

*Original Research*

# Distributions of Polycyclic Aromatic Hydrocarbons in Coal in China

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## Abstract

Groundwater level rises rapidly when mine drainage systems stop functioning after mine closures. Free-form polycyclic aromatic hydrocarbons (PAHs) in residual pillaring and abandoned mining levels could continue to migrate because of eluviation leaching. Moreover, other aquifers are polluted with mine water through mining-induced fractures, faults, and poorly sealed drill holes. Therefore, the distributions of 16-PAHs in raw coal mined in China and the factors influencing these distributions were analyzed to assist mine closures. The results showed that the average concentration of PAHs was  $10.540 \pm 7.973$   $\mu\text{g/g}$  in the raw coal samples, and PAHs with low molecular weights had the highest abundances, accounting for 44% of the total PAH concentration obtained. The highest concentration of 16-PAHs was observed in bituminous coals, followed by that in lignite, and the lowest is anthracite. The influence factors analysis reveals that carbon content, volatile matter, H/C, and O/C have a significant effect on PAH content in raw coals. The volatile matter and molar ratio of H/C play a leading role in the changing process in 16 PAHs, accounting for more than 60% of the total contribution.

**Keywords:** raw coal, polycyclic aromatic hydrocarbons, distribution, influence factor, degree of contribution

## Introduction

As China's main energy source, coal production and consumption account for 72% and 62%, respectively, of primary energy. The total output of raw coal was 3.41 billion tonnes (t) in 2016, down 9.0% from the same period a year earlier, and about 90% of coal comes from underground mines. In recent years, many coal mines have been closed because of the depletion of coal resources, resource integration, and coal mine

production safety. Consequently, groundwater levels in these mines rise rapidly after the mine drainage systems stop working. Moreover, other aquifers are polluted with mine water through mining-induced fractures, faults, and poorly sealed drill holes. In northern China, the Ordovician or Cambrian aquifer, which is the main coal seam floor, is an important water source [1]. This water resource is at risk of being polluted when the groundwater above becomes polluted. For example, due to the influence of the coal mine water in Zibo mining area of Shandong Province, and Yangquan mining area in Shanxi Province, the karst groundwater has been polluted in different degrees [2-3]. Investigations show that around 40% of total groundwater in northern China

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has been being affected by different mining activities [1]. Groundwater pollution in the abandoned mining areas, i.e., heavy metals [4], inorganic pollution [5], and organic pollution [6], is affected by chemical, biological, and physical factors [7]. Therefore, Polycyclic aromatic hydrocarbons (PAHs) are among the most common and typical organic pollutants and thus receive substantial attention from fields that investigate environmental pollution in mine areas.

A persistent organic pollutant, PAH is a hydrocarbon composed of two or more aromatic rings. At the start of the last century, PAH was initially a widespread concern because it causes skin cancer in coke production workers [8]. Industrial processes such as pyrogenesis or combustion of fossil fuels (coal and oil) or biomass, and oil spills are the main sources of PAHs. Moreover, natural factors such as forest fires and volcanic activities can also produce PAHs [9-11]. The release of PAHs during pyrolysis is greatly dependent on pyrolysis temperature, gas/coal ratio, and coal quality [12]. Most PAHs generated by human activities are discharged in gaseous form, but eventually turn into solid form or attach to particulate matter. PAHs not only pollute water and soil by precipitation and settlement but also threaten

human health through the food chain [13-15]. Moreover, most PAHs can cause cancer, abnormality, and mutation, and thus organisms exposed to PAHs for a long time are endangered [16-18]. A survey result indicated that lung cancer mortality increases by 5% in areas where benzo [a] pyrene concentration increases to  $0.001 \mu\text{g}/\text{m}^3$  [19]. At present, research on PAHs has been mainly focused on the following three areas: (i) the sources and distributions of PAHs in the environment, i.e., especially in the atmosphere [20-23]; (ii) the geochemical processes of PAHs in soils and sediments, including degradation, adsorption, and migration [24-27]; and (iii) the toxicity and risk assessment of PAHs in different environments [28-30].

Raw coals are generated from plant materials through a series of coalification under certain temperatures and pressures. In the *Chinese Classification of Coals* (GB/T 5751-2009), the raw coals are divided according to degree of coalification into lignite, bituminous, and anthracite coal, out of which the coalification degree of lignite is lowest, and of anthracite the highest. The main structural unit of coal is aromatic. The aromatic compounds in coal exist in two forms, namely macromolecular aromatic compounds, which



Fig. 1. Location of the sample sites in China.

Table 1. Industrial analysis and elemental analysis of raw coal.

Number	Coal Mine	Industrial Analysis (%)				Elemental Analysis (%)				
		M <sub>ad</sub>	A <sub>d</sub>	V <sub>daf</sub>	FC <sub>d</sub>	O	C	H	N	P
C1	Zhun Dong <sup>b</sup>	13.32	9.48	29.68	63.65	15.17	80.31	3.42	0.68	0.00
C2	Ha Mi <sup>a</sup>	16.58	9.09	40.69	53.92	20.28	75.00	3.61	0.82	0.00
C3	Zha Lainuoer <sup>a</sup>	17.44	19.21	43.18	45.91	22.73	72.68	3.30	0.99	0.01
C4	Bai Yinhua <sup>a</sup>	9.90	12.32	45.04	48.19	20.61	71.90	4.89	1.58	0.00
C5	Shi Getai <sup>b</sup>	10.62	5.42	33.85	62.56	14.23	80.41	4.19	0.90	0.00
C6	Na Linmiao <sup>b</sup>	8.40	11.67	35.72	56.78	14.92	79.67	4.07	1.00	0.01
C7	Chang Sheng <sup>b</sup>	3.56	8.09	36.46	58.40	14.51	79.35	4.87	1.04	0.02
C8	Ding Jiaqu <sup>b</sup>	2.86	5.04	35.32	61.42	13.45	80.65	4.49	1.04	0.00
C9	Ha Lagou <sup>b</sup>	5.78	4.49	34.36	62.69	13.63	80.41	4.47	0.84	0.00
C10	Hai Hong <sup>b</sup>	4.55	4.32	32.55	64.54	12.66	81.56	4.62	1.00	0.00
C11	Shang Wan <sup>b</sup>	1.42	36.99	32.95	42.25	14.78	78.61	4.99	0.79	0.02
C12	Zhang Gou <sup>c</sup>	0.73	12.33	8.12	80.55	3.86	91.21	3.38	1.25	0.15
C13	Long Gu <sup>b</sup>	1.06	17.10	35.84	53.18	7.78	84.39	5.32	1.63	0.01
C14	Dong Tan <sup>b</sup>	2.36	9.48	38.00	56.12	10.66	82.15	5.17	1.57	0.01
C15	Liu He <sup>b</sup>	0.85	12.24	12.40	76.87	3.87	90.47	3.85	1.21	0.01
C16	Qi Shan <sup>b</sup>	2.14	30.17	40.12	41.81	12.76	79.51	5.36	1.79	0.01
C17	Qi Shan <sup>*b</sup>	1.98	31.64	35.79	43.90	11.98	80.79	5.31	1.40	0.01
C18	Quan Tai <sup>b</sup>	2.96	16.18	40.32	50.02	12.91	78.49	5.14	1.67	0.01
C19	Ding Ji <sup>a</sup>	1.24	38.02	40.65	36.79	13.90	78.71	5.52	1.40	0.01
C20	Zhang Shuanglou <sup>b</sup>	2.21	19.65	36.01	51.42	10.07	82.59	4.99	1.43	0.01
C21	Kong Zhuang <sup>b</sup>	2.58	20.39	36.36	50.67	8.26	83.42	5.11	1.43	0.01
C22	Zhang Ji <sup>b</sup>	1.70	30.07	39.18	42.53	12.66	80.36	5.02	1.34	0.02
C23	Ke Lang <sup>a</sup>	9.97	15.46	56.34	33.23	20.31	68.89	5.16	1.59	0.01
C24	Gong Qing <sup>b</sup>	0.69	10.85	22.64	68.49	3.40	89.90	4.77	1.68	0.01
C25	Gong Qing <sup>*b</sup>	0.75	19.81	20.48	63.29	3.22	89.55	4.59	1.71	0.02
C26	Xing Ying <sup>b</sup>	0.62	14.04	19.96	68.38	4.19	89.25	4.46	1.64	0.02
C27	Mao Ergou <sup>c</sup>	1.20	29.24	12.66	61.06	2.94	88.39	3.75	0.92	0.01
C28	Chang Sheng <sup>c</sup>	0.61	4.71	5.67	89.34	1.27	94.09	3.04	0.77	0.01
C29	Xu Jiayuan <sup>c</sup>	0.47	9.47	7.00	83.79	1.15	93.02	3.30	0.85	0.00

<sup>a</sup> Lignite<sup>b</sup> Bituminous Coal<sup>c</sup> Anthracite

\* Sample collected from different coal seams at the same coal mine

are interconnected by aliphatic hydrocarbons, and low-molecular-weight PAHs that exist as solids and liquids in coal [31]. PAH concentrations are affected by several factors, such as carbon content, volatile matter, H/C mole ratio, and O/C mole ratio, but the degree of their influence cannot be determined in the present study [31-32]. Furthermore, current research on PAHs are mainly

focused on the release of PAHs during combustion and pyrolysis of fossil fuels and thus knowledge on the occurrence and migration PAHs and environmental hazards from free PAHs in raw coal remains limited [33-34]. As groundwater levels rise quickly after the mine drainage systems of closed mines stop their operations, the free PAHs in the protective coal pillars

and remaining coal seams of these mines are released into the groundwater, thereby polluting aquifers with mine water through mining-induced fractures, faults, and poorly sealed drill holes. Therefore, in order to guide mine closures, we investigated the levels and distributions of PAHs in coal mines in China. We then identified the correlation between PAHs and the physical and chemical properties of coal and analyzed the extent of contribution of different influencing factors on PAH content.

## Materials and Methods

### Study Area and Sampling

China has 14 coal bases. The most notable of these bases are located in the Shendong region, southwestern region of Shandong, Huaibei-Huainan region, and East Inner Mongolia-Northeast region. The main coal-forming periods are Carboniferous, Permian, Jurassic, Cretaceous, and Tertiary. In this study, a total of 29 samples were collected from different coal bases. The sampling distribution is shown in Fig. 1. In each sample site, approximately 1.5 kg coal sample was collected and placed into a sealed bag and then brought to the laboratory. A few of the collected samples were stored in brown glass bottle at 4°C and used for PAH detection in the University of Science and Technology of China, and the remaining part was used for coal analysis in the Jiangsu Institute of Geology and Mineral Resources. All brown glass bottles were pre-combusted at 450°C for 4 h in order to remove the organic matter.

### Coal Analysis

Coal analysis of coal samples were performed on the basis of Proximate Analysis of Coal (GB/T212-2008) in the Jiangsu Institute of Geology and Mineral Resources. Table 1 shows the coal analysis data of 29 mines, including 5 lignite, 20 bituminous coal, and 4 anthracite. The results indicated that ash concentrations in the analyzed coal samples fall in the range of 4.32–38.02% with a mean of 16.10±9.93%. The volatile content of raw coal varied from 5.67% to 56.34% with a mean of 31.29±12.51%. The mole ratio of H/C and O/C varied from 0.388 to 0.899 and from 0.009 to 0.235, respectively, among all the samples.

### PAH Analysis

For the analysis of polycyclic aromatic hydrocarbon content in coal, samples were pretreated through the microwave-assisted solvent extraction method (MASE) described by Yuan [35]. NaP-d<sub>8</sub>, Chry-d<sub>12</sub>, and Ph-d<sub>10</sub> were added to 20 g of coal samples. Dichloromethane (240 mL) was then added to the mixture. The crude extract, collected by Soxhlet extraction, was concentrated to about 2 mL in a rotary

evaporator. After adding 10 mL of exchange solvent (n-hexane), the mixed decoction was concentrated to about 2 mL. The polycyclic aromatic hydrocarbons in the extract were purified with an adsorption column containing silica and alumina gel (2:1) as adsorbents. Subsequently, the PAHs were leached from the column with 70 mL of hexane and dichloromethane (7:3) at a flow rate of 1 mL/min. The eluant was concentrated to 1 mL, and then introduced into thermo trace ultra gas chromatography coupled with a Thermo DSQ II mass spectrometer (GC-MS) for PAH analysis. A 60 m x 0.25 mm (L x ID) DB5 capillary column with a 0.25 µm-thick film was used. Helium was used as carrier gas at a flow rate of 1 mL/min. The injected sample volume was 1 µL in splitless injection mode. The initial temperature of the capillary column was set at 50°C for 2 min. The dynamic temperature program used was as follows: 60–200°C at 5°C/min, 200–250°C at 2°C/min, 250–290°C at 20°C/min, and then held at 290°C for 20 min. The electron impact mode was 70 eV, and the mass spectra were recorded with selected ion-monitoring modes. Individual PAH concentrations in coal samples were calculated from a six-point calibration curve of standard solutions.

### Quality Control of Test Results

The quality assurance and quality control procedures for the sample analysis were recommended by the United States Environmental Protection Agency (EPA). Method blanks, spiked blanks, and sample duplicates were performed during analysis. In the method blank samples there are no detections of the PAHs. The rate of recovery of 16-PAHs in NIST reference samples (National Institute of Standards and Technology, 1941) ranged from 76% to 105%. Furthermore, individual PAH recoveries evaluated by standard solutions of NaP-d<sub>8</sub>, Chry-d<sub>12</sub>, and Ph-d<sub>10</sub> were 81.5%±8.3%, 88.5%±9.6%, and 91.7%±9.0%, respectively. The detection limit of the instrument for PAH analysis ranged from 0.04 ng/g to 0.51 ng/g.

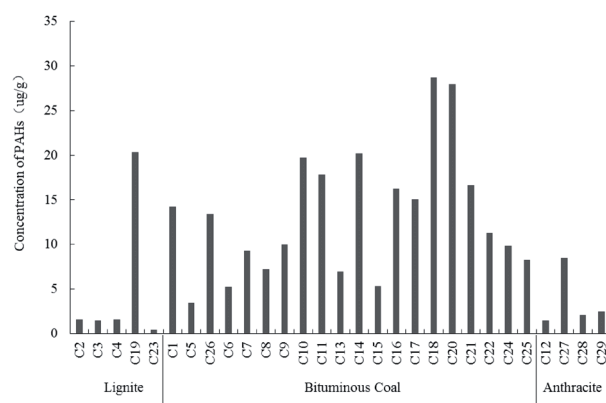


Fig. 2. Concentrations of 16-PAHs in coals from various coal mines in China.

Table 2. PAH concentrations ( $\mu\text{g/g}$ ) in different samples of raw coal.

PAH	Lignite (n = 5)				Bituminous Coal (n = 20)				Anthracite (n = 4)			
	Min.	Max.	Mean	SD	Min.	Max.	Mean	SD	Min.	Max.	Mean	SD
Nap	0.046	3.675	0.803	1.606	0.043	4.962	1.286	1.407	0.175	0.970	0.431	0.363
Acy	ND <sup>a</sup>	0.027	0.023	0.005	ND	0.708	0.103	0.182	0.000	0.013	0.009	0.006
Ace	0.009	0.578	0.138	0.247	0.022	2.326	0.471	0.611	0.036	0.202	0.089	0.078
Fle	0.031	3.535	0.739	1.563	0.077	4.501	1.271	1.489	0.131	0.483	0.240	0.166
Phe	0.061	4.136	0.901	1.809	0.100	6.244	2.285	1.973	0.417	0.696	0.589	0.120
Anth	0.018	0.499	0.126	0.209	ND	1.380	0.574	0.424	0.018	0.041	0.026	0.011
Fla	0.039	0.976	0.286	0.391	0.106	2.622	0.951	0.772	0.030	0.287	0.107	0.120
Pyr	0.043	2.048	0.485	0.875	ND	2.627	0.852	0.753	0.042	0.234	0.109	0.085
BaA	0.021	1.537	0.449	0.625	ND	3.661	1.081	0.995	0.022	0.069	0.035	0.023
Chr	0.021	1.292	0.356	0.539	0.347	2.570	0.959	0.540	0.066	1.117	0.445	0.467
BbF	0.012	0.193	0.088	0.073	ND	4.026	0.978	1.117	0.029	1.691	0.591	0.747
BkF	0.010	0.866	0.247	0.352	0.023	6.598	0.960	1.818	0.020	0.038	0.025	0.008
BaP	ND	0.769	0.264	0.342	ND	1.829	0.487	0.516	0.015	0.086	0.036	0.033
Ind	ND	0.412	0.200	0.184	ND	2.822	0.571	0.807	0.017	0.085	0.046	0.029
DaA	ND	0.203	0.093	0.089	ND	2.877	0.439	0.636	0.015	1.357	0.379	0.653
BgP	ND	0.037	0.020	0.012	ND	3.942	0.702	0.952	0.035	1.307	0.409	0.601
$\Sigma\text{PAHs}$	0.388	20.324	5.053	8.551	3.402	28.664	13.307	7.100	1.403	8.397	3.567	3.249

<sup>a</sup> ND is not detection

## Results and Discussion

### Levels of PAHs in Coal Mines

The concentrations of the 16 PAHs – including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fle), phenanthrene (Phe), anthracene (Anth), fluoranthene (Fla), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF),

benzo[a]pyrene (BaP), indeno[1,2,3-c,d]pyrene (Ind), dibenz[a,h]anthracene (DaA), and benzo[g,h,i]perylene (BgP) – in the coal samples are shown in Fig. 2, and the heights of the bars represent PAH content in the corresponding sample point. As shown in the figure, from lignite to anthracite, with the increase of coalification degree, the total PAH content (2-6 rings) changed along a quadratic curve. The total PAH concentrations in the coal ranged from 0.388  $\mu\text{g/g}$  (C23, Kelang Coal Mine) to 28.664  $\mu\text{g/g}$  (C18, Quantai Coal Mine) with a mean of  $10.540 \pm 7.973 \mu\text{g/g}$ . These obtained concentrations were consistent with those of naturally formed PAHs in the complex coal-forming process [36].

The monitoring results of PAHs in various coals is presented in Table 2. The concentrations of the 16 PAHs in the lignite, bituminous coal, and anthracite were  $5.053 \pm 8.551$ ,  $13.307 \pm 7.100$ , and  $3.567 \pm 3.249 \mu\text{g/g}$ , respectively. The highest PAH concentration was observed in the bituminous coal. The observed distribution characteristics are attributed to the complex coal metamorphism during coal formation and monitored PAH types. Organic matter in lignite was dominated by chain hydrocarbon, and PAH concentrations in lignite were low. Meanwhile, at intensified coalification, some aliphatic hydrocarbons in raw coal were transformed into aromatic compound through dehydrogenation and cyclization. The concentrations and kinds of

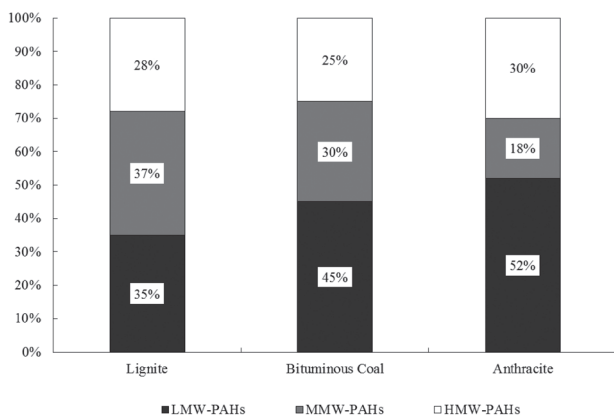


Fig. 3. Occurrence characteristics of PAHs in different coals.

Table 3. Occurrence characteristics of PAHs in coal collected from other mines in the world.

Country	Coal	Samples	$\Sigma$ PAHs	LMW-PAHs (%)	MMW-PAHs (%)	HMW-PAHs (%)	Reference
India	Butumious Coal	1	4.542	7.3	28.3	64.4	Verma et al. [32]
United States	Lignite	7	1.049	17.8	59.0	23.2	Stout et al. [36]
	Butumious Coal	6	5.660	74.6	19.4	6.0	
	Anthracite	2	1.064	63.4	19.7	16.9	
United States	Butumious Coal	3	17.773	29.8	34.8	35.4	Wang et al. [39]
	Anthracite	3	3.257	58.8	25.8	15.4	

PAHs in the bituminous coal increased continuously. However, concentrations of the 16 PAHs in anthracite decreased because of the reduction of extractable PAHs.

In order to assess the distribution of PAH homologue in different coal types, the 16-PAHs are divided into three groups according to their molecular weight, i.e. low-molecular-weight PAHs (LMW-PAHs: Nap, Acy, Ace, Fle, Phe, and Anth), medium-molecular-weight PAHs (MMW-PAHs: Fla, Pyr, BaA, and Chr), and high-molecular-weight PAHs (HMW-PAHs: BbF, BkF, BaP, Ind, DaA, and BgP) [37]. The average concentrations of LMW-PAHs (2-3 ring PAHs) were highest, which constituted 44% of the 16-PAH content, followed by MMW-PAHs (29%) and then HMW-PAHs (27%). Fig. 3 shows the occurrence characteristics of different rings of PAHs in raw coal. The results showed that the concentration of LMW-PAHs in lignite, bituminous coal, and anthracite were 35%, 45%, and 52%, respectively. In contrast, with the increase of coalification the percentage of MMW-PAHs decreases from 37% to 18%. PAH concentrations in raw coal in other countries are listed in Table 3. To compare with previous research, similar patterns were also found in coal in India and the United States, although the ratio is different. This may have been caused by the different coal origins of different countries [38].

### Regional Distribution of PAHs in Chinese Coal

In China, the regional distribution of PAHs showed distinct regional characteristics (Fig. 4). On the basis of different coal-forming eras, all coal mines were divided into three regions. As shown in the picture, coal distribution had significant regional characteristics, and the concentrations of 16 PAHs varied among different coal mining areas. The concentrations of the 16 PAHs in raw coal from China's eastern mining areas were significantly higher than those from other mining areas. The raw coal from Quantai Coal Mine (C18) contained the highest PAH concentration (28.66  $\mu\text{g/g}$ ), followed by Zhangshuanglou Coal Mine (C20). The enrichment of PAHs in China's eastern mining may be associated with the geological environment and the evolvement process of coal. The data analysis results revealed that most coal resources in the eastern coal mining areas were comprised of bituminous coal, therefore the concentrations of 16 PAHs were relatively high. By contrast, the major coal in northern and southern mining areas of China is lignite and anthracite, and the concentrations of PAHs in raw coals were low.

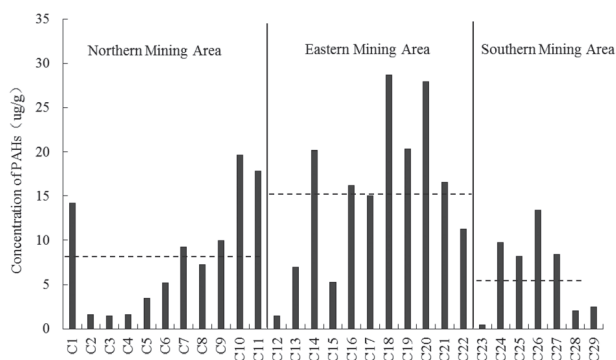


Fig. 4. Regional distribution of PAHs in Chinese raw coal.

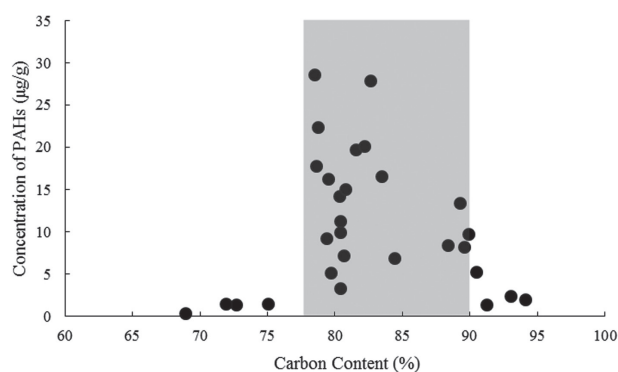


Fig. 5. Relationship between 16-PAHs and carbon content.

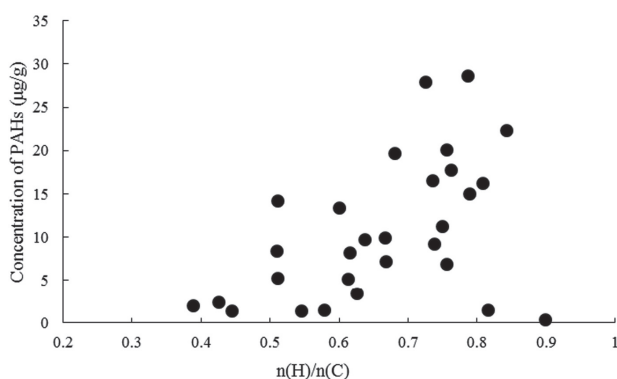


Fig. 6. Relationship between 16-PAHs and H/C molar ratio.

### Influencing Factors of PAH Distribution

It has been reported that the concentrations of 16 PAHs were influenced by the physicochemical properties of raw coal, including carbon content, volatile matter, H/C, and O/C [39-40]. The relationship between the 16 PAHs in the coal samples and carbon content is shown in Fig. 5. The result showed that PAH concentration increased gradually first and then sharply when carbon content increased from 70% to 82%. This increase may be attributed to coalification in which some PAHs formed through condensation reactions of

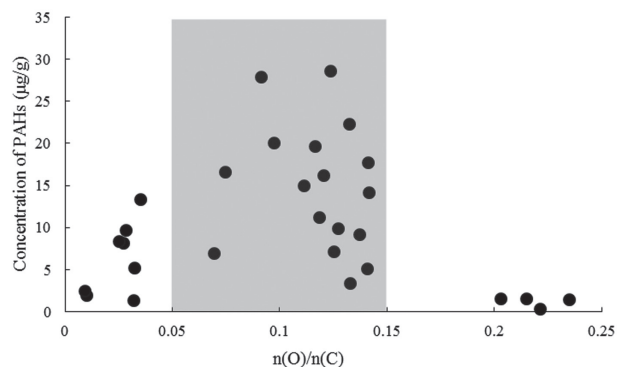


Fig. 7. Relationship between 16-PAHs and O/C molar ratio.

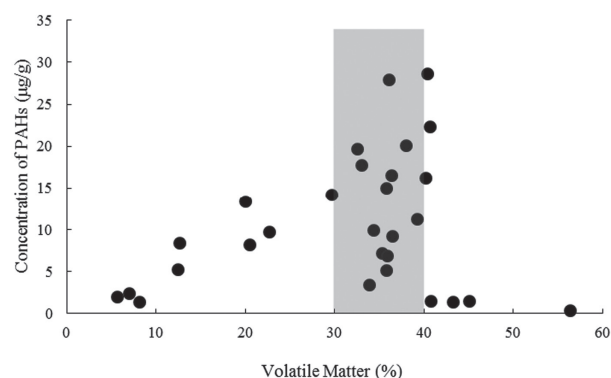


Fig. 8. Relationship between 16-PAHs and volatile component.

aliphatic hydrocarbons containing oxygen and sulfur groups. Therefore, the concentration and ring number of PAHs increased at increased carbon content in the course of coalification. After the carbon content reached 84%, some PAHs with high molecular weights became difficult to extract in the experiment, thus decreasing the concentrations of 16 PAHs. The amounts of the 16 PAHs extracted from the raw coals considerably increased at carbon contents ranging from 78% to 90%.

The molar ratio of H/C is one of the most important parameters of coalification degree. On the basis of the structural changes in the organic matter of coal, a low H/C ratio indicates a high degree of cyclization in coal structure, and a substantial amount of PAH is detected in raw coal. As shown in Fig. 6, the concentrations of the PAHs were associated with the molar ratio of H/C. At decreased molar ratio of H/C, the concentrations of the 16 PAHs increased slowly at the beginning, and then rapidly increased, and finally decreased after reaching a peak value. A substantial amount of PAHs were extracted from the raw coal samples with H/C molar ratios of around 0.75. This result was observed in the coal samples from Zhangshuanglou Coal Mine (C20) and Quantai Coal Mine (C18). In contrast to the trend in Fig. 5, the trend in the changes in the H/C molar ratios of the 16 PAHs were similar to the laws of PAHs with carbon content.

Similarly, a substantial amount of PAH was detected at O/C molar ratio of 0.05-0.15, and the maximum value was obtained at O/C molar ratio of around 0.11. The element oxygen usually exists in aliphatic hydrocarbons, and the higher the O/C molar ratio, the lower the amount of aromatic compounds in raw coal. During the evolution of the coal, the oxygen was constantly separated through oxidative dehydrogenation. Therefore, at a low O/C molar ratio, the aromatic compounds are the main compositions of organic compounds in the coal samples.

The 16-PAHs are part of a volatile component of raw coal, and the relationships between them are inevitably produced (Fig. 8). The volatile component in raw coal, referred to volatile organic matter and readily decomposable mineral, is an important indicator of the degree of coalification. As shown in Fig. 8, PAH concentrations are high at volatile component of

Table 4. Relative contribution of different factors on PAH content in coal.

Influence Factor	A	B	C = A*B	Relative Contribution (%)
Carbon content	-0.752	-0.052	0.039	10.77
Volatile matter	0.490	0.229	0.112	30.94
H/C	0.242	0.460	0.111	30.66
O/C	-1.351	-0.074	0.100	27.63

30-40%. A high volatile component content indicates the presence of more aliphatic hydrocarbons in raw coal (Lignite). In contrast, most organic compounds exist as macromolecules in anthracite with low content of volatile component and thus are difficult to extract.

#### Analysis of Contributions of Different Influencing Factors

The above analysis shows that carbon content, volatile matter, H/C, and O/C are related to the content of 16-PAHs in raw coal. The relative contribution of the four factors to PAH content was analyzed through convergence analysis performed in Eviews 6.0 [41]. As illustrated in Table 4, parameter A refers to multiple linear regression standard coefficients under different influence factors, and parameter B is linear regression standard coefficients among different factors and among the 16 PAHs. The relative contribution is the ratio of single influence (parameter C), the product of parameter A and parameter B, to total influence. The calculated result indicated that the volatile matter and molar ratio of H/C had a positive effect on PAH concentrations in raw coal, and the influence is highly evident. The relative contribution of volatile matter was the highest, accounting for 30.94%, followed by H/C molar ratio, which accounts for 30.66%. Overall, the volatile matter and molar ratio of H/C plays a leading role in the changes in the PAHs, and the total proportion was over 60%. Our results were found to be consistent with the previous studies. Wang (2010) analyzed the effect of carbon content, H/C, and O/C on 16-PAHs content in Chinese and American coal, and the relationships were fitted by Gauss-fit. The result showed that the effect of H/C on PAHs content was clearly greater than carbon content and O/C [39]. This result may be due to the changes in the chemical structures of organic compounds during coal metamorphism and evolution process. Coal evolution is a continuous process of aliphatic hydrocarbon condensation. In this process, the volatile matter continues to decline during the separation of O elements, and the molar ratio of H/C continues to grow. Accordingly, the volatile matter and molar ratio of H/C can directly reflect the changes in PAHs in raw coal.

#### Conclusions

Twenty-nine coal samples were collected from different coal bases to analyze the distributions of polycyclic aromatic hydrocarbons in coal and the factors influencing these distributions. The total concentration of PAHs (mean value) was  $10.540 \pm 7.973$   $\mu\text{g/g}$ . In this concentration, LMW-PAHs had the highest concentrations, constituting 44% of average share in the PAHs. Overall, the highest concentrations of extractable PAHs were detected in bituminous coals, followed by the concentrations

in lignite. The lowest concentration was obtained in anthracite. The concentrations and kinds of PAHs in bituminous coal increased continuously. Furthermore, the influences of carbon content, volatile matter, H/C, and O/C on PAH content in raw coals are significant. The volatile matter and molar ratio of H/C play a leading role in the changing process in the PAHs, accounting for more than 60% of the total contribution, followed by the molar ratio of O/C and carbon content.

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#### Conflict of Interest

The authors declare no conflict of interest.

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