

# Polycyclic aromatic hydrocarbon in urban soil from Beijing, China

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**Abstract:** Polycyclic aromatic hydrocarbons (EPA-PAHs) in the urban surface soils from Beijing were determined using gas chromatography and mass spectrometry (GC-MS). It is significantly complementary for understanding the PAHs pollution in soil of integrated Beijing city on the basis of the information known in the outskirts. The total concentration of 16 EPA-PAH was from 0.467 to 5.470  $\mu\text{g/g}$  and was described by the contour map. Compound profiles presented that the 4-, 5- and 6-ring PAHs were major compositions. The correlation analysis showed that PAHs have the similar source in the most sampling sites and BaP might be considered as the indicator of PAHs. Characteristic ratios of anthracene (An)/(An+phenanthrene (Phe)), fluoranthene (Flu)/(Flu+pyrene (Pyr)) and benzo[a]pyrene (BaP)/benzo[g,h,i]perylene (BghiP) indicated that the PAHs pollutants probably mainly originated from the coal combustion and it was not negligible from vehicular emission. The level of PAHs in our study area was compared with other studies.

**Keywords:** PAHs; urban soil; source; Beijing

## Introduction

Polycyclic aromatic hydrocarbons (PAHs) in the environment are attracting increasing attention for their widespread occurrence and their toxic, mutagenic, and carcinogenic potentials (Freitag *et al.*, 1985), sixteen un-substituted PAHs have been listed by the US Environmental Protection Agency (EPA) as priority pollutants. PAHs originated mainly from incomplete combustion of fossil fuels and other organic substances during industrial activities, residential heating, power generation, incineration, and vehicle emissions (Garban *et al.*, 2002; Dyke *et al.*, 2003; Mastral *et al.*, 2003). Due to the consumption of the large amount of energy sources, the urban environment is a source region of PAHs.

The rapid development of Beijing has resulted in significant stress to Beijing environments. Among them, severe air pollution was caused by the enormous daily traffic, industrial and domestic emissions. The total-PAH concentration in the aerosol of Beijing was 178.99  $\mu\text{g/g}$  in summer and 749.26  $\mu\text{g/g}$  in winter (Okuda *et al.*, 2002). The benzo[a]pyrene (BaP) concentration in outdoor air in residential areas of Beijing was approximately 15 times higher than that in residential areas of Tokyo (Ando *et al.*, 1996) and BaP from the urban center site (Temple of Heaven) in autumn even exceeded the Chinese National Standards about 9 folds (Li *et al.*, 2001). However, the data of PAHs about the air is not abundant, which is partly attributed to the difficulties and high costs associated with air sampling. It was reported that PAH concentrations in soil were associated significantly with the corresponding levels in air (Vogt *et al.*, 1987; Wild and Jones, 1995; Wild *et al.*, 1990), house dust

(Chuang *et al.*, 1995), urban street dust (Takada *et al.*, 1990) and plants (Wang and Meresz, 1982), therefore, PAH determination in soil may provide important information on the environmental pollution state (Trapido *et al.*, 1999), which will be helpful to the future development of study areas. A few studies have been published for soil in the typical areas in Beijing (e.g., highways, traffic line, residential and industrial areas) (Chu *et al.*, 2003; Ge *et al.*, 2004; Ma *et al.*, 2003; Tang *et al.*, 2005; Zheng *et al.*, 1997). The grid sampling and analysis of the surface soil from outskirts of Beijing had been done for PAHs pollutants (Ma *et al.*, 2005), but soil in the urban area with the many emission sources was excluded.

The aim of the study mainly concentrated on PAHs in the surface soil from the urban region of Beijing. The concentration, the spatial distribution, the compound profiles, and the possible sources of PAHs were examined and evaluated. The difference of PAHs between urban and outskirts was compared. Our study was significant for understanding the regional background of PAHs levels in the integrated Beijing, differing from types of anthropogenic input and evaluating the exposure level of sensitive populations living in Beijing.

## 1 Material and methods

### 1.1 Material and reagents

The mixed standard solution of 16 EPA-PAHs (naphthalene (Na); acenaphthylene (Acy); acenaphthene (Ace); fluorene (Fl); phenanthrene (Phe); anthracene (An); fluoranthene (Flu); pyrene (Pyr); benz[a]anthracene (BaA); chrysene (Chr); benzo[b]fluoranthene (BbF); benzo[k]fluoranthene (BkF); benzo[a]pyrene (BaP); indeno[1,2,3-cd]pyrene (InP);

dibenz[a,h] anthracene (DBA); benzo[g,h,i]perylene (BghiP)) each at 1000  $\mu\text{g/ml}$  were purchased from Aldrich (WIS, USA) and further diluted to obtain the desired concentration. The surrogate, 2-fluorobiphenyl (2-FBP) was purchased from Supelco (Bellefonte, PA, USA). All solvents were of analytical purity (Beijing Chemical Factory, China) and redistilled in all-glass system before use. Silica gel (100-200 mesh) was purchased from Qingdao Ocean Chemical Company (China) and was activated in drying oven at 130°C for 16 h. Anhydrous sodium sulfate (Beijing Chemical Factory, China) was heated at 600°C for 12 h.

### 1.2 Sampling and preparation

The main city zone of Beijing was surrounded by four ring roads by now. 30 soils at different locations within the four-ring road were sampled. The study area was divided into regular grids of  $(4 \times 4) \text{ km}^2$ , within which the topsoil samples (5–30 cm) were collected. Each of the composite soil samples was made of 16 sub-samples obtained in a  $(1 \times 1) \text{ km}^2$  grid using a stainless steel hand auger. The detailed sampling locations are shown in Fig.1. The soil was air-dried in fume hood at room temperature, thoroughly mixed, sieved to 30 mesh and stored in glass bottles at -4°C until further processing. The remaining water content in the soil was determined gravimetrically after drying individual sample in an oven at 105°C for 12 h. All the results were reported as dried weight.

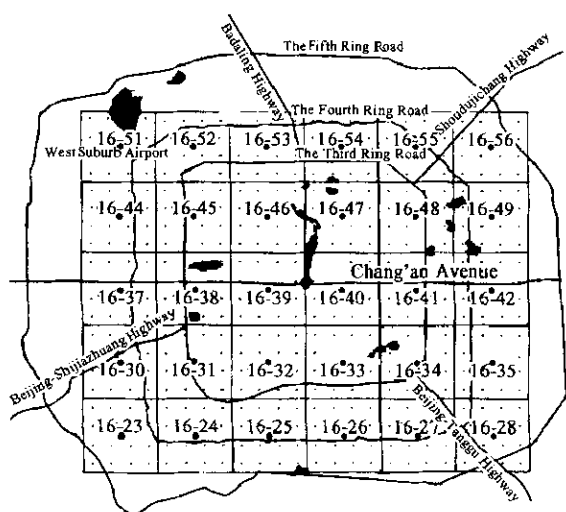


Fig.1 Geographic location of samples

### 1.3 Sample extraction and clean-up

Five gram was ground with anhydrous sodium sulfate into free flowing powder. The sample was ultrasonically extracted in centrifuge tube with 30 ml of acetone-hexane (1/1, v/v) for 5 min and then the extract was separated by centrifugation. The process was repeated three times. The solvents were combined and evaporated using K-D apparatus with a gentle

stream of nitrogen, then hexane was added as solvent. The concentrated extract was transferred to chromatograph column (30 cm  $\times$  10 mm I.D.) containing 10 g of activated silica gel with 1 g of anhydrous sodium sulfate on the column top. The silica gel was soaked in dichloromethane and filled in the column. The column was pre-eluted with 40 ml of hexane before loading the sample. The concentrated extract was transferred to the column top and was eluted by 25 ml of *n*-hexane and followed by 50 ml of hexane-dichloromethane (1/1, v/v). The second fraction was collected and evaporated, then was adjusted to 0.2 ml for determination.

### 1.4 PAHs determination

The determination of PAHs was carried out with a Hewlett Packard 6890 gas chromatography-5973 mass selective detector (GC-MS) system. The separation was performed on a fused silica capillary column (DB-5, 30 m  $\times$  0.25 mm I.D., and 0.25  $\mu\text{m}$  film thickness). The carrier gas was helium. The ionization was performed in the electron impact mode at 70 eV and the mass range scanned was from 50 to 550 amu under full scan acquisition mode. The GC oven temperature was programmed as follows: initial temperature 70°C held for 1 min, increased to 290°C at a rate of 4°C/min held for 2 min, then increased to 300°C at a rate of 10°C/min, maintained for 20 min. 1  $\mu\text{l}$  of sample was injected in splitless mode. Peak identification of PAHs was made by comparison of retention times and mass-spectrum with corresponding standards.

Quality control was based on the following procedures. Method blank was firstly run. The matrix blanks, the recoveries of PAHs and the limits of detection were based on the measures of the underground 20-metre-depth soil samples from the rural area of Beijing. The average recoveries for  $\Sigma 16$  EPA-PAHs in five times were from 86.7%–105.7% with less than 10% relative standard deviations. Before extraction, the soil sample was spiked with known amounts of 2-FBP as surrogate to compensate for the losses of PAHs. Recoveries for 2-FBP surrogate were 70%–115% (mean 89%) and the values were satisfactory and no correction was applied to the samples. The limits of detection (LODs) for PAHs as three times response of signal-to-noise in matrix blank sample, ranged from 1.00 for BaA to 8.64 ng/g for Na. All data have been blank corrected.

Correlation analysis was performed with SPSS 11.5 for Windows. The characteristic ratio of individual PAH was drawn using origin 6.1. For the contour map, Surface Mapping System (Ver. 7.0, Golden software, CO, USA) was used.

## 2 Results and discussion

### 2.1 PAHs concentrations in soil

The minimum, maximum, mean, median values and standard deviations of PAHs concentrations are listed in Table 1 and all were calculated by dried weight. The mean concentration of individual PAH varied from 0.007  $\mu\text{g/g}$  for Acy to 0.249  $\mu\text{g/g}$  for Flu. BaP, as one important representative toxic PAHs, was 0.023-0.402  $\mu\text{g/g}$  with a median value of 0.075  $\mu\text{g/g}$  and a mean value of 0.105  $\mu\text{g/g}$ .  $\Sigma$  2—3 ring PAHs referred to the total concentrations of Na, Acy, Ace, Fl, Ph and An and ranged from 0.088 to 1.288  $\mu\text{g/g}$  with 0.323  $\mu\text{g/g}$  as mean.  $\Sigma$  4—6 ring PAHs referred to the total concentrations of Flu, Pyr, BaA, Chr, BbF, BkF, BaP, InP, DBA and BghiP and varied from 0.325 to 4.683  $\mu\text{g/g}$  with 1.315  $\mu\text{g/g}$  as mean. The concentrations of  $\Sigma$  7 Car-PAHs was the sum of BaA, BaP, BbF, BkF, Chr, DBA and InP (US EPA, 2002) and was between 0.151 and 2.200  $\mu\text{g/g}$  with 0.677  $\mu\text{g/g}$  as the mean. The concentration of the  $\Sigma$  16 EPA-PAHs ranged from 0.467 to 5.470  $\mu\text{g/g}$  with 1.637  $\mu\text{g/g}$  as the mean concentration and 1.251  $\mu\text{g/g}$  as the median. The contour map of the  $\Sigma$  16 EPA-PAHs concentrations in the soil samples is shown in Fig.2. The map showed the spatial distribution characteristic of  $\Sigma$  16 EPA-PAHs. It seemed that the levels of  $\Sigma$  16 EPA-PAHs were more than 1.000  $\mu\text{g/g}$  in the most of the sites and those about the 13% sites exceeded the range of the majority of urban soil concentrations (0.600—3.000  $\mu\text{g/g}$ , WHO, 2000). As seen in Fig. 2, sites with the higher level of PAHs were not located in the urban center with dense vehicles and populations, but fell in the southwest and southeast regions beyond the Four-ring road and site with the highest level was found at station 16—33 (5.470  $\mu\text{g/g}$ ) located in the south of the Third-ring road. These may be accounted for by the fact that there were more PAHs inputs in these areas due to a large number of factories located, while types of industrial activities are the major factor of the consumption of energy source in Beijing. A large amount of waste gas/particle with PAHs from incomplete combustion of fossil fuel in industrial activities was emitted to atmosphere, then PAHs at atmosphere were transferred to soil via dry/wet deposition. Due to the correlation of PAHs between in soil and in air (Vogt *et al.*, 1987; Amagai *et al.*, 1999; Trapido *et al.*, 1999), it indicated that PAHs level in air in these regions should give prominence to be focused on.

The mean concentration of the individual PAH in urban soil from Beijing (0.007—0.249  $\mu\text{g/g}$ ) was greatly in excess of the background value in soil from the biosynthesis and forest fire (0.001—0.01  $\mu\text{g/g}$ ; Edwards, 1983) except for Acy, and much higher than those in the soil from the exurb of Beijing (Miyun and Fangshan districts) (0.002—0.009  $\mu\text{g/g}$ ; Ge *et al.*, 2004). Compared with the surface soil from the

Table 1 Statistics description of PAHs in the soil of Beijing ( $\mu\text{g/g}$ )

Compound	Min.	Max.	Mean	Median	SD
Na	0.024	0.474	0.076	0.062	0.078
Acy	nd	0.032	0.007	0.006	0.006
Ace	nd	0.086	0.012	0.008	0.015
Fl	0.006	0.065	0.017	0.014	0.012
Phe	0.047	0.542	0.184	0.156	0.114
An	0.006	0.109	0.026	0.020	0.023
Flu	0.069	0.993	0.249	0.193	0.190
Pyr	0.051	0.750	0.183	0.144	0.144
BaA	0.026	0.412	0.121	0.084	0.096
Chr	0.053	0.665	0.205	0.153	0.157
BbF	0.036	0.543	0.169	0.130	0.126
BkF	0.018	0.308	0.099	0.075	0.076
BaP	0.023	0.402	0.105	0.075	0.088
InP	0.012	0.181	0.057	0.044	0.039
DBA	nd	0.138	0.033	0.026	0.032
BghiP	0.010	0.330	0.093	0.069	0.074
$\Sigma$ 2—3 ring PAHs	0.088	1.288	0.323	0.261	0.226
$\Sigma$ 4—6 ring PAHs	0.325	4.683	1.315	0.990	0.996
$\Sigma$ 7 Car-PAHs	0.151	2.200	0.677	0.493	0.515
$\Sigma$ 16 EPA-PAHs	0.467	5.470	1.637	1.251	1.147

Notes: nd = not detected; SD = standard deviation

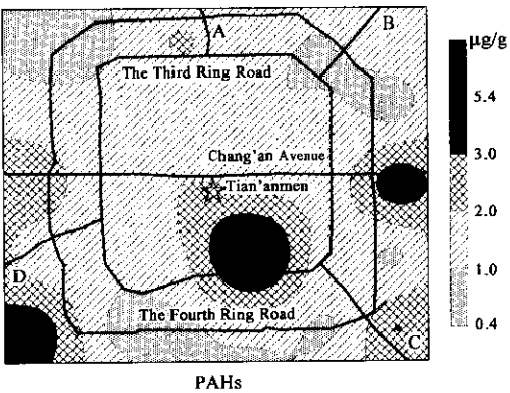


Fig.2 The contour Map of  $\Sigma$  16 EPA-PAHs in the urban soil of Beijing

outskirts of Beijing (0.017—0.132  $\mu\text{g/g}$ ; Ma *et al.*, 2005), the concentration with the high molecular weight mass was obviously higher. However, the change trend of PAHs in urban area was basically consistent with that in the outskirts area (Fig.3), possibly suggesting the similar sources of PAHs in two areas. BaP was identified in all the samples. The mean (0.105  $\mu\text{g/g}$ ) was nearly two-fold higher than that in the outskirts of Beijing (0.055  $\mu\text{g/g}$ ; Ma *et al.*, 2005) and even higher than that in the typical industrial and commercial region of Tokoyo in Japan (Matsushita *et al.*, 1980).

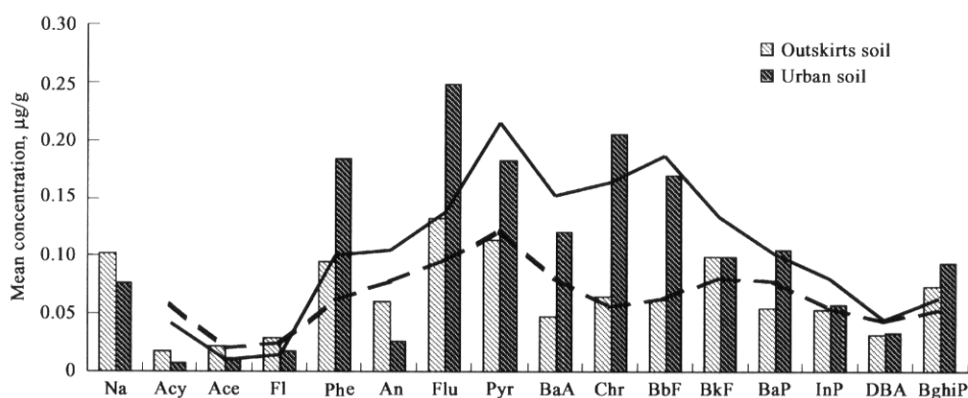


Fig.3 Composition of PAHs in soil from the outskirts and urban of Beijing

$\Sigma$  16 EPA-PAHs was ten-fold higher than the natural and background level of PAHs at remote or rural sites ( $0.100 \mu\text{g/g}$ ; Trapido *et al.*, 1999), suggesting that the urban soil of Beijing has been seriously contaminated by these pollutants. On the basis of the guideline of Dutch government,  $\Sigma$  16 EPA-PAHs was much higher than the target value set for unpolluted soil ( $0.02$ – $0.05 \mu\text{g/g dw}$ ; VROM, 1994) and that in site 16–33 even exceeded  $4 \mu\text{g/g}$  intervention value set for soil sanitation (Van Brummelen *et al.*, 1996). Compared with the reports published in other urban region, the result was significantly lower than those presented in urban soil from New Orleans ( $3.731 \mu\text{g/g}$  as median; Mielke *et al.*, 2001) and Estonian ( $2.200 \pm 1.396 \mu\text{g/g}$ ; Trapido *et al.*, 1999), was very close to the determined values from Tallinn, Helsingi, Vilnius, Chicago and London ( $1.092 \mu\text{g/g}$  as median; Saltiene *et al.*, 2002), but it was higher than those in the adjacent countries to China and the adjoining cities to Beijing, e.g. Tokushima in Japan ( $0.611 \mu\text{g/g}$  as mean; Yang *et al.*, 2002), tropical metropolis Bangkok ( $0.012$ – $0.380 \mu\text{g/g}$ ; Wilcke *et al.*, 1999), Chiang-Mai in Thailand ( $0.824 \mu\text{g/g}$  as mean; Amagai *et al.*, 1999) and Tianjin City of China ( $0.818 \mu\text{g/g}$  as mean; Wang *et al.*, 2003). It was reported that PAHs in soils from urban areas were approximately 2–10 higher than those in rural areas (Lodovici *et al.*, 1994; Tremolada *et al.*, 1996; Wagrowski and Hites, 1997). Here, the mean of  $\Sigma$  16 EPA-PAHs ( $1.637 \mu\text{g/g}$ ) was nearly 4 times higher than that in the rural area ( $0.464 \mu\text{g/g}$ ; Ma *et al.*, 2005) and was much higher than those from the exurb of Beijing ( $0.058$ – $0.141 \mu\text{g/g}$ ; Ge *et al.*, 2004). It appeared that the pollution situation of PAHs in Beijing urban soil was more serious than those in the adjacent areas.

## 2.2 PAHs compound profiles

The relative PAH composition in soil is displayed in Fig.4. Flu were the most abundant components (16%), followed by Chr (13%), Pyr (11%), Phe (11%) and BbF (10%). BaP formed 6% of  $\Sigma$  16 EPA-PAHs. PAHs by ring size were predominant by 4- and 5-ring

PAHs. In general, the  $\Sigma$  4–6 ring PAHs consisted of about 79.0% of  $\Sigma$  16 EPA-PAHs, while  $\Sigma$  2–3 ring PAHs only accounted for 21%. The seven potential carcinogenic PAHs represent 29%–51%. This might be related with the fact that PAHs with higher rings tend to be strongly adsorbed to the soil and lower molecular PAHs was apt to be depleted preferentially. The composition in this study area was similar to those in urban atmospheric particulates of Beijing (Huang *et al.*, 2001; Zeng *et al.*, 2001, 2002; Liu *et al.*, 2003), thus our study should be beneficial to understand the pollution of PAHs in the air. As reported, soil played a role of accumulator of PAHs in the air and PAHs concentrations in soil were associated significantly with the corresponding levels in air (Vogt *et al.*, 1987; Amagai *et al.*, 1999; Trapido *et al.*, 1999).

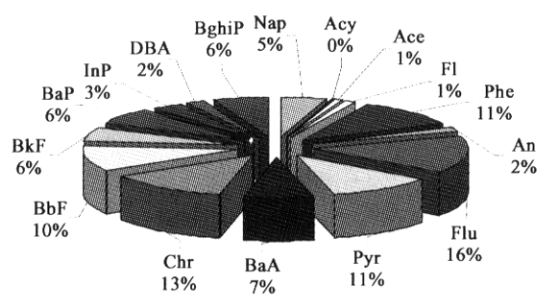


Fig.4 Relative PAH composition (% w/w) of 16 EPA-PAHs

A liner regression analysis was done among the individual PAH and  $\Sigma$  16 EPA-PAHs. Most of the individual PAH yielded a good correlation ( $r > 0.5$ ). There existed stronger correlations between lower molecular PAH (Na, Acy, Ace, Fl, Ph and An,  $r > 0.6$ ) and between the high molecular PAH (Flu, Pyr, BaA, Chr, BbF, BkF, BaP, Per, InP, DBA and BghiP,  $r > 0.8$ ), respectively. There was also a better correlation between individual PAH and  $\Sigma$  16 EPA-PAHs ( $r > 0.6$ ) except for Na and Ace. The better correlation might attribute to the similar source of PAHs at different sampling sites (Ma *et al.*, 2005; Weiss *et al.*, 1994; Wilcke and Zech, 1997). BaP, the

most potent carcinogenic PAHs, was significantly correlated with  $\Sigma 16$  EPA-PAHs ( $r=0.95$ ) and may be used to estimate total PAHs concentrations in the soil due to the similarity of each other.

2.3 Source of PAHs

Petrogenic and pyrolytic sources are by far the most important source of PAH inputs to the environment (Baek *et al.*, 1991). Petrogenic input is closely related to petroleum products (e.g., oil spills, road construction materials) and pyrolytic sources include combustion processes (e.g., fossil fuel combustion, forest fires, shrub and grass fires). Due to the lack of the characteristic parameters of PAHs in soil, those at atmosphere were generally used to discuss the PAHs source in soil (Trapido, 1999; Saltiene *et al.*, 2002; Zhang *et al.*, 2004; Ma *et al.*, 2005; Tang *et al.*, 2005). The ratios of low to high molecular weight PAHs (LPAHs/ HPAHs), anthracene to anthracene plus phenanthrene ( $An/(An+Phe)$ ) and fluoranthene to fluoranthene plus pyrene ( $Flu/(Flu+Pyr)$ ) was generally used as a tool for discriminating the petroleum/combustion sources of PAHs (Sicre *et al.*, 1987; Budzinski *et al.*, 1997; Zhang *et al.*, 2004). In general, the petroleum source contain relatively higher concentrations of 2—3 ring PAHs compounds (Tolosa *et al.*, 1996), while a large proportion of high molecular weight parent PAHs is typical characteristic of a combustion origin (Budzinski *et al.*, 1997).  $An/(An+Phe) < 0.10$  usually is considered as an indication of petroleum while a ratio of  $>0.10$  indicates a dominance of combustion (Budzinski *et al.*, 1997). Most crude oil samples have  $Flu/(Flu+Pyr)$  ratio below 0.40 and combustion generated PAHs are above 0.40. In our study, the HPAHs were definite dominant and the LPAHs/HPAHs ranged from 0.13—0.72 with the average ratio of 0.28,  $An/(An+Phe)$  was 0.05—0.18 with a mean of 0.12 and  $Flu/(Flu+Pyr)$  was 0.55—0.62 with a mean of 0.58, which suggested the definite predominance of combustion source in the urban soil of Beijing.

BaP/BghiP could be used to distinguish from traffic exhausts and coal combustion sources. The ratio of BaP/BghiP between 0.3—0.44 indicated the traffic exhausts and that between 0.9—6.6 showed the coal combustion (Sawicki, 1962). In addition, the ratio of  $Flu/(Flu+Pyr)$  between 0.40 and 0.50 was considered as being more characteristic of liquid fossil fuel (vehicle and crude oil) combustion whereas the ratio above 0.50 is characteristic of grass, wood or coal combustion (Yunker *et al.*, 2002). In the present study, the ratio of BaP/BghiP ranged from 0.3 to 5.1 with 1.2 as the mean and 90% values were in the range of 0.9—6.6. All values of  $Flu/(Flu+Pyr)$  were  $>0.5$ . Plot of BaP/BghiP versus  $Flu/(Flu+Pyr)$  was shown in Fig.5. As seen in Fig.5, 90% of samples fell

in the combustion zone (I), suggesting the prominence of coal combustion around the study area. Three samples with  $Flu/(Flu+Pyr) > 0.5$  and  $BaP/BghiP < 0.9$  indicated the mixture (II) of coal combustion and traffic emission.

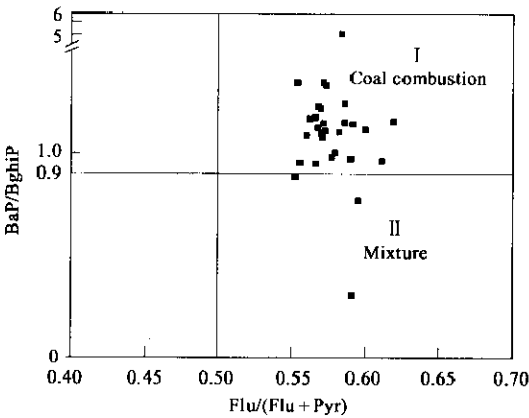


Fig.5 Plot of BaP/BghiP versus  $Flu/(Flu+Pyr)$

Through the analysis of PAHs characteristic ratio, it can be concluded that pyrolysis source, especially coal combustion, was the main contamination sources of PAHs in soil from the urban area of Beijing. However, due to the PAHs source in soil were discussed by the characteristic parameters at atmosphere, some error were probably made by the different environmental medium. This result was not in accordance with those in other metropolitan areas, which generally considered traffic as the main source of PAHs in environment (Rogge *et al.*, 1993; Van Metre *et al.*, 2000), but it was fit for the real condition in Beijing and could be accounted for by the energy consumption and the structure of energy sources in Beijing. Beijing's energy consumption pointed to a total of over  $41 \times 10^6$  t of standard coal in 2000, of which coal and hard coke took up 74.2 percent forming the main cause of pollution to Beijing's atmosphere and local environment (Zeng *et al.*, 2001, 2002; Liu *et al.*, 2003; Zhang *et al.*, 2004). However, He *et al.* (2001) reported that the vehicle in Beijing grows at a rate of 15% per year through the 1990s, and have exceeded  $2.0 \times 10^6$  in 2000. Traffic exhaust had been considered as one of the predominated sources of contamination to PAHs in Beijing (Zeng *et al.*, 2001, 2002; Liu *et al.*, 2003) and Okuda *et al.* (2002) even concluded that traffic exhaust was major pollution source in the atmosphere of Beijing. In our study, the mean of BaP/BghiP was 1.2 and was only slightly higher than the transition point of coal combustion/traffic exhaust (0.9). In addition, BghiP, as a known marker of traffic exhausts (Baek *et al.*, 1991), with an average of 6.2% of the  $\Sigma 16$  EPA-PAHs, also reflected the importance effect of the traffic source in urban soils of Beijing. This indicated that controlling the coal usage should be emphasized on but the effect of increasing vehicles should not be

ignored for cutting the pollution of PAHs in Beijing.

### 3 Conclusions

This work revealed the soil contamination information by PAHs residues in urban area of Beijing. It extends our understanding of the current PAHs contamination status in whole the Beijing area. The absolute concentrations of  $\Sigma 16$  EPA-PAHs and BaP showed that pollution of PAHs in urban soil of Beijing was serious but most fell in the range of the majority of urban soil concentrations. PAHs in the most sites had a similar source. PAHs characteristic ratios showed that the coal combustion was the dominant sources of PAHs and traffic exhausts was not negligible. This study suggested that the pollution of PAHs in urban area of Beijing should emphasize on controlling the coal combustion and traffic exhaust, especially the coal combustion.

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