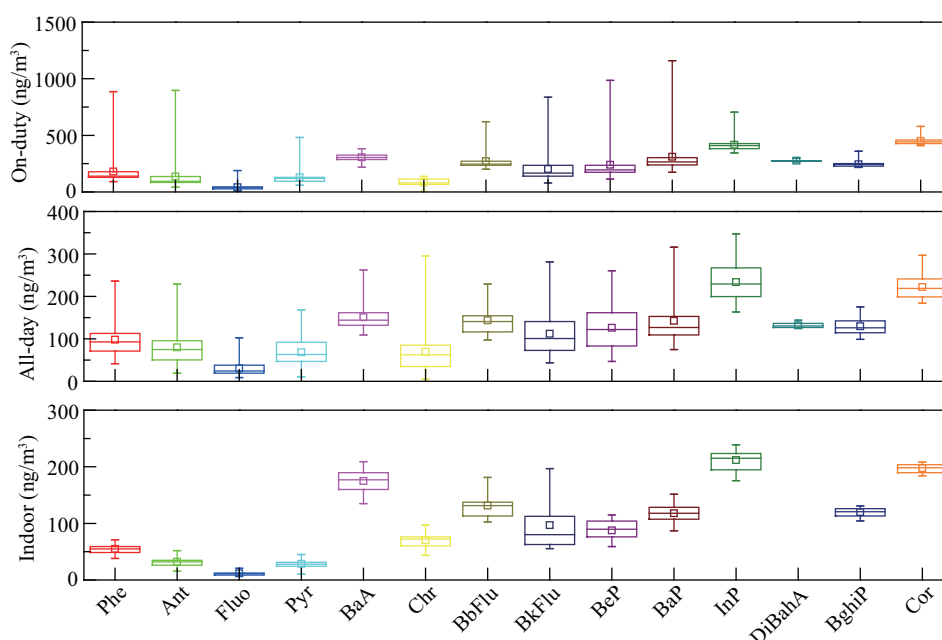


Fig. 2 PAHs exposure level of TAs in Tianjin.

Department of China (GB3095-1996) is less than  $10 \text{ ng/m}^3$  and the criterion of WHO is  $1 \text{ ng/m}^3$  (WHO, 1987). The exposure levels for TAs are far higher than those 2 standards and in a dangerous situation, and an intervention to control exposure and reduce risk is necessary.

Comparing the results of this study, the PAHs exposure level for the TAs in Tianjin was lower than the PAH exposure of highway toll station workers in Taiwan and PAH exposure levels of traffic police in Tianjin according to the research of Hu et al. (2007). The PAH exposure level for the TAs in Tianjin is higher than the PAH exposure level of traffic policemen in Budapest (Szaniszló and Ungváry, 2001) and in Beijing (Liu et al., 2007a, 2007b; Zhang et al., 2006). The highway toll station staff has a long exposure time and more direct exposure, and the Beijing traffic police during the summer study period encounter volatile gaseous compounds caused mainly by the traffic.

## 2.2 Characterization of the personal exposure to PAHs

Comparing the daily exposure profile and exposure profile on duty, most of the PAHs show the same level between on duty and all day, but compounds with 3–4 rings show a low abundance, which can be explained by the occupational contribution playing an important role in the total PAH exposure of the TAs.

Comparing the smoking status, there were obvious differences between males and females (Table 1). Males showed a higher PAH exposure level than females from the smoking status because most of the smokers were male. Environmental tobacco smoke (ETS) and cooking fumes (CF) are 2 more sources of PAH exposure. The source profiles of these 2 sources have been published in another paper by this author (Zhang et al., 2009). Smoking status and more time spent in cooking would raise the PAH exposure concentration up to a higher level. Some source characteristics can be determined from the relative exposure concentration of every PAH species. The exposure concentration and profile for females is similar to nonsmoker exposure concentration and profile of the males is similar to the exposure concentration and profile of a smoker. The exposure concentration increases with the times spent in cooking. After comparing the differences between the PAH exposure levels according to smoking status and gender, we find that most of the smokers are male, and the cooking fumes make an important contribution to the exposure increase from male to female. The concentrations of BaA, Chr, BbFlu, DiBahA and Cor show less change.

The diagnostic ratio of PAH isomeric compounds can be used for qualitative investigation of the source contribution. Five specific ratios were applied in this study to identify the source of PAHs: InP/(InP+BghiP), BaP/BghiP, Flu/(Flu+Pyr), Ant/(Ant+Phe) and BaA/(BaA+Chr). The diagnostic ratios in some other publications as well as in this study are shown in Table 2.

In this study, most of the InP/(InP+BghiP) ratios were approximately 0.63–0.64. This ratio in smokers was higher than the ratio in nonsmokers. The sample sites in this study were in urban Tianjin that had a number of gasoline engines on the roads, so the ratio in the on duty samples was lower than the ratio in the all day and indoor samples because of the contribution of gasoline engines. Emission from gasoline engines has a low ratio.

Most of the ratios of BaP/BghiP varied from 0.95 to 1.27. This ratio in smokers was higher than in nonsmokers, and males showed a higher ratio than females because of the ETS (BaP/BghiP in ETS is approximately 1.89) driving the ratio up. This ratio increased with the daily cooking frequency, because of the high value of CF. The ratio for on-duty had a larger value than the ratio for all day and indoor samples. Others sources can be found, but gasoline and diesel engine emissions were the major contributors to the exposure of TAs to PAHs just like ETS, etc.

For the ratio of Flu/(Flu+Pyr), all day samples and indoor samples were 0.30 higher than on-duty samples (0.24), and females and nonsmokers were 0.29 lower than males and smokers (0.31). This ratio increased with an increase in cooking frequency. Comparing the ratios for different sources with the literature, gasoline engines were found to be less than 0.5, and CF and ETS were found to be more than 0.5. The contribution of the gasoline engine decreased the ratio, but the contributions of CF and ETS increased the ratio. The frequency of cooking drives the ratio up to a higher level. Most of the smokers were male. Therefore, the ETS source produced the same contribution for males and smokers (an increase in this ratio).

For the ratio Ant/(Ant+Phe), the sequence of indoor < on-duty < all-day was shown (Table 2). Smokers (0.46) showed a higher ratio than nonsmokers (0.44), and females (0.43) showed a lower ratio than males (0.46). Cooking frequency had a positive effect on the increase in this ratio. The literature shows that gasoline engine combustion has the highest ratio, and natural gas combustion has the lowest value among the 8 sources. In this study, this ratio in indoor samples was lower than on-duty samples because of the contribution of the gasoline engine emission. Males and smokers showed a higher ratio than females and nonsmokers because of the contribution of ETS. Cooking times had a bidirectional effect on this ratio because the combustion of natural gas in kitchens will drive the ratio down, but the CF source emissions will drive the ratio higher.

For BaA/(BaA+Chr), the ratios in this study were higher than the sources reported by others with on-duty > indoor > all-day. Males and smokers were lower than females and nonsmokers. Cooking frequency had a negative effect.

## 2.3 Risk assessment due to exposure to PAHs

Assessment of the health risk PAHs pose to TAs can cite the cancer incidence and then promote occupational



**Table 1** Difference of exposure level (ng/m<sup>3</sup>) based on the gender, smoke status and cooking frequency (*n* = 33)

PAHs	Gender		Smoke		Cooking frequency		
	Female ( <i>n</i> =10)	Male ( <i>n</i> =23)	Smoker ( <i>n</i> =20)	Nonsmoker ( <i>n</i> =13)	1 times per day ( <i>n</i> =4)	2 times per day ( <i>n</i> =18)	3 times per day ( <i>n</i> =11)
Phe	82.47 (49.8) <sup>a</sup>	104.77 (30.93)	106.86 (32.67)	84.4 (45.14)	85.07 (37.84)	98.38 (22.72)	98.01 (84.99)
Ant	63.02 (53.88)	87.49 (33.49)	89.67 (35.37)	65.31 (48.89)	66.41 (40.57)	80.25 (25.1)	80.07 (91.84)
Fluo	22.74 (28.05)	32.68 (13.37)	33.68 (13.97)	23.5 (25.65)	21.27 (11.71)	27.2 (15.95)	29.67 (49.39)
Pyr	55.73 (37.41)	74.05 (25.69)	76.04 (26.59)	56.89 (36)	57.29 (21.8)	66.07 (25.02)	68.5 (67.57)
BaA	154.51 (47.4)	149.07 (19.09)	151.13 (19.72)	150.24 (43.08)	142.8 (20.67)	147.28 (24.34)	150.77 (67.94)
Chr	62.15 (86.75)	72.84 (32.33)	77.39 (30.17)	57.96 (79.05)	61.63 (28.9)	60.67 (34.67)	69.5 (133.1)
BbFlu	146.36 (42.1)	142.51 (31.36)	145.05 (32.11)	141.56 (38.91)	155.38 (21.01)	139.18 (31.27)	143.68 (71.95)
BkFlu	89.93 (47.51)	121.38 (55.83)	125.52 (57.26)	90.81 (46.37)	148.01 (44.19)	101.85 (34.31)	111.85 (120.67)
BeP	111.98 (51.04)	131.94 (50.47)	137.85 (52.44)	107.51 (51.19)	116.76 (57.43)	125.77 (46)	125.89 (74.56)
BaP	120.66 (54.93)	152.02 (51.31)	155.99 (54.11)	121.8 (53.97)	126.3 (69.69)	139.28 (45.73)	142.52 (51.37)
InP	232.81 (56.86)	234.22 (43.35)	239.31 (44.5)	225.69 (52.98)	233.51 (35.28)	226.37 (49.64)	233.78 (58.83)
DiBahA	132.82 (7.92)	131.17 (4.84)	131.73 (4.84)	130.79 (7.92)	126.74 (2.65)	129.17 (6.98)	131.47 <sup>b</sup>
BghiP	129.86 (23.44)	129.23 (17.92)	130.82 (18.33)	127.27 (21.75)	132.38 (16.18)	127.51 (19.58)	129.42 (25.17)
Cor	219.18 (36.03)	222.98 (24.11)	225.52 (24.28)	215.59 (33.66)	219.62 (20.22)	217.99 (30.98)	222 (25.04)
Σ14 PAHs	1624.23 (579.68)	1786.35 (388.63)	1826.57 (402.58)	1599.33 (542.87)	1693.15 (389.61)	1686.96 (366.76)	1737.13 (960.96)

<sup>a</sup> Mean and standard deviation (in parentheses).

<sup>b</sup> Only one sample was detected in this PAHs species.

**Table 2** Diagnostic ratios based on the gender, smoking status and cooking frequency

Diagnostic ratio	InP/(InP+BghiP)	BaP/BghiP	Flu/(Flu+Pyr)	Ant/(Ant+Phe)	BaA/(BaA+Chr)
On-duty ( <i>n</i> =25)	0.63	1.27	0.24	0.43	0.79
All-day ( <i>n</i> =33)	0.64	1.10	0.30	0.45	0.68
Indoor ( <i>n</i> =10)	0.64	0.99	0.30	0.37	0.71
Female ( <i>n</i> =10)	0.64	0.93	0.29	0.43	0.71
Male ( <i>n</i> =23)	0.64	1.18	0.31	0.46	0.67
Smoker ( <i>n</i> =20)	0.65	1.19	0.31	0.46	0.66
Nonsmoker ( <i>n</i> =13)	0.64	0.96	0.29	0.44	0.72
Cooking 1 times per day ( <i>n</i> =4)	0.64	0.95	0.27	0.44	0.70
Cooking 2 times per day ( <i>n</i> =18)	0.64	1.09	0.29	0.45	0.71
Cooking 3 times per day ( <i>n</i> =11)	0.64	1.10	0.30	0.45	0.68
Gasoline engine	0.18 <sup>a</sup>	0.5–0.6 <sup>b</sup>	< 0.5 <sup>c</sup>	0.50 <sup>d</sup>	0.49 <sup>e</sup>
Diesel engine	0.35–0.7 <sup>f</sup>	0.3–0.4 <sup>g</sup>	> 0.5 <sup>c</sup>	0.35 <sup>d</sup>	0.68 <sup>e</sup>
Coal/coke	0.33 <sup>h</sup>	> 1.25 <sup>b</sup>	0.53 <sup>i</sup>	–	0.50 <sup>h</sup>
Coal burning	0.56 <sup>f</sup>	0.9–6.6 <sup>j</sup>	0.57 <sup>k</sup>	0.24 <sup>d</sup>	0.46 <sup>k</sup>
Wood combustion	0.62 <sup>l</sup>	–	0.51 <sup>k</sup>	0.16 <sup>k</sup>	0.43 <sup>m</sup>
Natural gas combustion	0.32 <sup>k</sup>	–	0.49 <sup>k</sup>	0.12 <sup>k</sup>	0.39 <sup>k</sup>
CF (cooking fume) <sup>n</sup>	0.40	1.33	0.69	0.48	0.43
ETS (environmental tobacco smoke) <sup>n</sup>	0.52	1.89	0.77	0.39	0.30

<sup>a</sup>Wang et al., 2008; Del.Rosario Sienra et al., 2005; <sup>b</sup>Ravindra et al., 2008; <sup>c</sup>Ravindra et al., 2008; Ravindra et al., 2006; <sup>d</sup>Guo et al., 2003; <sup>e</sup>Khalili et al., 1995; Vasilakos et al., 2007; <sup>f</sup>Wang et al., 2008; Del.Rosario Sienra et al., 2005; Ravindra et al., 2008; Ravindra et al., 2006; Vasilakos et al., 2007; Saarnio et al., 2008; <sup>g</sup>Bourotte et al., 2005; <sup>h</sup>Tang et al., 2005; Saarnio et al., 2008; <sup>i</sup>Akyüz and Çabuk, 2008; <sup>k</sup>Galarnau, 2008; <sup>l</sup>Del Rosario Sienra et al., 2005; Ravindra et al., 2008; Akyüz and Çabuk, 2008; Mantis et al., 2005; <sup>m</sup>Akyüz and Çabuk, 2008; Mantis et al., 2005; <sup>n</sup>Zhang et al., 2009.

–: no reported.

protection. A Monte Carlo simulation was performed to assess the risk of exposure of TAs to PAHs based on the ILCR. After running the simulation 1000, 5000 and 10,000 times, the risk and sensitivity were described in **Table 3**.

All of the constants were listed and evaluated according to the reference. IR (breath volume) obeys a triangular

distribution according to the report of USEPA (1997). The minimum, most probable and maximum values are 1.1, 1.3 and 1.5 m<sup>3</sup>/hr. Chen and Liao (2006) have summarized the potency of the BaP cancer slope factors (Collins et al., 1991; OEHHA, 1993; Thyssen et al., 1981), and they concluded that the CSF of BaP obeys a lognormal

**Table 3** Monte Carlo simulation \*

Parameter	Input	Trial number	Risk ( $\times 10^{-4}$ )	Sensitivity of risk
Breath volume	Triangular (1.3, 1.1–1.4)	1000	1.062 $\pm$ 0.80	CSF: 66.0%; TEC <sub>BaP</sub> : 32.2%; breath volume: 1.2%; body weight: –0.6%
Body weight	Lognormal (65, 5)	3000	1.034 $\pm$ 0.76	CSF: 63.1%; TEC <sub>BaP</sub> : 35.7%; breath volume: 1.1%; body weight: 0%
TEC <sub>BaP</sub>	Lognormal (263.22, 110.79)	5000	1.047 $\pm$ 0.75	CSF: 63.0%; TEC <sub>BaP</sub> : 36.1%; breath volume: 0.7%; body weight: –0.2%
CSF	Lognormal (3.14, 1.80)	8000	1.055 $\pm$ 0.79	CSF: 64.4%; TEC <sub>BaP</sub> : 34.6%; breath volume: 1.0%; body weight: 0%
EF	250	10000	1.053 $\pm$ 0.79	CSF: 63.8%; TEC <sub>BaP</sub> : 35.3%; breath volume: 0.8%; body weight: –0.1%
ED	25			
AT	70			

\* Output was calculated by  $\frac{(\text{CSF} \times \text{TEC} / 1000000) \times \text{breath volume} \times \text{EF} \times \text{ED}}{(\text{body weight} \times \text{AT} \times 365)}$

distribution (3.14, 1.80). After an exploratory study, we found that the TEC<sub>BaP</sub> obeys a lognormal distribution (263.22, 110.79). All of the parameters (Table 3) were applied in the Monte Carlo simulation.

After 1000, 3000, 5000, 8000 and 10000 iterations, the results showed that 8000 trials are sufficient to ensure the stability of the results. The cancer risk to the TAs was  $1.055 \times 10^{-4}$  (95% CI (confidence interval):  $2.81 \times 10^{-5}$  to  $2.52 \times 10^{-4}$ ) which is higher than in the primary studies by Hu et al. (2007). Hu et al. (2007) reported that the risk posed by PAHs for policeman in Tianjin was approximately  $2.1 \times 10^{-5}$  to  $6.7 \times 10^{-5}$ . For both the traffic policeman and the traffic assistants, the PAH inhalation exposure risk was remarkably larger than the negligible risk level (approximately  $10^{-6}$ ). This risk scale was 2–3 times the risk of sintering workers in Taiwan (Lin et al., 2008), less than the workers in a carbon black factory (Tsai et al., 2001), and approximately 10% that of workers in 2 foundries (Liu et al., 2010) in Taiwan. The risk was 2–3 fold higher than the risk of Beijing traffic police officers (approximately  $4.16 \times 10^{-4}$ – $5.83 \times 10^{-4}$ ) (Liu et al., 2007b).

The results of the sensitivity analysis showed that the CSF is the most important contributor (64.4%) to the uncertainty of this forecast, and the next contributors are the TEC<sub>BaP</sub> (34.6%), breath volume (1.0%) and body weight (0%). Most of the uncertainty of the risk assessment is due to these factors. In the future, a more detailed toxicology database can show less uncertainty in the evaluation. Use of a PAH concentration monitor is another way to strength the simulation.

## 2.4 Exposure source apportionment

Few studies focus on exposure source identification (Anderson et al., 2001; Brinkman et al., 2009; Miller et al., 2002; Yakovleva et al., 1999), and PMF and PCA are the main methods applied. The earlier studies focused on the personal exposure to VOCs and particulate matter.

In this study, 13 PAHs (except for the DiBahA, which had a low detected ratio in 33 samples) were rearranged by principal component analysis using PASW Statistics 18.

After a one-tailed *t*-Test, most of the individual PAHs show a significant correlation. The Kaiser-Meyer-Olkin statistics value is 0.75 (more than 0.7). The Bartlett's Test of Sphericity statistics value is 657 and the significance is 0.00 ( $p < 0.05$ ). All of the above information indicates that this database is fit for factor analysis. Based on the "Principal Component Analysis" method, 4 factors were extracted. A total 33 samples were rearranged according to the 4 factors shown in Table 4. The total explained variances of Factors 1 to 4 were 25.14%, 24.06%, 20.16% and 18.85%, respectively.

In Factor 1, 5-ring compounds (BkFlu, BeP and BaP) are the predominant compounds. This factor is associated

**Table 4** Principal component analysis of 33 all day PAHs exposure samples in Tianjin

Compound (rings)	Factor 1	Factor 2	Factor 3	Factor 4
Phe (3)	0.22	0.15	0.01	<b>0.94</b>
Ant (3)	0.22	0.15	0.02	<b>0.94</b>
Fluo (4)	0.37	0.43	0.36	<b>0.62</b>
Pyr (4)	0.43	0.41	0.45	<b>0.61</b>
BaA (4)	0.12	<b>0.77</b>	0.13	0.28
Chr (4)	0.23	<b>0.81</b>	0.16	0.40
BbFlu (5)	0.56	0.63	0.23	0.20
BkFlu (5)	<b>0.81</b>	0.38	0.00	0.17
BeP (5)	<b>0.73</b>	0.29	0.43	0.33
BaP (5)	<b>0.82</b>	–	0.30	0.36
InP (6)	0.13	<b>0.69</b>	<b>0.64</b>	–
BghiP (6)	0.22	<b>0.61</b>	<b>0.69</b>	–
Cor (7)	0.21	0.11	<b>0.89</b>	0.19
Variance (%)	25.14	24.06	20.16	18.85
Cumulative variance (%)	25.14	49.21	69.37	88.22
Eigenvalues	2.77	3.14	2.36	3.12
Potential sources	DE	GE and ETS	CC	CF and PV

DE: diesel exhaust, GE: gasoline exhaust, ETS: environmental tobacco smoke, CC: coal combustion, CF: cooking fumes, PV: petroleum volatilized from gasoline stations, –: no detected or no calculated.

The bold values are the components that have a factorial weight more than 0.6. Negative values are not been shown in this table.

with diesel exhaust emissions (Guo et al., 2003; Khalili et al., 1995). In factor 2, the high factor loading compounds were BaA (0.77) and Chr (0.81). All of the dominant compounds were 4-ring PAHs. From the literature, this factor is associated with gasoline exhaust emissions (Shah et al., 2005) and environmental tobacco smoke (Zhang et al., 2009). The heaviest weighted species (6–7-ring compounds, BghiP and Cor) represented Factor 3. The higher molecular weight PAHs originated mainly from pyrolytic processes such as coal combustion. In Factor 4, the dominant compounds were Phe (0.94) and Ant (0.94) which are low molecular weight compounds and come from unburned diesel oil and gasoline (Dachs et al., 2002). Khalili et al. (1995) have reported that the PAHs with small numbers of rings could be tested in diesel and gasoline samples. Some publications reported that PAHs from cooking fumes have low molecular weight species as the main constituents (Zhang et al., 2009).

The exposure of the TAs to PAHs by inhalation may come from many sources, not simply occupational exposure (automobile exhaust), public exposure (industrial emissions, such as coal combustion) and indoor emission sources (such as cooking activities and tobacco smoke). For the sake of controlling exposure and decreasing the risk, indoor emissions are as important as outdoor emissions.

### 2.5 Limitations

First, the results of the study have shown that there is a great variability in TAs' particulate-bound PAHs exposure. In addition, the limited sample size makes this study only a preliminary study of TAs' particulate-bound PAH exposure, and more robust results should be obtained on the basis of a wider range investigation.

Secondly, some deviation can come from the limited summer samples in the calculation of the lifetime cancer risk and individual exposure source apportionment. Based on the Winter/Summer value of particulate-bound PAH concentration varying from 2 to 10 (Hu et al., 2007a), the lifetime cancer risk may be underestimated. Also, the contribution of different sources was restricted in summer.

Thirdly, the gaseous parts of PAHs (gPAHs) were not included in this study. Although the TEFs of gPAHs (such as Nap) were smaller than those of large MW PAHs (5–6 rings), the actual impact to total lifetime cancer risk cannot be assessed.

Lastly, the diagnostic ratio method and PCA method are qualitative identification pPAH sources. More refined exposure source apportionment should be based on quantitative evaluation methods (such as, PMF and CMB).

## 3 Conclusions

In the present study, we find that the total exposure to 14

PAHs in PM<sub>10</sub> is approximately  $2871 \pm 928$  ng/m<sup>3</sup> (on-duty),  $1622 \pm 457$  ng/m<sup>3</sup> (all-day) and the indoor PAH level is  $1257 \pm 107$  ng/m<sup>3</sup>. The scarcest and most abundant species of 14 PAHs in on-duty, all-day and indoor samples are Fluo and Cor, Fluo and InP, Fluo and InP, respectively. The BaP concentration of TAs inhaled in Tianjin were 310.61 ng/m<sup>3</sup> (on-duty), 142.52 ng/m<sup>3</sup> (all-day) and 117.83 ng/m<sup>3</sup> (indoor), values that are higher than both the SEPA and WHO standards for air quality. The PAH exposure concentration is higher for smokers than nonsmokers, and the concentration is higher for males than females. The exposure levels for TAs are higher than SEPA air quality standards. Summarizing the ratio analysis, we can find that the exposure can come from various source emissions. After a Monte Carlo simulation, results show that the PAH exposure can create a cancer risk of approximately  $1.055 \times 10^{-4}$  (95% CI:  $2.81 \times 10^{-5}$  to  $2.52 \times 10^{-4}$ ), higher than the result of Hu et al. (2007) in Tianjin and approximately 2–3-fold that of the result of Liu et al. (2007c) in Beijing. The CSF (64%) and TEC<sub>BaP</sub> (35%) contribute the major uncertainty to this risk. After source rearrangement by PCA, 4 factors are derived from the 33 exposure samples. Factor 1 stands for the diesel exhaust, Factor 2 for gasoline exhaust, Factor 3 for coal combustion and Factor 4 for cooking fumes and petroleum volatilized from gasoline stations. Except for the mobile exhaust, coal combustion and cooking play important roles in total PAH exposure, suggesting a need for a total control of multi-source PAH exposure to control the exposure and protect the health of TAs.

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