

Application of spatial analysis and multivariate analysis techniques in distribution and source study of polycyclic aromatic hydrocarbons in the topsoil of Beijing, China

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Abstract Surface soil samples were collected from 161 sites throughout the downtown and suburban area of Beijing, China. The samples were analyzed for polycyclic aromatic hydrocarbons (PAHs) concentrations. Through Kriging analysis, five heavily contaminated zones were identified in the study area. Sources of PAHs in the soil were apportioned using principal factor analysis and multiple linear regression. Three factors were identified representing coal combustion/vehicle emission, coking emission, and petroleum sources, respectively. The relative contributions of the three sources were 48% for coal/vehicle emission, 28% for coking emission, and 24% for petroleum sources. The contributions of total PAHs from the three sources were 16.4, 4.63 and 3.70 ng g⁻¹, respectively. Spatial analysis indicated that the contribution of coal/vehicle sources was higher in the downtown area than in the suburban area, the petroleum sources had a high contribution in the urban area, and the contribution of coking sources was high in the suburban area. The results indicated that PAH contamination in the surface soil in Beijing was closely related to the spatial characteristics of energy consumption and functional zoning. Improvement of the energy consumption structure and relocation of industries with heavy pollution are effective ways to control PAH contamination in surface soil in the area.

Keywords PAHs · Surface soil · Source apportionment · Beijing · China

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of toxic and carcinogenic pollutants, and are identified as high priority chemicals in “Regionally-based Assessment of Persistent Toxic Substances” in Region VII (including China) conducted by United Nations Environmental Program and Global Environmental Facility. The US Environmental Protection Agency (EPA) has promulgated 16 parent PAHs as priority pollutants. Due to the slow rates of photochemical decomposition and biodegradation, PAHs are considered persistent substances in the environment (Ma et al. 2005). Despite some natural sources, PAHs in the environment are mainly produced by anthropogenic sources including high-temperature reactions such as incomplete combustion and pyrolysis of fossil fuels and biomasses, and direct release from petroleum and related products (Garban et al. 2002).

Soil is an important sink for PAHs in the environment (Wild and Jones 1995). It was reported that the primary input of PAHs to topsoil is from air-to-surface precipitation (Wang et al. 2002), and the PAH concentrations in soil are closely related to the corresponding levels in air (Trapido 1999). Previous studies demonstrated the importance of soil PAHs contributing to root vegetables (Tao et al. 2004), and vegetables are one of the largest contributors to the human intake of PAHs (Dennis et al. 1983). In addition, evaporation and storm leaching are also sources of PAHs in atmosphere and groundwater concentrations (Cousins et al. 1999). Therefore, knowledge of PAH contamination in surface soil, especially in agricultural soil, is important to prevent direct and indirect exposure to PAHs.

According to the study conducted by Xu et al. (2006), biomass burning, domestic coal combustion, and the coking industry are the major sources of PAHs in

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China, and the emission profile featured a relatively higher portion of high molecular weight (HMW) species with carcinogenic potential due to the large contribution of PAHs from domestic coal and the coking industry. It is widely known that each PAH emission source has a specific source fingerprint. Researchers surmized that the relative abundance and predominance of certain PAH species might be used to distinguish emissions from particular pollution sources (Park and Kim 2005). Both source- and receptor-oriented approaches could be used to evaluate source contributions. The receptor-oriented approach usually infers the contribution from various sources by determining the best-fit to a linear combination of equations for the emission sources needed to reconstruct the measured composition of a sample or by using multivariate analysis (Hopke 1985).

Many studies have reported on the PAHs in the soils of some countries (Jones et al. 1989; Wilcke et al. 1999), however, studies on accumulation and distribution of PAHs in the soils of China are rare (Wang et al. 2003; Zuo et al. 2007; Cai et al. 2007, 2008). Beijing, as the capital city of China, is facing many environmental problems due to the rapid industrialization and urbanization of the past 30 years. The PAH contamination in the topsoil of this area is of particular concern because the agricultural soils in suburban areas are commonly used for vegetable production. The objective of the present study is to understand the distribution and identify the major sources of PAHs in surface soil by spatial analysis techniques, principal components analysis (PCA) and multivariate linear regression based on the measured PAH concentrations in 161 surface soil samples collected from both urban and suburban areas of Beijing. The contribution of each source was quantified, and compared with the source emissions estimated on fuel consumption and emission factors. Based on the calculated source contributions at each sampling site, the Kriging interpolation method was applied to investigate the spatial distribution of each source.

Materials and methods

Samples collection

A total of 161 topsoil samples (0–10 cm) were collected in the whole Beijing area from April to June 2004. At each sampling site, fresh soil samples were collected from five locations in an area of about $100 \times 100 \text{ m}^2$ (at the four corners and the center), and thoroughly mixed to form a composite soil sample. Soil samples were air dried at room temperature and sufficiently ground in a grinder (Fritsch Pulveriste 2) to pass through a 1 mm-mesh sieve. Fine roots were removed from the sieved samples using a glass

bar rubbed on silk. The samples were then stored at -18°C prior to analysis.

Sample extraction and cleanup

The extraction and analysis were carried out according to a procedure developed by Tao et al. (2002). Ten grams of soil samples mixed with 10 g of baked anhydrous sodium sulfate were extracted using accelerated solvent extraction (Dionex ASE 300). Extractions were performed with 34 ml of solvent (1:1 mixture of hexane and dichloromethane).

The extracts were concentrated with a rotary evaporator, solvent-exchanged to hexane, and then passed through silica column. A glass column (10 mm i.d. \times 350 mm length) was first filled with dichloromethane, then packed with 10 g of silica gel (presoaked in dichloromethane) and about 25 mm length of anhydrous sodium sulfate. The extra dichloromethane was released while keeping the silica gel and the anhydrous sodium sulfate soaked. The extracts from soil samples were transferred to the silica columns and eluted with 25 ml of hexane and then 50 ml of hexane and dichloromethane (3:2).

GC–MS analysis and quantification

The final extracts were transferred and diluted with *n*-hexane and were brought exactly to 1.0 ml by nitrogen blowdown, and then transferred into vials before instrumental analysis. The concentrations of PAHs in the extracts were determined by an Agilent 6890 GC coupled with an Agilent 5973 mass spectrometer and a 7683 auto sampler (Agilent Technology). A $30 \text{ m} \times 0.25 \text{ mm i.d.} \times 0.25 \text{ mm}$ 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^{-1} up to 260°C , with a final holding time of 20 min. Helium was used as the carrier gas. A $1.0 \mu\text{l}$ 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^{-1} . 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 230°C .

The identity of each PAH was confirmed using a standard PAH mixture (610/525/550 in methanol from Chem Service, US) containing the 16 PAHs. The standard PAH mixture was analyzed by GC/MS (Agilent 6890 GC, 5973 MSD) at full scan mode. The molecular ions were selected as the target ion for quantification and another two or three characteristic ions were selected for confirmation. The following 16 USEPA priority PAHs were analyzed:

naphthalene (NAP), acenaphthene (ACE), acenaphthylene (ACY), fluorene (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).

QA/QC measures

All data were subject to strict quality control procedures. Quantification was done using an external calibration method. Recoveries were 108% for NAP, 63.7% for ACE, 79.6% for ACY, 91.7% for FLO, 130% for PHE, 80.7% for ANT, 98.5% for FLA, 97.2% for PYR, 93.1% for BaA, 83.4% for CHR, 97.7% for BbF, 119% for BkF, 69.2% for BaP, 82.0% for DahA, 75.3% for IcdP, and 70.7% for BghiP. A 100 ml solvent blank was concentrated to 200 μ l, and then analyzed in order to monitor the solvent background. No noticeable interferences on the target compounds were identified for the solvent blank. Procedural blanks were determined by going through the extraction and cleanup procedures using glass beads instead of soil samples, with a 1:1 mixture of acetone and dichloromethane used for extraction. The target analytes ranged from not detected to 505 ng g⁻¹. Therefore, all data were blank corrected. The relative difference for individual PAHs identified in method duplicate samples was less than 30%. The detection limits were 2.6×10^{-2} – 5.1×10^{-1} ng g⁻¹ for a 10 g sample.

Data analysis methods

Kriging interpolation

A simple pollution assessment based on the limited sampling data is insufficient because PAH compounds are unevenly distributed in surface soil. Thus spatial interpolation technology is necessary to map and assess PAHs distribution. Among various spatial interpolation methods, Kriging analysis is an unbiased optimal method. It was first used in the estimation of ore reserves in mining and has been extensively applied since then in many other fields (Burgess and Webster 1980). Based on the theory of regionalized variable developed by Matheron (1963) and Krige (1966), Kriging analysis can provide theoretically optimal estimates without bias and with minimal and known variance. In this study, two-dimensional ordinary Kriging was applied in the interpolation process, and spherical models were used to fit all experimental variograms.

Multivariate analysis

Multiple variant techniques, including principal component analysis (PCA) and multiple linear regression analysis (MLRA), were used to quantify PAH sources for the soil samples. PCA is a procedure that determines a few linear combinations of original variables that can be used to summarize a data set (Larsen and Baker 2003). In the present study, PCA was conducted with varimax rotation. Principal components with eigenvalue >1 were retained. The principal components were explained by factor loadings of the PAH species and used to identify source emission composition.

Based on the identified principal components, MLRA is used to quantify the source contribution. The factor scores were regressed against the standard normalized deviate of the sum of PAHs. Based on the regressed equations, the mean contribution for each principal factor can be calculated. The regression coefficients were used to convert the factor scores to the estimates of source mass contributions (Larsen and Baker 2003). In this study, stepwise regression procedure was applied for the MLRA.

Results and discussion

Concentration and spatial distribution of PAHs

All samples were analyzed for the occurrences of 16 PAHs. The maximum and minimum of Σ 16 PAHs were 5,230 and 5.510 ng g⁻¹, respectively. Normality testing showed that the measured PAH concentrations followed normal logarithmic distribution. Basic statistical information of all samples is listed in Table 1.

As shown in Table 1, BghiP is the dominant PAH compound detected in the soil samples, with a geometric mean value of 32.9 ng g⁻¹. BaP and FLA had the largest ranges among the 16 PAH compounds, from 0.640 to 1,330 ng g⁻¹ and 0.260–1,310 ng g⁻¹, respectively. ACY had the smallest range, from 0.030 to 22.6 ng g⁻¹. Four-ring and 5-ring PAHs contributed 33.7 and 25.6% of the total concentration, respectively, while 3-ring PAHs had the lowest proportion of 11.7%. The high molecular weight PAHs (4–6 ring) had a total contribution of 75.5%, which is a bit higher than the ratio reported by Ma et al. (2005).

Kriging interpolation was applied to elucidate the spatial distribution of PAHs in surface soil in the study area. The results were exhibited in Fig. 1. According to the criteria for PAHs in soil reported from Maliszewska-Kordybach (1996), the Σ 16 PAH concentrations of <200, 200–600, 600–1000 and >1000 ng g⁻¹ could be attributed to four groups including none-contaminated, weakly contaminated,

Table 1 The basic statistical information of 16 PAHs (ng g⁻¹)

Compounds	Mean	Geometric mean	Standard deviation	Minimum	Maximum	Median
NAP	35.8	22.9	37.5	nd	255	25.1
ACE	5.43	2.77	8.58	nd	68.1	2.64
ACY	2.59	1.47	3.51	0.03	22.6	1.59
FLO	6.28	3.84	7.61	0.07	74.3	4.36
PHE	15.9	8.62	25.8	0.16	188	8.33
ANT	4.07	1.68	9.02	0.04	70.8	1.59
FLA	31.9	12.2	110	0.26	1310	10.6
PYR	20.1	8.99	38.9	0.18	257	7.77
BaA	22.3	7.08	70.9	0.16	806	6.08
CHR	37.6	17.9	68.3	0.49	437	14.9
BbF	51.7	23.3	95.8	0.45	566	20.6
BkF	21.8	8.54	44.4	0.16	307	7.72
BaP	74.3	24.0	175	0.64	1330	22.4
IcdP	53.4	20.9	116	0.72	775	19.1
DahA	15.6	5.32	35.6	nd	235	5.03
BghiP	87.0	32.9	186	1.12	1100	29.5
2-ring PAHs	50.1	34.4	50.3	0.40	323	33.6
3-ring PAHs	51.8	23.6	129	0.45	1420	21.2
4-ring PAHs	153	68.2	296	1.92	2010	56.7
5-ring PAHs	143	51.9	309	1.70	2090	45.5
6-ring PAHs	87.0	32.9	185	1.12	1100	29.5
Σ16PAHs	485	227	907	5.60	5230	193

n 161, *nd* not detected

contaminated and heavily contaminated, respectively. The spatial distribution of the Σ16 PAH is shown in Fig. 1f, and 5 heavily contaminated zones (HCZs) were identified (I–V).

HCZ I located in the downtown areas, and 3–4 ring PAHs have the highest values in this zone. Relatively low concentrations of 2- and 6-ring PAHs were observed in this area. HCZ II located in the southeast suburb of Beijing city. In Fig. 1f, no clear boundary was identified between HCZ I and II. But for PAHs of different rings, the highest values of 2-, 5- and 6-ring PAHs could be identified in HCZ II. HCZ III is to the north of the downtown area. Relatively high concentrations of 4–6 ring PAHs were seen in this area. HCZ IV is located in the southwest suburb of Beijing city. PAHs of different rings had high concentrations in this area. HCZ V is north of Beijing city containing relatively high concentrations of 4–5 ring PAHs.

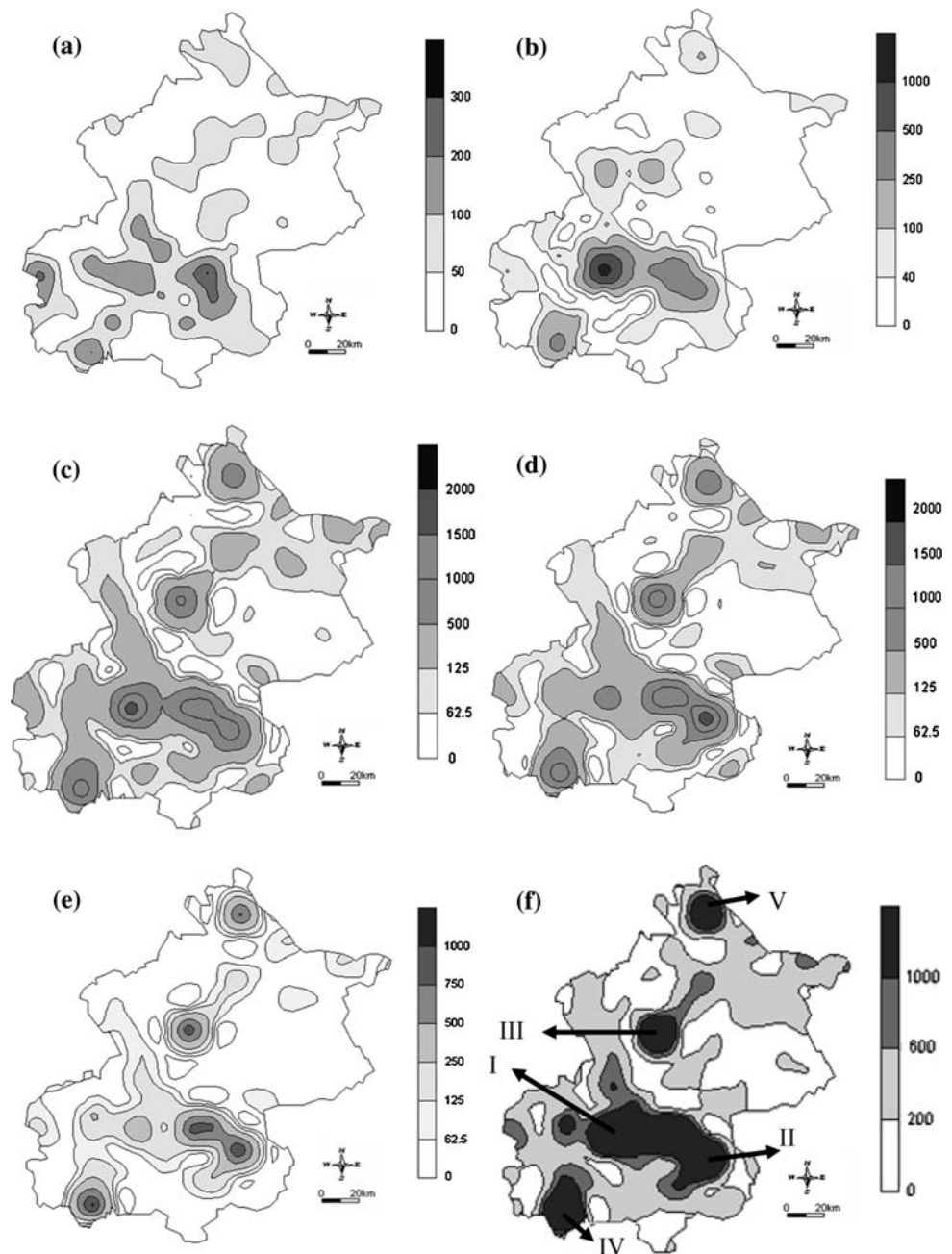
Besides the five HCZs mentioned above, other areas were weakly contaminated. As shown in Fig. 1a–e, the 2-, 3- and 6-ring PAHs were mainly distributed in certain small areas, while the 4- and 5-ring PAHs had a relatively dispersed distribution throughout the sampling area. The spatial distribution of different PAH species reflects the variation of source contributions since PAHs in soil originate from various sources.

PAH source apportionment

Since PAH concentrations followed lognormal distribution, all data were log-transformed (23 samples were removed as outliers using the method of Five-Number Summary) prior to the multivariate analysis. The Henry's law constants of the 16 PAHs are summarized in Table 2 (Ten Hulscher et al. 1992). From Table 2, we could see that, compared with other PAH compounds, NAP is a very volatile compound and the abundance of this compound may mask the variability of the remaining species. Therefore, concentrations of 15 parent PAHs excluding NAP were considered in PCA and MLRA. Three principal components were retrieved for the soil samples. The variance loading of the first three factors is 50.6, 21.5 and 20.2% of the total variability respectively, and the accumulative variance of the three factors is 92.3%. The three factors were used to identify the source information. The loadings of the three factors are shown in Fig. 2. For each factor, the PAHs with relatively high loadings were marked with darker shading.

As shown in Fig. 2, factor 1 is highly weighed in HMW PAHs, including FLA, PYR, BaA, CHR, BbF, BkF, BaP, IcdP, DahA, and BghiP. Among these species, FLA, PYR, CHR and BkF are typical markers for coal combustion (Mastral et al. 1996; Harrison et al. 1996). CHR, BbF, BkF, BaP, IcdP, DahA and BghiP indicate vehicular emission

Fig. 1 Spatial distribution of PAH concentrations for different rings and $\Sigma 16$ PAHs: **a** 2-ring PAHs, **b** 3-ring PAHs, **c** 4-ring PAHs, **d** 5-ring PAHs, **e** 6-ring PAHs, **f** $\Sigma 16$ PAHs



(Simcik et al. 1999; Motelay-Massei et al. 2007). Therefore, factor 1 can be attributed to composite sources of coal combustion and vehicular emission. Coal is the major type of energy consumed in Beijing. According to Xu (2005), Beijing consumed 34.2 million metric tons of standard coal equivalent in 2003, among which the coal consumption accounted for 46.8%. Coal is heavily used for domestic heating in winter, which lasts for 4 months. Coal consumption is one of the primary contributors to PAHs in surface soil. Another important source is vehicular emission. Since 1990, the number of vehicles in Beijing has increased at an annual rate of 15% and reached

2.14 million in 2005. Beijing is facing serious traffic congestion and the associated air pollution.

Factor 2 is mostly associated with ACE, FLO and PHE, which are indicators for coking sources (Simcik et al. 1999). Coking is an important industrial sector in Beijing, thus factor 2 can be considered as indicator of coking industry emission.

Factor 3 is dominated by ANT and ACY. In previous studies, ANT and ACY are identified as a typical tracer of a petroleum emission source, including leakage and the spill of petroleum-related products during production and transportation (Motelay-Massei et al. 2007; Larson and

Table 2 Henry's law constant of 16 PAHs (Ten Hulscher et al. 1992)

PAHs	Henry's law constant (Pa m ³ /mol)
Naphthalene	64.2
Acenaphthylene	17.8
Acenaphthene	45.4
Fluorene	18.6
Phenanthrene	6.33
Anthracene	10.5
Fluoranthene	1.49
Pyrene	1.53
Benz(<i>a</i>)anthracene	0.861
Chrysene	0.183
Benzo(<i>b</i>)fluoranthene	0.983
Benzo(<i>k</i>)fluoranthene	0.0853
Benzo(<i>a</i>)pyrene	0.239
Indeno(1, 2,3- <i>cd</i>)pyrene	0.0124
Dibenz(<i>a,h</i>)anthracene	0.0310
Benzo(<i>g,h,i</i>)perylene	0.00770

Baker 2003). This factor accounted for 20.2% of the total variance, indicating that a petroleum emission is a significant source of PAHs in the study area.

Ratios of PAH isomer are also widely applied for PAH source identification. Previous studies have demonstrated that PAH isomer pairs with similar molecular weight were useful for PAH source apportionment (Cai et al. 2007;

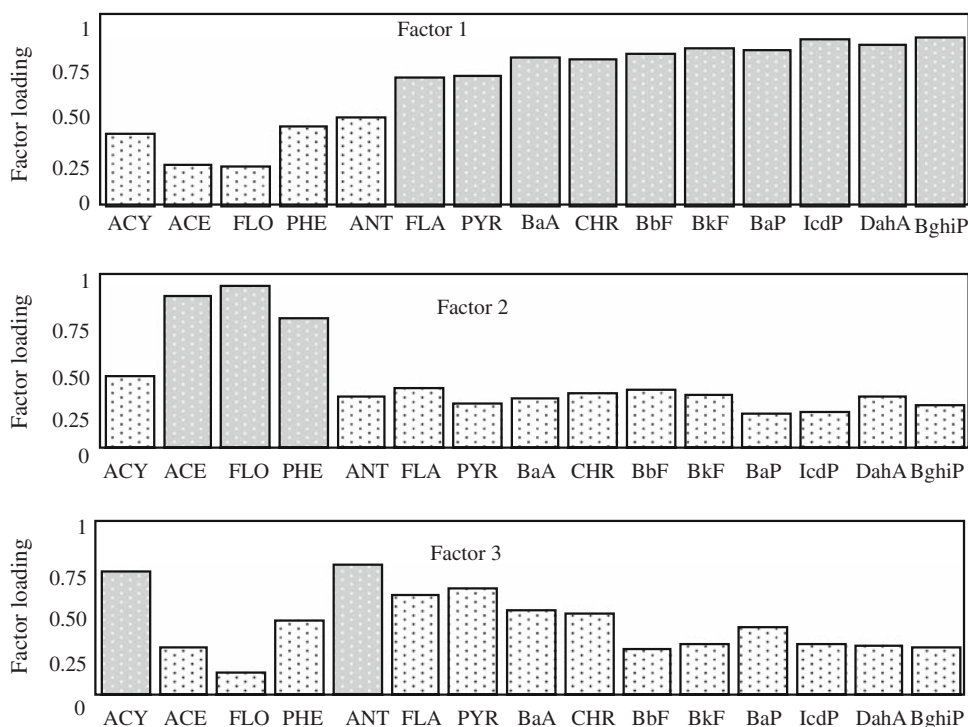
Budzinski et al. 1997; Yunker et al. 2002). According to Budzinski et al. (1997), a ratio of ANT/(ANT + PHE) < 0.1 indicates petroleum, while the ratio > 0.1 signals a dominance of combustion. In this study, the ANT/(ANT + PHE) ratios are between 0.010 and 0.77 (78% of the 161 samples are larger than 0.1), suggesting a dominance of combustion. Another widely used indicator for 3- and 4-ring PAHs is FLA/(FLA+PYR). The ratio lower than 0.4 signals petroleum, 0.4–0.5 implies petroleum combustion, and higher than 0.5 indicates combustion of coal, straw and wood (Yunker et al. 2002). In this study, the FLA/(FLA+PYR) ratios ranged from 0.45 to 0.98 (88% of the 161 samples are larger than 0.50), indicating combustion of coal, straw and wood. Comparison of the PCA results and isomer ratios shows that combustion of fossil fuels, including coal and petroleum oil, is the major source of PAHs in surface soil in the study area.

Estimation of source contribution

Multiple linear regression was performed stepwise using the absolute factor scores as independent variables, and the total $\Sigma 15$ PAH concentrations as the dependent variables. The regressed equation is:

$$\sum \ln PAHs = 0.797 \text{ factor1} + 0.449 \text{ factor2} + 0.385 \text{ factor3}$$

The contributions of three major sources were calculated based on the regression coefficients. The mean percent contributions are summarized in Fig. 3a.

Fig. 2 Factor loadings of factor analysis based on the measured soil PAHs in Beijing

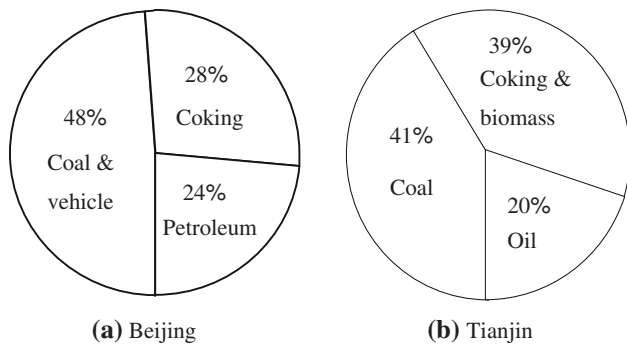


Fig. 3 Contributions of PAHs sources in surface soil in **a** Beijing and **b** Tianjin (Tianjin data were cited from Zuo et al. 2007)

As illustrated in Fig. 3a, the contributions of coal/vehicular emission, coking and petroleum sources were 48, 28 and 24%, respectively. Coal/vehicular emission was the dominant PAH source, accounting for nearly half of the total contribution to the surface soil in Beijing. The source contributions of PAHs in surface soil in Tianjin (Zuo et al. 2007) were cited in Fig. 3b for comparison. Tianjin is one of the largest industrial cities in China, bordered by Beijing to the northwest. The energy consumption structure of Tianjin is similar to that of Beijing. In Fig. 3b, coal and petroleum contributed over 60% of the total source contribution. The remaining 39% were from either biomass burning or coking. In both cities, coal is the primary contributor to PAHs in surface soil. In Beijing, the percent contribution of coking is less than that of Tianjin, indicating that the environmental impact of coking in Beijing is relatively small.

With the mean percent contributions, the source mass contributions at each sampling site can be calculated (Larsen and Baker 2003). The mass contribution of $\Sigma 15$ PAHs in surface soil was 16.4 ng g^{-1} for coal/vehicular emission, 4.63 ng g^{-1} for coking emission, and 3.70 ng g^{-1} for petroleum sources. The calculated mass contributions were compared with the source emission estimated based on the amount of fuel consumed and emission factors from the literature (Xu et al. 2006). The comparison is presented in Fig. 4. The profile of the calculated source contributions in surface soil is generally consistent with the profile of the estimated source emission. According to the source emission estimation, coal combustion is the largest source of PAHs, followed by coking emission. The percentage of PAHs released from coal combustion and the coking industry was 83.2% in Beijing in 2003, which was close to the calculated mass contribution of coal combustion and coking emission. The total PAHs generated from sources of petroleum, wood combustion and biomass burning accounted for 16.8% of the total emission. However, wood combustion and biomass burning could not be distinguished from the petroleum source through PCA.

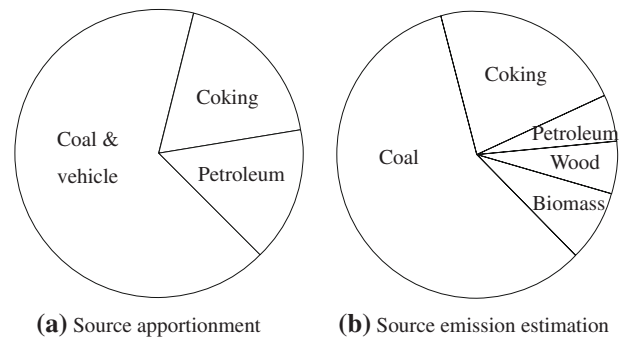


Fig. 4 Source contributions of PAHs in surface soil in Beijing: **a** source apportionment; **b** source emission estimation (Source emission data were cited from Xu et al. 2006)

Therefore, the calculated mass contribution of the petroleum source through PCA/MLRA was 5.9 times that of the data estimated from source emission.

Spatial interpolation of source contributions

A simple assessment of source contribution at the limited sampling sites is insufficient because the PAHs contributions from the identified sources are unevenly distributed in the study area. Thus, the ordinary Kriging method was applied to map and assess the spatial feature of the PAH contribution from each source. The Kriging interpolation maps for each source are presented in Fig. 5.

Figure 5a illustrates the spatial contour of the contribution from coal and vehicular sources. High concentrations were mainly found in downtown areas of Beijing, indicating a large contribution from coal and vehicular sources in this area. The downtown area of Beijing is the business and finance center of China, and features a large population density and high traffic volume. Pollution from vehicular emission and coal combustion for heating in winter is high in this area. A high contribution of this source is also observed in the northeastern, southeastern and western areas, where industrial zones are located. Currently, coal-burned boilers are widely used to provide steam, hot water and heating in factories in the industrial zones. In the southeastern area, it was reported that the major sources for PAHs in total suspended particles (TSP) included gasoline, diesel and coal combustion (Zhang et al. 2007), which is consistent with the source profile for PAHs in the surface soil. Atmospheric TSP may act as an important input pathway for PAHs in surface soil in this area.

The spatial distribution of coking emission is presented in Fig. 5b. The points with high PAHs concentration released from a coking source corresponded to the locations of main coking and gas plants in Beijing, including Beijing Coking Plant, Shougang Coking Plant, Beijing Chaoyang Shuangqiao Gas Plant, Huairou Huogezhuang

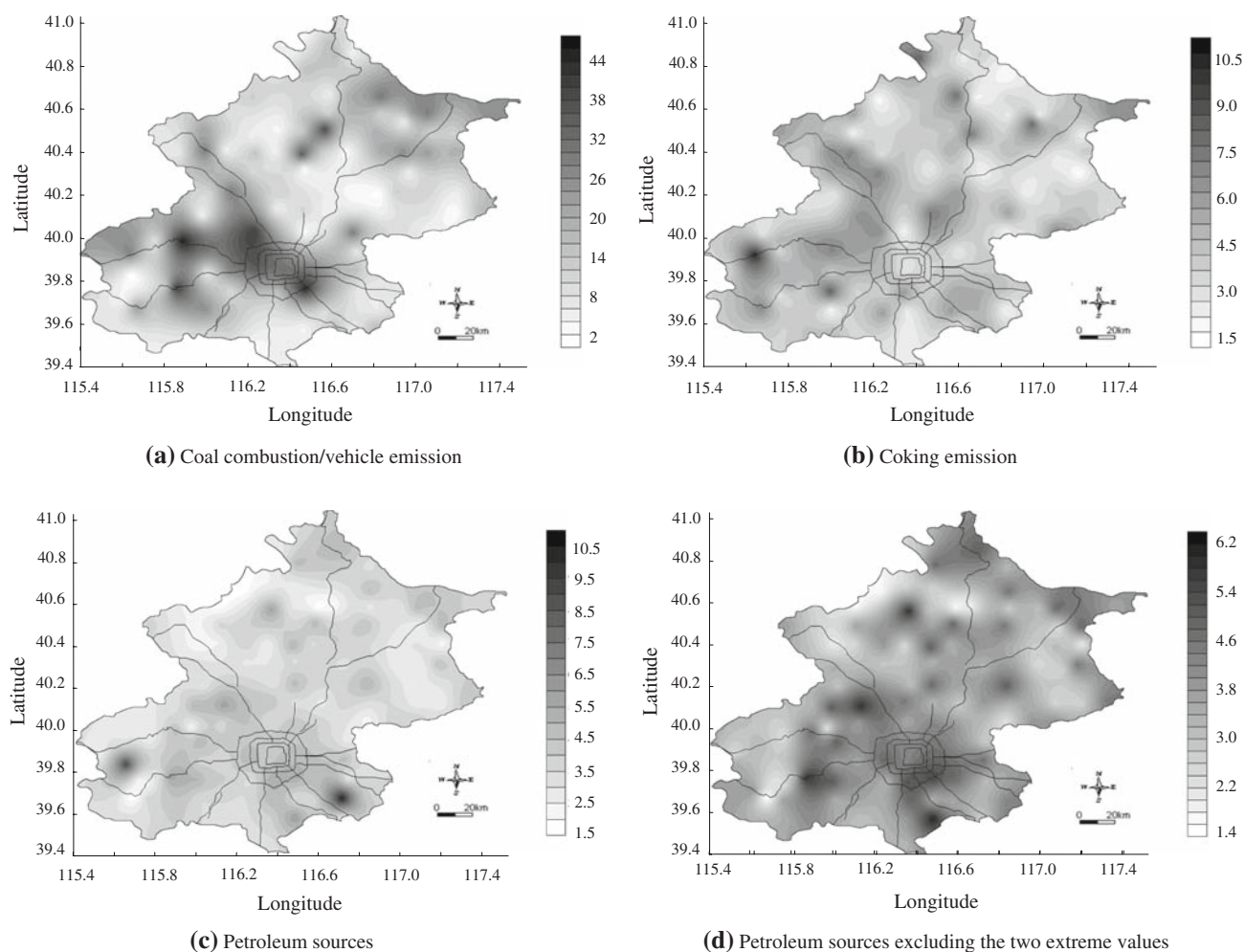


Fig. 5 Distribution of PAHs source contributions in surface soil in Beijing (ng g^{-1}): **a** Coal combustion/vehicle emission, **b** coking emission, **c** petroleum sources, **d** petroleum sources excluding the two

extreme values. (Line the ring road in downtown area and the national highway in suburban area)

Gas Plant, Tongzhou Coking Plant, Beijing Xiluyuan Gas Plant, and Pinggu Fuhua Coking Plant. The emissions from these coking and gas plants cause serious air pollution to the area. Currently, natural gas is being supplied in Beijing to reduce the demand for gasoline and coal. The coking and gas plants are being relocated to neighbor provinces. With the decrease of gas and coke production, the PAH pollution from these sources are expected to decrease significantly.

The distribution of petroleum contribution is illustrated in Fig. 5c, d. In Fig. 5c, two points with extremely high concentrations originated from petroleum sources observed in the southeastern and western suburban areas. Industrial zones are located in these areas. However, the spatial characteristics of petroleum sources at other locations may be masked by the two extreme points. Kriging interpolation is thus re-conducted excluding the two extreme values, and the updated contour map is presented in Fig. 5d. In Fig. 5d, the contribution of petroleum sources is seen distributed

widely throughout the urban and suburban areas of Beijing. This indicates that PAH contamination from petroleum sources, including leakage of vehicle fuel tanks, transportation spillage of petroleum products, and leakage from gas stations, widely effect Beijing.

Based on the source identification and spatial distribution analysis above, control of PAH contamination in surface soil in Beijing should be implemented with abatement of emission from the three sources. According to Xu (2005), the consumption of coal and oil (gasoline + diesel) in 2003 is 16 million tonnes of standard coal equivalent and 4.2 million tonnes of standard coal equivalent, respectively. Control of oil consumption in transportation and replacement with cleaner energy can reduce the PAH contamination in surface soil, especially of the HMW species. For the control of low molecular weight PAHs, effective measures include reduction of coal consumption, and improvement of storage and transportation of petroleum-related products.

Conclusion

The maximum and minimum of $\Sigma 16$ PAHs in topsoil of Beijing were 5,230 and 5.510 ng g⁻¹, respectively, and BghiP and ACY were the dominant and lowest compounds. Four-ring and 5-ring PAHs contributed 33.7 and 25.6% of the total concentration, respectively. Five HCZs were identified using Kriging interpolation.

Multivariate analysis showed that, the dominant sources of PAHs in surface soil in Beijing were coal combustion and vehicular emission, with a composite contribution of 48% of the total sources. Other sources included coke production and petroleum, with source contribution of 28 and 24%, respectively. The calculated mass contribution of 15 PAHs from coal combustion/vehicular emission, coking emission, and petroleum related products was 16.4, 4.63 and 3.70 ng g⁻¹, respectively.

The spatial distribution of source contributions was affected by the functional zoning and energy consumption structure. Generally, PAH contamination of surface soil was more serious in downtown areas than in suburban areas. Coal combustion and vehicular emission were the main PAH sources in the downtown areas. PAHs generated from coking emission were mainly distributed in the suburban areas. Contribution from petroleum sources was widely distributed in the Beijing area. Based on the identified PAH sources, control of PAH contamination of soil in Beijing could be reality.

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