Contamination of polycyclic aromatic hydrocarbons (PAHs) in urban soils in Beijing, China

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Abstract

Soil contamination with polycyclic aromatic hydrocarbons (PAHs) is an increasing problem in many countries, including China. An extensive and systematic survey has been undertaken to evaluate the contamination with PAHs of urban soils in Beijing, China. Soil samples were collected from campuses of universities, schools and kindergartens, public squares, fallow land and roadsides, and were analyzed for 16 PAHs by GC–MS. There was a high variability in the total PAHs (Σ PAHs) concentrations, ranging from less than 366 to 27,825 ng g⁻¹. The highest Σ PAHs concentrations were found at roadsides and industrial sites. Soil organic carbon (SOC) is one of the important factors that can influence the concentrations of PAHs in soils. It was found that concentrations of Σ PAHs were significantly correlated with that of soil organic carbon. To trace the sources of PAHs, the ratios of phenanthrene to anthracene and fluoranthene to pyrene were used to identify pyrogenic and petrogenic sources, respectively. In most cases, PAHs in soils in urban areas of Beijing were pyrogenic. These sources included motor vehicle exhausts, industrial activities and coal burning. These data can be further used to assess the health risk associated with soils polluted with PAHs.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), a group of stable chemicals, are ubiquitous organic contaminants in environments, such as sediments and soils (Hoffman et al., 1984; Jones, 1991; Wild and Jones, 1993; Baumard et al., 1998; Ribes et al., 2003). They have been listed as priority pollutants by both the US Environmental Protection Agency (EPA) and European Union (EU). While PAHs can occur naturally, mostly they are originated from anthropogenic processes, such as burning of fossil fuels and other organic substances (Simoneit, 1977; Wakeham et al., 1980a,b). PAHs containing two or more rings usually have high stability in the environments. Due to the high hydrophobicity and stable chemical structure, PAHs are not very soluble and can be adsorbed rapidly onto soil

particles, particularly on soil organic matter (Means et al., 1980). PAHs in soils can be dispersed by surface runoff and dust production; soils can therefore be considered as one of the pollution sources for PAHs contamination in air and sediments (Mai et al., 2003). Soil types and properties such as organic carbon play the most important role in the adsorption of PAHs in soils (McGroddy and Farrington, 1995; Jonker and Smedes, 2000).

It has been well established that PAHs have carcinogenic, mutagenic and teratogenic effects on animals (Hoffman and Wynder, 1971; Grimmer, 1983; Perera, 1997). This has led to intensive research into their chemical and biological properties in the environments, and the mechanisms by which these physiological effects are produced. For the assessment of environmental risks associated with soil contamination with PAHs, it is important to evaluate the spatial distribution and pathways (sources) of PAHs in soils. Molecular indices based on individual compound concen-

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trations were developed to assess the various origins of these compounds (Solco, 1986; Sicre et al., 1987).

As anthropogenic activities are the main sources of PAHs, the levels of PAHs in soils in urban areas are approximately a factor of 2–10 higher than those in rural areas (Lodovici et al., 1994; Wagrowski and Hites, 1997). PAHs contamination in soils close to highways has been found to be similar to those in urban areas (Wang and Meresz, 1982). Beijing, located in northern China, is a densely populated city with a heavy load of vehicular traffics and major industrial plants within the metropolitan area. However, up to now, little information is available on the PAHs contamination in urban soils of Beijing (Chu et al., 2003). The purpose of this study is therefore to identify the levels of PAHs in topsoils in some key areas of Beijing in order to establish an environmental evaluation, which will be served as essential reference information for future studies, such as remediation technologies, health risk assessment and urban development.

2. Materials and methods

2.1. Area description and sampling procedure

Soils used in this study were collected from eight main municipal zones in urban areas of Beijing (Fig. 1). These sampling areas could be categorized into different functional sections, such as campuses of universities, schools and kindergartens, public squares, fallow land and roadsides where people always play and rest on. At each site, six samples (at the depth of 0–5 cm) were collected randomly, and were bulked together to form one composite sample. All samples were air-dried at room temperature

after transported to the lab within 4 h. Air-dried soil samples were crushed to pass 2-mm mesh sieve and homogenized, then sealed in Kraft paper envelopes or in solvent-cleaned glass containers and stored at -20°C until analysis.

2.2. Chemical analysis

All solvents purchased from Beijing Chemical Factory were of analytical grade and redistilled in all glass system before use. The stock reference standard mixture of 16 PAHs (Supelco Co., Cat. No. 47930-U) includes naphthalene, acenaphthylene, 2-bromonaphthalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(a)pyrene, indeno(123-cd)pyrene, dibenz(ah)anthracene, and benzo(ghi)perylene at a concentration of 2000 mg L^{-1} , respectively. Silica gel (60–100 mesh) purchased from Qingdao Haiyang Chemical Co. (Shangdong, China) was activated at 130°C for 10 h, then kept in a desiccator.

Ten grams of the soil sample was weighed precisely and mixed with 2 g anhydrous sodium sulfate. The sample was Soxhlet-extracted for 24 h with 160 ml dichloromethane/acetone solvent (7:9 v/v). The solvents were removed using a rotary evaporator and the volume was reduced to ca. 1 ml under nitrogen. The extract was then cleaned using silica adsorption chromatography capped with anhydrous sodium sulfate and eluted with 60 ml mixture of hexane and pentane (6:4 v/v). Afterward, this extract was reduced to 1 ml under nitrogen for analysis of PAHs.

The final concentrated extracts from the environmental samples were analyzed using a Hewlett Packard 6890 gas chromatography equipped with a splitless injector, HP-5 capillary column (60 m $\times$ 0.25 mm i.d. with 0.25 μm film thickness) and a 5973 mass spectrometer detector. The carrier gas was helium (ultra high purity) at a flow rate of 1.1 ml/min, while oven was programmed from 90°C (held for 2 min) initially to 200°C (held for 5 min) at a rate of 10°C/min and then to 300°C (held for 15

Fig. 1. Schematic map showing the soil sampling sites in the urban areas of Beijing City.

min) at a rate of 3 °C/min. The injector temperature was held at 290 °C, and the detector temperature was 310 °C. For identification, selected ion monitoring (SIM) mode was carried out using the molecular ions selective for individual PAHs. Chromatographic data were collected and processed using HP Chemstation software. A representative chromatogram of sample was shown in Fig. 2.

Experiments on recovery were carried out by spiking known concentration standards (2000 ng g⁻¹) in uncontaminated soil (previously found containing very low amounts of PAHs), and showed that the average recovery ranged from 81% for naphthalene to 110% for benzo(ghi)perylene. The concentration of those to produce a signal-to-noise of 3:1 in blank sample was defined as detection limit (LOD). The LOD for PAHs ranged from 2000 ng kg⁻¹ for naphthalene to 6000 ng kg⁻¹ for benzo(ghi)perylene.

All soils collected in this study were brown soils. Soil pH was measured to be 7.03 to 9.15 in 0.01 mol l⁻¹ CaCl₂ at a soil/solution ratio of 1:1. The content of soil organic carbon (SOC) was determined in triplicate using the methods described by Nelson and Sommers (1982). The standard deviation of measurements was ±3% for soil organic carbon (SOC).

3. Results and discussion

3.1. PAHs contamination levels in urban soils

A wide range of soil PAHs concentrations was observed (Table 1), from 219 ng g⁻¹ to 27,825 ng g⁻¹ (dry weight), which included those PAHs considered as carcinogens by the US Environmental Protection Agency. The mean concentration of total PAHs in soil samples was 3917 ng g⁻¹ of the sum of the 16 PAHs. The highest pollution levels were observed for soil samples in roadside showing heavy traffics at sites BJ-4 (14,574 ng g⁻¹) and BJ-5 (27,825 ng g⁻¹). These high pollution levels are comparable with those found in Beijing's highways (1470 to 6610 ng g⁻¹) (Chu et al., 2003), but were significantly lower than values reported on in the Handan city of China (1150 to 480 mg kg⁻¹) (Sun et al., 2003). PAHs concentrations near some petrol stations were also high. These stations are always jammed with motor vehicles and close to industrial activities, which explained the relatively high PAHs concentrations at these sites. PAHs in

soils collected in this study include compounds with a wide range of molecular weights, from naphthalene to benzo(ghi)perylene. Fig. 3 shows the variation in percentages of PAH compounds with different ring numbers in soils. The BJ-1 to BJ-7 sampling sites are located in the main traffic areas, and the major organic pollutants are mostly dominated by PAHs with 3–5 rings, such as pyrene. Sampling sites from CY-1 to CY-5 and from YS-11 to YS-18 had much less anthropogenic activities, which resulted in the dominance of PAHs with 2 rings. Generally speaking, the concentrations of PAHs with 6 rings were relatively low in most soil samples.

Some environmental standards (criteria or guidance) for PAHs in soil were released by the Netherlands Ministry of Housing, Spatial Planning, and the Environment (De Vries and Bakker, 1998), Canadian Council of Ministers of the Environment (CCME, 1996a,b), British Columbia Ministry of Environment, Lands and Parks (BCME, 1996), and The Ontario Ministry of Environment and Energy (MOEE, 1996), for assessing risk at contaminated sites, clean-up goals, and monitoring remediation efforts, as shown in Table 2. About 61% of collected soils in Beijing were associated with PAHs concentrations over 1 µg g⁻¹, which was set to be the soil target value for total 10 PAHs concentration by the Netherlands, indicating their potential risk to human health. However, such environmental standards for PAHs in soil are not established yet in many countries, including China.

3.2. Source of the PAHs contamination

Combustion processes and release of un-combusted petroleum products are the two main sources of anthropogenic PAHs found in the environment. In general, parent PAHs are largely produced by incomplete combustion of fossil fuels and plant materials or natural diagenesis (Yunker et al., 1996). To distinguish the natural and anthropogenic PAHs inputs in soils, we used PAHs compositions (parent, alkyl and vascular plant PAHs) and their ratios as a way to discriminate PAHs sources (Yunker et al., 1999; Sanders et al., 2002). The usefulness of these indices relies on the fact that during low temperature processes such as catagenesis of organic matter leading to the formation of petroleum, the PAHs distribution is governed by thermodynamic properties. For high temperature processes, such as pyrolysis of organic matter, their distribution is

Fig. 2. A representative chromatogram of 16 PAHs from a soil sample. (1=naphthalene, 2=acenaphthylene, 3=2-bromonaphthalene, 4=acenaphthene, 5=fluorene, 6=phenanthrene, 7=anthracene, 8=fluoranthene, 9=pyrene, 10=benzo(a)anthracene, 11=chrysene, 12=benzo(b)fluoranthene, 13=benzo(a)pyrene, 14=indeno(123-cd)pyrene, 15=dibenz(ah)anthracene, 16=benzo(ghi)perylene).

Table 1
Total polycyclic aromatic hydrocarbons and selected PAH ratios in each sample

Sampling sites		Total PAHs (ng g ⁻¹)	Phe/Ant index	Flu/Pyr index	SOC (%)	Dominant Origin
CY-1	University	4018.4	10.5	1.2	2.5	Pyro ^a and Petro ^b
CY-2	University	1725.1	ND ^c	1.3	1.7	Pyro
CY-3	Bus station	8036.7	10.5	1.2	4.3	Pyro and Petro
CY-4	Residential area	3450.2	ND	1.3	2.3	Pyro
CY-5	Playground	219.7	1.0	ND	0.8	Pyro
RD-1	Public squares	575.4	7.9	1.3	1	Pyro
RD-2	Residential area	4269.4	18.6	1.4	2.9	Pyro and Petro
RD-3	Residential area	5284.0	8.0	1.5	3.8	Pyro
RD-4	Residential area	365.8	6.5	1.4	0.9	Pyro
RD-5	School	943.0	15.1	1.4	1.1	Pyro and Petro
RD-6	School	658.8	14.1	1.4	1.2	Pyro and Petro
RD-7	School	939.1	5.5	1.3	1.2	Pyro
RD-8	Roadsides	12,597.9	10.6	1.2	4.9	Pyro and Petro
RD-9	Bus station	2068.4	8.8	1.2	1.7	Pyro
RD-10	Fallow land	2534.4	11.4	1.2	2.6	Pyro and Petro
YS-11	Public squares	1137.8	1.6	0.3	1.1	Pyro and Petro
YS-12	Residential area	543.3	1.5	0.2	0.9	Pyro and Petro
YS-13	Playground	1053.2	14.6	1.3	1.3	Pyro and Petro
YS-14	Kindergarten	1350.4	13.2	1.3	1.3	Pyro and Petro
YS-15	Residential area	1005.7	15.6	1.5	1.2	Pyro and Petro
YS-16	Roadsides	1538.4	21.7	2.1	1.6	Pyro and Petro
YS-17	Public squares	1321.9	6.6	1.1	1.5	Pyro
YS-18	Schoolyard	1431.4	3.4	0.3	2	Pyro and Petro
YS-19	Roadsides	2373.0	2.8	0.2	2.6	Pyro and Petro
BJ-1	Roadsides	1601.1	10.8	1.3	1.5	Pyro and Petro
BJ-2	Roadsides	1590.2	8.6	1.3	1.4	Pyro
BJ-3	Roadsides	2894.9	6.6	1.2	2.4	Pyro
BJ-4	Roadsides	14,574.0	8.3	1.0	5	Pyro
BJ-5	Service station	27,825.0	ND	1.2	6.2	Pyro
BJ-6	Roadsides	10,844.9	10.4	1.1	4.7	Pyro and Petro
BJ-7	Roadsides	2661.8	9.7	1.3	2	Pyro

^a Pyrogenic source.

^b Petrogenic source.

^c Not detectable.

governed by kinetic characteristics. Therefore, PAHs distribution is dependent on temperature (Alberty and Reif, 1988). The ratios of phenanthrene/anthracene (Phe/Ant) within the 3-ring PAHs group

and fluoranthene/pyrene (Flu/Pyr) within the 4-ring PAHs group were used to form molecular indices (Colombo et al., 1989; Baumard et al., 1998). Phenanthrene is more thermochemically

Fig. 3. The distribution of PAHs with different ring numbers in different soil samples collected from urban areas of Beijing, China.

stable than anthracene, so at low temperature molar fraction of phenanthrene produced is much higher than that of anthracene. High temperature courses, such as the incomplete combustion of organic materials (coal burning, wood burning, vehicular exhaust emission), are characterized by low Phe/Ant ratio values. The discrimination of the origins of PAHs was also performed according to the value of Flu/Pyr.

Usually, Phe/Ant ratio < 10 and Flu/Pyr ratio > 1 indicate that PAHs come from pyrogenic source and Phe/Ant > 15 and Flu/Pyr < 1 indicate petrogenic origins of PAHs (Baumard et al., 1998). The ratios of Phe/Ant and Flu/Pyr are listed in Table 1. The Flu/Pyr ratios in all samples ranged from 0.2 to 2.1 and Phe/Ant ratios ranged from 1.0 to 18.6. Data from each sample were grouped according to the origins of these PAHs in soils. It can be seen that there was a strong pyrogenic influence on soil PAHs in all soils sampled in Beijing. The molecular indices at site RD-8 were 10.6 and 1.2 for Phe/Ant and Flu/Pyr, respectively, indicated a possible influence of petrogenic inputs.

3.3. Effect of soil organic carbon (SOC)

Numerous studies have shown that PAHs are strongly retained by the soil matrix (Chung and Alexander, 1998, 2002). The partitioning concept of soil sorption of organic contaminants implies that the sorption of hydrophobic organic molecules is determined by the organic carbon content of the substrate (Karickhoff and Brown, 1979; Chiou et al., 1979, 1998). The organic matter content is considered to be a very important variable related to PAHs pollution of soils (Boehm et al., 2002). Soil organic carbon varied greatly between soil samples ranging from 0.8% to 6.2% with a mean of 2.4% (Table 1). The relationship between total concentrations of PAHs in soils and soil organic matter has been extensively investigated. In the present study, soils contained a wide range of SOC, with the lowest (0.8%) being found in soil CY-5, and

Fig. 4. The relationship between soil organic carbon and total concentrations of PAHs in soil samples at different locations in urban areas of Beijing.

the highest (6.2%) in soil BJ-5 (Table 1). A regression analysis was conducted to investigate the relationship between the concentrations of the Σ PAHs (16 compounds, ng g^{-1} d.w.) and the amounts of soil organic carbon (SOC, %). The experimental results showed a positive exponential relationship (Fig. 4). This result demonstrated that higher amounts of PAHs mainly occur in soils with higher SOC, which can be described by the following equation:

$$\Sigma\text{PAHs} = 610.85 \text{ SOC}^{1.8837}, n = 31, r^2 = 0.944$$

where Σ PAHs is the total concentrations of 16 PAHs in soils (ng g^{-1}), and SOC is the amounts of soil organic carbon (%). This relationship illustrates that the level of soil contamination with PAHs

Table 2

Some published Soil environmental standards for PAHs in soils ($\mu\text{g g}^{-1}$)

Compound name	RIVM TV	BC Ag	BC UP	BC R	BC C	BC I	CCME Ag	CCME RP	CCME C	CCME I	Ont Ag (C)	Ont RP (C)	Ont IC (C)
Acenaphthene											15	15	15
Acenaphthylene											100	100	100
Anthracene											28	28	28
Benzo(a)anthracene		0.1	1	1	10	10	0.1	1	10	10	6.6	6.6	6.6
Benzo(a)pyrene							0.1	0.7	0.7	0.7	1.2	1.2	1.9
Benzo(b)fluoranthene		0.1	1	1	10	10	0.1	1	10	10	12	12	12
Benzo(ghi)perylene											12	12	12
Chrysene											12	12	17
Dibenz(ah)anthracene		0.1	1	1	10	10	0.1	1	10	10	1.2	1.2	1.9
Fluoranthene											40	40	40
Fluorene											340	340	340
Indeno(123-cd)pyrene		0.1	1	1	10	10	0.1	1	10	10	12	12	19
Naphthalene		0.1	5	5	50	50	0.1	0.6	22	22	4.6	4.6	4.6
Phenanthrene		0.1	5	5	50	50	0.1	5	50	50	40	40	40
Pyrene		0.1	10	10	100	100	0.1	10	100	100	250	250	250
PAH (total of 10) ^a	1												

Abbreviations: BC Ag: British Columbia Agricultural; BC UP: British Columbia Urban Park; BC R: British Columbia Residential; BC C: British Columbia Commercial; BC I: British Columbia Industrial; CCME Ag: Canadian Council of Ministers of the Environment Agricultural; CCME RP: Canadian Council of Ministers of the Environment Residential/Parkland; CCME C: Canadian Council of Ministers of the Environment Commercial; CCME I: Canadian Council of Ministers of the Environment Industrial; Ont Ag (C): Ontario Agricultural land use (coarse- $\mu\text{g/g}$); Ont RP (C): Ontario Residential/Parkland land use (coarse- $\mu\text{g/g}$); Ont IC (C): Ontario Industrial/Commercial land use (coarse- $\mu\text{g/g}$); RIVM TV: The Netherlands soil/sediment (mg/kg dry material) target value.

^a The total of anthracene, benzo(a)anthracene, benzo(k)fluoranthene, benzo(a)pyrene, chrysene, phenanthrene, fluoranthene, indeno(123-cd)pyrene, naphthalene and benzo(ghi)perylene.

may be pre-determined by soil organic carbon contents, despite the fact that the input of PAHs to each site is rather different.

4. Conclusions

Data from the current study demonstrated that urban soils from Beijing, China, were contaminated with PAHs ranging from 366 to 27,825 ng g⁻¹. Based on the molecular indices (phenanthrene/anthracene and fluoranthene/pyrene ratios) of PAHs in soils, it was suggested that PAHs in soils from urban areas of Beijing had strong pyrogenic source. PAHs with 2–4 rings were the major fractions of the total soil PAHs. Total concentrations of PAHs in soils were strongly correlated with soil organic carbon (SOC), implying that SOC was the key factor determining the retention of PAHs in soils.

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