

Original Research

Polycyclic Aromatic Hydrocarbons in Soil Around Coal-Fired Power Plants in Shandong, China

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Abstract

This study comprehensively investigated the concentrations, distribution patterns, sources, and health risks of 16 polycyclic aromatic hydrocarbons (PAHs) found in soils around two thermal power plants in Shandong, China. The total concentrations of the 16 total PAHs (Σ_{16} PAH) in the Longguang (LG) and Xinyuan (XY) thermal power plants were 1,031.78-2,744.06 and 1,383.8-2,924.8 $\mu\text{g}/\text{kg}$, respectively, with mean values of 1,925.2 and 2,303.1 $\mu\text{g}/\text{kg}$, respectively. Seven carcinogenic PAHs accounted for 45.5% and 55.7% of Σ_{16} PAHs in soils around LG and XY thermal power plants, respectively. A similar composition pattern of PAHs was observed in soils around the two power plants. The contributions of four-ring PAHs, namely, fluoranthene (FLA), pyrene (PYR), benz[a]anthracene (BaA), and chrysene (CHR), were high; thus, the four-ring PAHs dominated in soils around the two power plants. With the increase of distance, most individual PAH concentrations decreased, and Σ_{16} PAHs concentrations decreased significantly from 200 m to 500 m. However, the contributions of two- to three-ring PAHs increased with distance, whereas the contributions of four-ring PAHs decreased. Four PAH sources in soils around the two thermal power plants were identified by positive matrix factorization. Coal combustion was the main source of soil PAHs. Health-risk assessment based on toxic equivalency factors of benzo[a]pyrene indicated no risk of PAH contamination in soils around LG power plant and a low risk in those around XY power plant. This study highlights the distribution and sources of PAHs in soil around thermal power plants.

Keywords: PAHs, coal-fired power plant, soil, positive matrix factorization, risk assessment

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds including two or more aromatic

rings [1], and human health can be threatened by their teratogenicity, carcinogenicity, and mutagenicity [2]. They can be eliminated via biological degradation [3] and photodegradation [4], and their content in the environment can generally be maintained at a low level. However, in recent years, this balance has been destroyed due to anthropogenic activities, which are the main sources of

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PAHs [5]. Consequently, PAHs are now ubiquitous in the environment, and their content in the environment has increased [6]. PAHs are most probably derived from diffuse sources, such as dry and wet deposition from the atmosphere [2], and they tend to accumulate in soil.

With the development of industrialization, coal has become one of the most abundant and important sources of global energy. In 2015 global coal production was 8.00 billion tons, and total coal production in China was 3.65 billion tons [7]. In the same year, coal consumption in China was 3.97 billion tons, which accounted for about half of global coal consumption [8]. The share of coal in energy consumption in China has reached 64%, which is far higher than the global average level of 30% [9]. Coal combustion is an important source of PAHs, particularly some carcinogenic species. Most studies on pollution from coal-fired power plants have focused on fly ash around large industrial coal-fired plants [10]. These studies show that phenanthrene is the dominant PAH and that more than 80% of the PAHs found in ash samples around coal-fired plants exist as three- and four-ring PAHs [11]. In addition, the total PAH content of fly ash is higher than that of bottom ash [12]. The high concentrations of PAHs detected in leaves of shrubs near coal-fired power plants could pinpoint conspicuous air contamination [13]. A number of reports have shown that coal tar is an important source of urban PAH and that high PAH concentrations in soil and water have been detected in the vicinity of coal or coal gangue dumps [14-15]. Obvious differences have also been observed in the PAH content of soil in the northern and southern regions of China. The content of PAHs in soil in the southern region of China is generally lower than that in the northern region, probably because of coal firing in the latter [16]. However, research on the composition, variation with distance, sources, and

health risks of PAHs in soil affected by coal-firing is scarce.

Shandong Province (34.61–37.91°N and 115.08–122.41°E) is one of the largest industrial producers in northern China. It is dominated by petroleum, chemical, electronic, fertilizer, tire, and rubber industries. It has a temperate climate with an average annual temperature of 11–14°C and annual precipitation 550–950 mm. In China, Shandong has the greatest number or the highest installed capacity of coal-fired power plants. The Longguang (LG) thermal power plant and Xinyuan (XY) thermal power plant are located in Weifang and Zaozhuang, Shandong, respectively. The LG thermal power plant utilizes 12 MW generator sets, whereas the XY thermal power plant utilizes 30 MW generator sets. Both power plants have been in operation for more than 10 years, and they share the same geographical position and climate conditions. Given these similarities, we chose the two power plants for this study. The study measured the concentrations of PAHs in soils around the LG and XY coal-fired power plants. Then we aimed to (1) analyze the variation of PAHs with distance, (2) fingerprint the sources of PAHs, and (3) evaluate the human health risks of PAHs in soils around coal-fired power plants. The results will broaden the understanding of characteristics of PAHs in contaminated soils near thermal power plants.

Materials and Methods

Soil Sampling

Thirteen surface soil samples (0–20 cm) from around the LG and XY power plants were collected in July 2015 (Fig. 1). Four samples were collected from

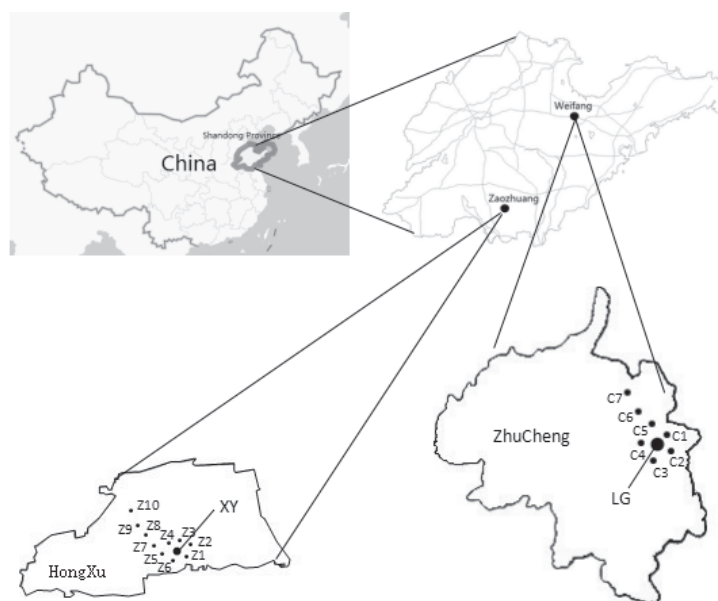


Fig. 1. Schematic map showing soil sampling sites around Longguang and Xinyuan power plants in Shandong.

the area close to the LG power plant, six samples were collected from the area close to the XY power plant, and three samples were collected at distances of 200, 500, and 1000 m from the LG and XY power plants, respectively. All the samples were freeze-dried, homogenized, sieved into 20-mesh-sized particles, and then kept in a refrigerator at -4°C.

Sample Extraction and Cleanup

Exactly 2 g of soil samples and 10 mL dichloromethane (DCM) were put into a 30 mL centrifuge tube, which was sonicated for 1 h. The mixture was centrifuged at 4,000 r/min for 10 min, and the extract was collected. The same extraction process was repeated by adding 10 mL DCM. The two extracted solutions were combined and concentrated to approximately 1 mL through rotary evaporation. The silica gel was used to clean up the concentrated solution. The glass chromatography column (20 cm×10 mm i.d.) was packed from the bottom with 4 g activated silica gel (200-300 mesh) and 4 g anhydrous sodium sulfate. Elution was performed with 20 mL DCM and n-hexane (v:v = 1:1). The eluent was further concentrated nearly to dryness and solvent-exchanged to 2.0 mL methanol for analysis.

High-Performance Liquid Chromatography Analysis

PAH concentration was measured through high-performance liquid chromatography equipped with an ultraviolet and fluorescent detector (HPLC-UV-FLD, Shimadzu). Then, 16 PAHs were separated with an Inertsil ODS-P column (250 × 4.6 mm, 3.5 μm particle size, 1,000 nm pore size) with the mobile phase of methanol-water (80:20, by volume) through gradient elution with a flow rate of 1.0 mL/min. Detection was carried out with the FLD wavelength switching program at excitation wavelengths of 265, 260, 290, and 250 nm, and emission wavelengths of 420, 430, and 500 nm. UV detection with a double wavelength pattern was performed at wavelength channel 1 of 254 nm and wavelength channel 2 of 220, 280, and 210 nm. The column temperature was set to 40°C, and the injection volume was 20 μL.

A composite standard solution of 16 PAHs, including naphthalene (NAP), acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLU), 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), dibenzo[a,h]anthracene (DBA), indeno[1,2,3-cd]pyrene (IPY), and benzo[ghi]perylene (BPE) was purchased from AccuStandard Company (USA). All solvents were HPLC grade.

Quality Assurance and Quality Control

The method blank was applied and showed no detectable amounts of PAHs. The sample duplicate was analyzed, and the variation in PAH concentrations was less than 9%. The recoveries of the procedure of the 16 PAHs were controlled in the range of 60.2–119.3%, and the relative standard deviation was 0.6–14.8%.

The external standard calibration method was implemented, and determined instrument stability and confirmed the calibration curve by analyzing the standard mixture for every 10 samples. The limits of detection were in the range of 0.07 μg/kg to 2 μg/kg.

Source Identification Methods

In this study, the emission sources of PAHs in the soil samples were distinguished by diagnostic ratios and the positive matrix factorization (PMF) model. Diagnostic ratios have been commonly applied to distinguish the sources of PAHs [17]. However, the ratios cannot provide accurate information on the contributions of PAH sources [18]. Therefore, the PMF model, which was developed in 1994, was used in this study. The concrete calculation method for the parameters of the PMF model was based on the work of Reff et al. (2007) [19-20]. The current work used the U.S. EPA PMF 5.0 model [21]. The measured PAH concentration and the estimated uncertainty (u) of the concentration are required for the PMF 5.0 model. In the concentration file, the concentration of PAHs below the method detection limits (MDL) was substituted by half the detection limit. The uncertainty of the concentration was calculated by applying MDL to each PAH and the measurement uncertainties (MU) defined as 10% of the measured concentration values. If the concentration \leq MDL, the uncertainty u is calculated as

$$u = \frac{5}{6} \times \text{MDL} \quad (1)$$

...and when the concentration $>$ MDL, u is calculated as:

$$u = \sqrt{(\text{MU} \times \text{concentration})^2 + (\text{MDL})^2} \quad (2)$$

PAH Risk Assessment

Benzo[a]pyrene equivalent concentrations (BaP_{eq}) could be estimated by using benzo[a]pyrene toxic equivalency factors (TEFs). The carcinogenicity of the other PAHs relative to BaP was quantified to estimate BaP_{eq} [22]. BaP_{eq} was calculated according to Eq. 1 [23]:

$$\text{BaP}_{\text{eq}} = C_i \times \text{TEF}_i \quad (3)$$

...where C_i is the concentration of i th PAH (μg/kg) and TEF_i is the toxic equivalency factor of i th PAH.

Results and Discussion

PAH Concentrations and Composition

The total concentrations of the 16 PAHs (Σ_{16} PAHs) in soils around the LG power plant varied from 1,031.8 $\mu\text{g}/\text{kg}$ to 2,744.1 $\mu\text{g}/\text{kg}$, with a mean of 1,936.2 $\mu\text{g}/\text{kg}$ (Table 1). The concentrations of seven carcinogenic PAHs (Σ CPAHs; including BaA, CHR, BbF, BkF, BaP, IPY, and DBA) were in the range of 541.2 $\mu\text{g}/\text{kg}$ to 1,355.7 $\mu\text{g}/\text{kg}$, accounting for 45.5% of Σ_{16} PAHs. In terms of the concentrations of the PAHs of different rings, the concentrations of the four-ring PAHs were high with a mean of 862.3 $\mu\text{g}/\text{kg}$, followed by the five-ring PAHs. The concentrations of the six- and two- to three-ring PAHs were relatively low.

Σ_{16} PAHs in soils around the XY power plant varied from 2,252.4 $\mu\text{g}/\text{kg}$ to 3,924.8 $\mu\text{g}/\text{kg}$, with a mean of 2,594.4 $\mu\text{g}/\text{kg}$. Σ CPAHs were in the range of 1,181.4 $\mu\text{g}/\text{kg}$ to 1,966.4 $\mu\text{g}/\text{kg}$, with a mean concentration of 1,445.0 $\mu\text{g}/\text{kg}$, which accounted for

55.7% of Σ_{16} PAHs. The concentration of the four-ring PAH was relatively high with a mean of 1,138.5 $\mu\text{g}/\text{kg}$, followed by the concentrations of the five-ring and two- to three-ring PAHs. The concentration of the six-ring PAHs was relatively low.

Maliszewska-Kordybach established a standard classification method for PAH-contaminated soil according to the data of soil from several European countries and combined it with the risk of human exposure [24]. According to this standard, all the soil samples around the two power plants were heavily contaminated ($>1,000$ $\mu\text{g}/\text{kg}$).

Σ_{16} PAHs in soils around the point sources are presented in Table 2. The PAH concentrations in this study were lower than those around other coal-fired power plants, such as those in India (2538.24 $\mu\text{g}/\text{kg}$) [13], South Africa (9,730-61,240 $\mu\text{g}/\text{kg}$) [2] and Huainan (2,830-5,320 $\mu\text{g}/\text{kg}$) [25]. The PAH concentrations in the coking areas in Shanxi Province in China (171.67-3,176.79 $\mu\text{g}/\text{kg}$) and a coking plant in France (1,355.78 $\mu\text{g}/\text{kg}$) were comparable to those in this

Table 1. Concentrations of PAHs in soils around the LG and XY power plants ($\mu\text{g}/\text{kg}$).

Compounds	LG power plant					XY power plant				
	Min	Max	Mean	Median	SD	Min	Max	Mean	Median	SD
NAP	21.5	66.8	38.8	33.5	20.1	16.6	48.2	32.1	32.1	11.2
ACY	60.4	78.2	69.8	70.3	8.0	60.1	80.4	72.8	73.9	6.8
ACE	18.0	50.5	31.1	28.0	14.3	19.5	45.8	28.7	26.7	9.8
FLU	3.5	9.8	6.7	6.8	2.6	7.9	143.0	59.1	51.6	49.8
PHE	2.0	29.4	14.6	13.5	14.2	28.9	63.0	45.2	42.7	13.6
ANT	35.7	192.4	121.9	129.7	80.0	167.2	491.1	272.8	245.9	125.6
FLA	108.2	373.8	190.2	139.3	123.3	148.3	304.9	217.4	213.2	57.1
PYR	211.7	358.6	302.2	319.4	67.3	69.4	308.7	180.6	186.5	95.5
BaA	124.3	508.3	269.3	222.3	182.4	338.1	538.7	436.9	412.7	78.1
CHR	50.1	246.7	100.6	52.7	97.4	210.8	518.2	303.6	284.7	112.6
BbF	24.9	74.8	44.7	39.5	22.8	45.4	66.7	55.7	54.1	8.5
BkF	202.5	420.5	315.8	320.1	100.6	40.2	85.4	57.2	50.5	16.8
BaP	3.0	40.4	21.6	21.5	21.4	6.7	80.5	32.3	26.3	27.7
DBA	105.8	128.7	121.8	126.4	10.9	239.4	586.2	460.7	477.7	116.5
BPE	16.7	495.8	279.9	303.6	197.5	49.3	493.3	240.7	208.6	190.0
IPY	5.2	9.0	7.1	7.1	1.8	45.7	123.0	98.6	107.3	28.0
2~3 rings	154.0	423.6	282.9	277.0	117.3	352.8	851.5	510.6	463.9	185.6
4 rings	497.8	1419.7	862.3	765.8	400.6	945.4	1670.6	1138.5	1072.1	269.0
5 rings	355.0	641.5	503.9	509.6	136.3	380.8	816.9	605.9	605.8	141.3
6 rings	25.0	501.0	287.1	311.1	196.3	159.0	585.8	339.3	289.3	188.1
Σ CPAHs	541.2	1355.7	880.9	813.4	341.7	1181.4	1966.4	1445.0	1381.4	268.5
Σ_{16} PAHs	1031.8	2744.1	1936.2	1984.5	712.1	2252.4	3924.8	2594.4	2335.5	655.4

Table 2. Total concentrations of 16 PAHs ($\mu\text{g}/\text{kg}$) in soils around point sources.

Region/area	Sampling site types	Range	Mean	Reference
Anhui Province, China	Coal mines	130 ~ 3540	840	Wang et al.(2010a)
Xinzhou, China	Coal production area	n.d.~782	202	Zhao et al.(2014)
Shanxi, China	Coking areas	171.67~3176.79	982.18	Cui et al. (2015)
Shanxi, China	Coke production base	247~1410	691	Duan et al. (2015)
France	Coking plant		1335.78	Biache et al. (2008)
Indian	Coal-fired power plant	951.79~4362.35	2538.24	Sharma et al. (2009)
South Africa	Coal-fired power plants	9730~61240		Okedeyi et al. (2013)
Huainan, China	Coal-fired power plants	2830~5320		Wang et al. (2013a)
China	Thermal desorption plant	3180~3532	3338.2	Liu et al. (2015)
Milan, Italy	Manufacturing gas plant		1669300	Saponaro et al. (2002)
Gdańsk, Poland	Municipal solid waste	892~3514	1974	Melnyk et al. (2015)
Guangdong, China	E-waste recycling site	127~10600	1230	Wang et al. (2012)
Taizhou, China	E-waste recycling town	262.6~3420.2	1095.8	Tang et al. (2010)
Longtang, China	E-waste recycling site	25~4300	514.4	Huang et al. (2014)
Taizhou, China	E-waste recycling sites	330~790	566.67	Shen et al. (2009)
Abraka, Nigeria	Fuel stations	0.82~62.98	14.48	Emoyan et al.(2011)
Arizona, USA	Urban highway	67~10117	523	Marusenko et al. (2011)
Zhejiang, China	Municipal waste incinerator	2222.53~6883.91	3859.06	Shen et al. (2010)
Yangtze River Delta region, China	Industrial areas	471.30		Wang et al. (2017)
Zhucheng, China	Coal-fired power plant	1031.8~3744.8	1925.2	In this study
Zaozhuang, China	Coal-fired power plant	1383.8~3924.8	2303.1	In this study

study [26-27], but they were higher than those in soils from the coal production area in Anhui Province (840 $\mu\text{g}/\text{kg}$) [28], Xinzhou (n.d.-782 $\mu\text{g}/\text{kg}$) [18] and Shanxi (247-1,410 $\mu\text{g}/\text{kg}$) [29] in China. Furthermore, the PAH concentrations in this study were higher than those in most e-waste recycling sites in Guangdong (1,230 $\mu\text{g}/\text{kg}$) and Taizhou (262.6-3,420.2 $\mu\text{g}/\text{kg}$; 330-790 $\mu\text{g}/\text{kg}$) in China [30-32].

The PAH concentrations in this study and other areas around the power plants were lower than those of a thermal desorption plant (3,338.2 $\mu\text{g}/\text{kg}$), municipal waste incinerator in Zhejiang (222.53-6,883.91 $\mu\text{g}/\text{kg}$), and manufacturing gas plant (1,669,300 $\mu\text{g}/\text{kg}$) [33-35]. By contrast, they were higher than those of fuel stations (0.82-62.98 $\mu\text{g}/\text{kg}$), urban highway (523 $\mu\text{g}/\text{kg}$), industrial areas (471.3 $\mu\text{g}/\text{kg}$), and municipal solid waste site (1,974 $\mu\text{g}/\text{kg}$) [36-39]. Therefore, the PAH concentrations in soils around the coal-fired power plants in this study were in the middle range.

In soils around the LG power plant, BkF, PYR, BPE, and BaA were the major PAH compounds, accounting for 16.3%, 15.6%, 14.5%, and 13.9% of the total PAHs, respectively (Fig. 2). In soils around the XY power plant, DBA, BaA, CHR, and ANT were the major

PAH compounds, accounting for 17.8%, 16.8%, 11.7%, and 10.5%, respectively. A similar composition pattern of PAHs was observed in soils around the two power plants. The contributions of the four-ring PAHs, including FLA, PYR, BaA, and CHR, were high, resulting in the

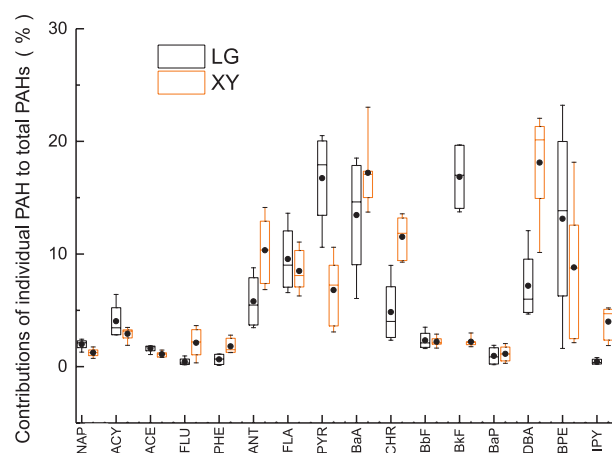


Fig. 2. Contributions (%) of individual PAH compounds (lines on top, bottom, and middle denote max., min., and median values, respectively; dot denotes the mean value).

four-ring PAHs being predominant with a concentration of 44.6% in the LG power plants and 44.0% in the XY power plants. Although the contributions of BkF and DBA in the two power plants differed, the contributions of the five-ring PAHs were in the range of 20-30%, followed by those of the four-ring PAHs. Even with the contributions of BPE (one of six-ring PAHs) being high in the two power plants, the contributions of the six-ring PAHs were relatively low, accounting for 13.0% of that in the LG power plant and 12.8% of that in the XY power plant. By comparison, the contributions of the two- to three-ring PAHs in the two power plants were also less than 20%.

BaA showed high concentrations in soil samples from around the two thermal power plants, whereas BaP and PHE showed a small proportion. DBA in soil samples around the XY thermal power plant and BkF in soil samples around the LG thermal power plant showed large proportions, but they showed a smaller proportion in soil samples around another thermal power plant. Previous studies found that the most abundant PAHs around a power plant in South Africa were CHR, BkF, and IPY [3]. The finding of the current study is similar to this result.

PHE, FLA, BbF, NAP, PYR, and CHR were dominant in soil samples from a coke production base [28], whereas ANT, ACE, DBA, PYR, and CHR were dominant in soil samples from around a fuel station [36]. Some of the major compounds identified in the coke production base and fuel station were consistent with the results obtained for the two power plants [29, 36]. In soils around an e-waste recycling area, the most abundant PAHs were NAP, PHE, and FLU [30]. In all samples around a thermal desorption plant in China, the most abundant PAHs were FLU and ANT [33]. In all the soil samples around a manufacturing gas plant site, the most abundant PAHs were FLA, PYR, BaP, and IPY [35]. Given the availability of various sources, the composition of PAHs in soils from different sampling site types is disparate.

Variation of PAHs with Distance

With increasing distance, most PAH compound concentrations decreased, and \sum_{16} PAH concentrations decreased significantly from 200 m to 500 m (Fig. 3). The mean \sum_{16} PAH concentrations in soils from a distance of 1,000 m from the LG and XY power plants were 1,593.45 $\mu\text{g}/\text{kg}$ and 1,475.87 $\mu\text{g}/\text{kg}$, respectively. The contributions of PAHs with different rings presented a difference with distance. The contributions of two- to three-rings PAHs increased with distance from 14.6% to 27.3% in soils near LG and from 19.7% to more than 35% in soils near XY. However, the contributions of the four-ring PAHs decreased from 45% to 30-35%. The contributions of the five-ring PAHs in soils near LG increased with distance from 26.0% to 33.1%, but near the XY power plant, the contributions decreased from 23.4% to 16.9% (Fig. 4). The contributions of the six-ring PAHs decreased from 14.8% to 10.2% and from 13.1% to 11.2%

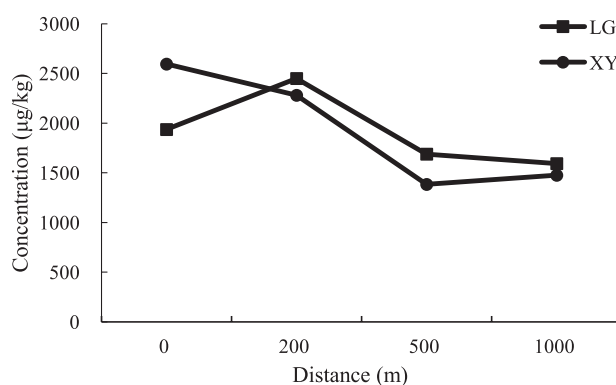


Fig. 3. Variation of 16 PAH concentrations with distance in soils from the LG a) and XY b) power plants.

in soils near the LG and XY power plants, respectively. Despite the changes in contributions, the four- and five-ring PAHs still accounted for a large percentage at a distance of 1,000 m from the two power plants.

Low molecular weight PAHs (LMW PAHs, \leq three rings) exhibit high vapor pressure, exist mainly in the gaseous phase, and can be transported for a long distance [40]. Thus, the percentage of two- to three-rings PAHs increases with distance [41]. Moreover, LMW PAHs migrate more easily than high molecular weight PAHs (HMW PAHs, \geq four rings) [42]. The samples sites around the XY power plant were situated at altitudes of 67, 66, 65, and 58 m at a distances of 0, 200, 500, and 1,000 m, respectively. The decrease in altitude with distance may help LMW PAHs migrate in the soil [43]. On the contrary, HMW PAHs cannot easily be migrated and may even accumulate in soils found adjacent to the power plants. A garbage transfer station was situated at a distance of 700 m away from the LG power plant, and a transportation center was located at a distance of about 600 m from the XY power plant. Previous studies found that these point sources may release a great deal of LMW PAHs, such as ANT and FLU, resulting in the high contributions of LMW PAHs [37, 39].

Source Identifications and Contributions

The ratios of ANT/(ANT+PHE) and BaA/(BaA+CHR) have been proposed as an indicator of petrogenic or pyrogenic sources [44]. The ratio of ANT/(ANT+PHE) < 0.10 is identified as petrogenic emissions, whereas the value > 0.10 is characteristic of combustion (pyrogenic emissions) [45]. The ratio of BaA/(BaA+CHR) < 0.20 is identified as petrogenic emissions, whereas a value > 0.35 is characteristic of combustion (pyrogenic emissions) [46]. The ratios of ANT/(ANT+PHE) and BaA/(BaA+CHR) in soils around the LG and XY power plants both indicated that the PAHs derived from combustion (Fig. 5). Coal firing is the primary energy source for thermal power plants [47]. It is consistent with the fact that the main PAH contribution in soils around the thermal power plants in

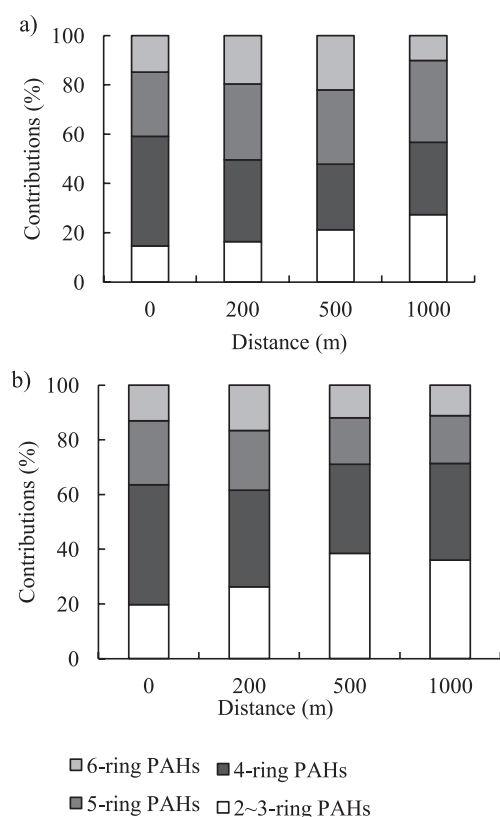


Fig. 4. Variation of contributions of PAHs (%) with distance in soils around the LG a) and XY b) power plants.

this work was due to the abundance of coal combustion and diesel combustion. Similar results were found in other areas in China. The ratios of $ANT/(ANT+PHE)$ and $FLA/(FLA+PYR)$ indicated that the main sources of soil PAHs in Beijing and Tianjin were biomass burning; in Dalian, the main source was petroleum combustion [48–49]. However, several points on Fig. 5 are relatively decentralized, which indicates that except for coal combustion, some other sources also were conducive to PAH contamination in soils from the study area [18].

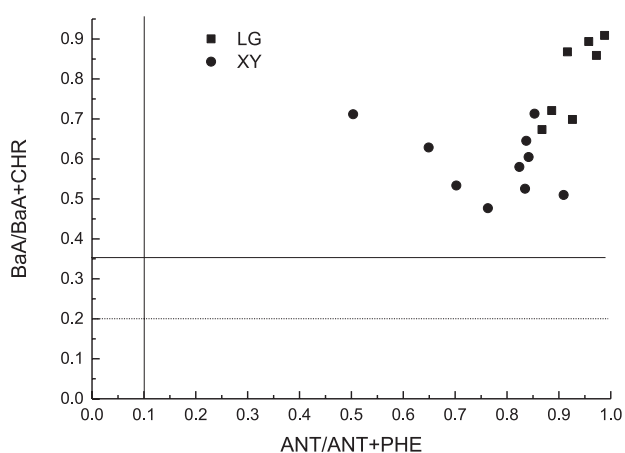


Fig. 5. Molecular diagnostic ratios of PAHs in soil in the LG and XY power plants.

The PMF model provided the source profiles and the contribution of each factor [50], are presented in Fig. 6 to distinguish the sources of PAHs in soils around the LG and XY power plants.

In soils around of the LG power plant, factor 1 characterized by NAP, ACE, BaP, ANT, and FLA contributed 23.2% to total PAHs. Incomplete combustion could generate Nap, and Nap could serve as an indicator of petroleum sources [51]. BaP, ANT, and FLA indicate the combustion of coal [52]. Therefore, Factor 1 is labeled as an incomplete coal combustion source. Factor 2, accounting 33.3% of the total PAHs, was mainly associated with IPY, FLU, CHR, and DBA. These compounds indicate fuel combustion [51], and diesel emissions are characterized by CHR and DBA [53–54]. Therefore, Factor 2 could be denoted as a traffic source factor. Factor 3, accounting for 21.1% of total PAHs, has high loadings of PHE, BaP, ANT, and FLU. Coal combustion could be traced by these compounds [55]. PHE is typically derived from coal or coal combustion [54]. ANT and FLU are predominant in coal combustion signals [56]. Therefore, Factor 3 is assigned as a coal source factor. Factor 4, accounting for 22.4% of total PAHs, has high loadings of BPE, CHR, PYR, and BkF. BPE is related to gasoline emissions [52]. CHR, BkF, and PYR were identified as diesel emissions [57]. Therefore, Factor 4 indicates a gasoline and diesel emissions source. Therefore, the primary source of PAHs in soil samples around the LG power plant is coal combustion.

In soil samples around the XY power plant, Factor 1, which contributed 23.0% of the total PAHs, was weighted predominately by DBA, FLU, BPE, and CHR. This characteristic is due to vehicle emissions generating high concentrations of CHR, BaP, and HMW PAHs, such as BPE and BkF [55]. Factor 1 could be denoted as a vehicle emissions factor. Factor 2, explaining 25.7% of the total PAHs, had high loadings of BaP, FLU, ACE, NAP, and IPY. LMW PAHs are generally delivered from petroleum and its products, as well as purified oil products [58–59]. Fossil fuel combustion and coking would produce HMW PAHs [46]. Factor 2 was both highly loaded with LMW and HMW PAHs, which is consistent with coal burning as the primary source of the mixed source pollution. Factor 3 contributed 16.6% of the total PAHs and has high loading of PYR, BkF, BaP, ANT, and BbF. Fossil fuel combustion could produce PYR [60]. BaP and BbF were the main contributors for this factor, and both of them are makers of diesel combustion [52, 61]. Factor 3 was chosen to represent a pyrogenic source. Factor 4, accounting 34.7% of the total PAHs, was highly loaded with BkF, BaA, ANT, IPY, PHE, and DBA. BkF, BaA, and ANT were related to the coal combustion source [54]. Therefore, Factor 4 was assigned as a coal source factor.

The main source of PAHs in soil samples around the two power plants is coal combustion, which produces heat to make the water evaporate and the steam turbine rotate to produce electrical power in coal-fired power

plants [62]. This process could explain why the coal combustion source is the main source in the LG and XY power plants.

The LG power plant is located in the center of town next to a fair. Thus, the traffic in this area is heavy. A transportation center is situated in the northeastern

portion of the XY power plant. The frequent transportation activities could explain the traffic source, accounting for the large loading. The boilers of thermal power plants generally use diesel for ignition, and power plants are equipped with a diesel oil tank, which may result in the spilling of diesel [63].

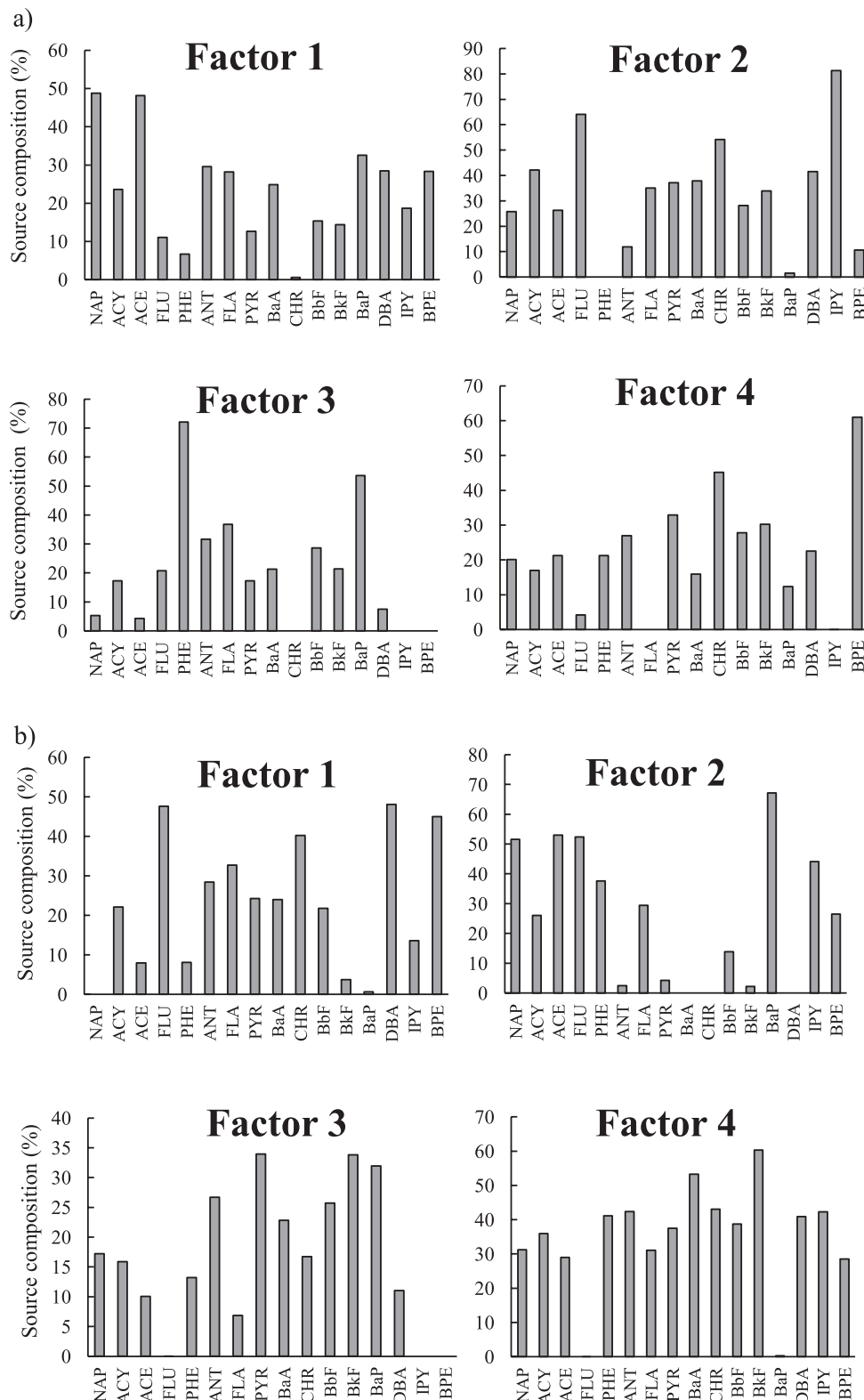


Fig. 6. Source profiles of PAHs in soil around the LG (a) and XY (b) power plants obtained with the PMF model.

Health-Risk Assessment

In soil samples around the LG power plant, BaP_{eq} concentrations of the 16 PAHs ranged from 164.6 µg/kg to 268.3 µg/kg, with a mean value of 211.9 µg/kg (Table 3). In soil samples around the XY thermal power plant, BaP_{eq} concentrations of the 16 PAHs ranged from 347.5 µg/kg to 756.2 µg/kg, with a mean value of 563.9 µg/kg. The BaP_{eq} concentrations in this study are higher than those in the urban and suburban areas of Nanjing (445 and 215 µg/kg) [9], the industrial district in Jinan (262 µg/kg), urban areas of Shanghai (428 µg/kg), and the farmland soil of Shouguang (23.1 µg/kg) [64-66]. By contrast, the BaP_{eq} concentrations in the study areas are lower than those in the steel mill area in Fujian Province (852 µg/kg) [67].

The Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health indicates that the safe value of BaP_{eq} in soil is 600 µg/kg [68]. The results showed that all concentrations of the samples from around the LG power plant were below the safe value, and 16.7% of the samples around the XY power plant showed concentrations above the safe value. Therefore, no risk of PAHs was found in soils around the LG power plant, and a low risk of PAHs was observed in soils around the XY power plant. The generator sets of the XY thermal power plant (30 MW) were bigger than those of the LG thermal power plant (12 MW). Therefore,

Table 3. Toxic equivalent concentration (BaP_{eq}) (µg/kg) of PAHs in soils from study areas.

PAHs	TEFs	BaP _{eq}	
		LG	XY
NAP	0.001	0.04±0.02	0.03±0.01
ACY	0.001	0.07±0.01	0.07±0.01
ACE	0.001	0.03±0.01	0.03±0.01
FLU	0.001	0.01±0.00	0.06±0.05
PHE	0.001	0.01±0.01	0.05±0.01
ANT	0.01	1.22±0.08	2.73±1.26
FLA	0.001	0.19±0.12	0.22±0.06
PYR	0.001	0.30±0.07	0.18±0.10
BaA	0.1	26.93±18.24	43.69±7.81
CHR	0.01	0.01±0.01	0.30±0.11
BbF	0.1	4.47±2.28	5.57±0.85
BkF	0.1	31.58±10.06	5.72±1.68
BaP	1	21.63±21.42	32.32±27.69
DBA	1	121.80±10.85	460.70±116.48
BPE	0.01	2.80±1.98	2.41±1.90
IPY	0.1	0.71±0.18	9.86±2.80
∑ ₁₆ PAHs		211.9±43.3	563.9±130.5

a large coal-fired power plant would cause serious pollution of PAHs. Human health would be threatened by contaminated soil, and thus close attention should be paid to soil and human health.

Conclusions

The PAHs in the surface soil samples around the two thermal power plants were heavily contaminated. The four-ring PAHs dominated all samples around the two power plants. With a gradual increase in distance, most individual PAH concentrations in soil samples decreased, and the total PAH concentrations decreased significantly from 200 m to 500 m. The contributions of the two- to three-ring PAHs increased with distance, whereas the contributions of the four-ring PAHs decreased. The ratios ANT/(ANT+PHE) and BaA/(BaA+CHR) showed that these PAHs were emitted from combustion. The sources of PAHs in the two thermal power plants, as obtained with the PMF model, were coal combustion, traffic, diesel emission, and mixed sources; coal combustion was the main source. Risk assessment showed no risk of PAHs in soils around the LG power plant, but a low risk of PAHs in soils around the XY power plant.

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Conflict of Interest

The authors declare no conflict of interest.

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