Distribution of Polycyclic Aromatic Hydrocarbons in Coal Gangue with Different Metamorphic Degrees

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Abstract. The contents of 16 US Environmental Protection Agency (EPA) polycyclic aromatic hydrocarbons (PAHs) in coal gangue samples with different metamorphic degrees (R₀,max=1.21%, 1.47%, 1.84%, 2.05%, 3.30%) have been measured by high performance liquid chromatography (HPLC), and the characteristic distributions of PAHs in samples are studied. The results suggest that all the contents of high molecular weight polycyclic aromatic hydrocarbons (HPAHs, 4-6ring), low molecular weight polycyclic aromatic hydrocarbons (LPAHs, 2-3ring), total polycyclic aromatic hydrocarbons and the toxicity of PAHs in samples increase first and then decrease as the increase of vitrinite reflectance. The proportion of LPAHs in the ∑16PAHs decreases first and then increases, while HPAHs is opposite. Furthermore, Ant/(Ant+Phe) ranges from 0 to 0.03, Flua/(Flua+Pyr) ranges from 0 to 0.37, BaA/(BaA+Chr) ranges from 0.04 to 0.14, and total index range from 0.64 to 1.66 in samples, all of which could provide a reference for judging the source of PAHs.

1. Introduction
Polycyclic aromatic hydrocarbons (PAHs) are organic compounds composed of two or more benzene rings which contain only two elements, carbon and hydrogen, and have characterized as non-polarity, hydrophobicity and refractory degradation [1]. Simultaneously, PAHs also have the characteristics of "carcinogenic, teratogenic and genetic mutations" [2][2]. In addition, PAHs have adverse impact to human body by destroying hematopoietic and lymphatic systems, degrading the spleen, thymus and diaphragm lymph nodes, and inhibiting bone growth, when people contact with water, air, food, daily necessities polluted by PAHs [6]. The US Environmental Protection Agency (EPA) has included 16 PAHs in the blacklist of priority controlled toxic organic pollutants, including: naphthalene (Nap), acenaphthene (Ace), acenaphthylene (Acpy), fluorine (Flu), phenanthrene (Pha), anthracene (AnT), fluoranthene (Flua), pyrene (Pyr), benzo (a) anthracene (BaA), chrysene (Chr), benzo [b] fluoranthene (BbF), benzo [k] fluoranthene (BkF), benzo (a) pyrene (BaP), dibenzo (a, h) anthracene (DbA), benzo (g, hi) perylene (BghiP), indeno (1, 2, 3-cd) pyrene, (In [1, 2, 3-cd] P) [7].
Coal gangue is the main solid waste residue in coal mining and processing [8]. Sun et al manifested that a coal gangue yard contains more than 10 types of carcinogenic PAHs, and its organic pollution has an impact on the environment within a range of at least 1800 meters [9]. Wang et al suggested that coal gangue contains various PAHs such as Nap, Pyr, Ace, FluA, Flu, Pha, AnT, BbF, and BaP [10].

PAHs can be divided into low-molecular-weight PAHs (LPAHs, 2-3 rings) and high-molecular-weight PAHs (HPAHs, 4-6 rings) [11]. The content of HPAHs and LPAHs is related to the maturity of coal [12]. Ruwei Wang et al considered that high molecular weight (HMW) PAHs dominate in low-grade coal, and low molecular weight (LMW) PAHs dominate in high-rank coal by studying 14 Chinese and American coals [13], which was consistent with the results of Stout's PAH model shifting from HPAHs in low-grade coal to LPAHs in high-rank coal [14].

Many scholars devoted themselves to studying the characteristic parameters of PAHs in pollutants. Yunker et al inferred the sources of PAHs in the Fraser Basin through the ratios of Ant/(Ant+Phe), Flua/(Flua+Pyr), BaA/(BaA+Chr), InP/(InP+BghiP), and evaluated the consistency of distribution of these sources and the applicability of various commonly used PAHs ratios as indicators [15]. H.H.Soclo studied the sources of PAHs in Cotonou (Benin) and Aquitaine (France) Areas by using six molecular indices Phe/An, Flt/Py, Chry/BaA, LMW/HMW, Per+/+PAH and Per+/+pentaresene) [16]. Maria Rosaria Mannino and Santino Orecchio proposed to distinguish the sources of PAHs by the total index, total index = [Flua/(Flua+Pyr)]/0.4 + [Ant/(Ant+Phe)]/0.1 + [BaA/(BaA+Chr)]/0.2 + [InP/(InP+BghiP)]/0.2 [17, 18]. Xueqin Wen et al studied the characteristic parameters of PAHs in low temperature spontaneous combustion samples and calculated the ratios of Ant/(Ant+Phe) (all equal to 1), Flua/(Flua+Pyr) (0.85-0.97), BaA/(BaA+Chr)(0.99-1), InP/(InP+BghiP) (all equal to 1) and total index (22.125-22.425) [19].

The purpose of this paper is to study the changes in the total amount of PAHs, HPAHs and LPAHs, and the relationship between toxic equivalent and metamorphic degree. Studying the eigenvalues of PAHs in coal gangues from different coal ranks plays an important role in preventing pollution, judging pollution sources and improving pollution control efficiency. Therefore, this article also studied the characteristic parameters of 16 PAHs in coal gangue with different metamorphic degree, and searched for the range of characteristic index.

2. Sampling area
There are five sampling location in Taiyuan City and Jincheng City, Shanxi Province. Four sampling location of all, Malan Coal Mine (ML), Ximing Coal Mine (XM), Tunlan Coal Mine (TL) and Duerping Coal Mine (DEP), located in Xishan coal field in Taiyuan, Shanxi. The four samples were collected from the No. 8 coal seam of the Taiyuan Formation, interbeded with limestone, mudstone and sandstone. The fifth sampling location, Gushuyuan Coal Mine (GSY) located in the Jincheng mining area, the southeastern part of Shanxi Province, which belongs to the Qinshui coalfield. The fifth sample is also taken from No.8 coal seam of Taiyuan Formation, which is mainly composed of sandy mudstone and limestone.

3. Sample processing
PAHs were extracted from the samples by Soxhlet, with dichloromethane for 8h, and purified by a rotary evaporator, in which the dichloromethane was evaporated and the organics remained in the flask. PAHs were obtained by column chromatography and elution of dichloromethane and n-pentane. It was purified with methanol to a 2ml, filtered and stored in a brown vial before determination.

HPLC (Water2695) is used to determine sixteen PAHs. The measurement conditions are shown in Table 1.

China Di ma Technology Company provided the standard sample of PAHs (100µg.ml^-1 dissolved in methanol) to determine the standard curve of PAHs. The standard solution of 100µg/ml was diluted to different concentrations, measuring the PAHs respectively curves at different concentrations, recording the peak areas of 16 of PAHs, and the primary equations of different concentrations and
peak areas of PAHs were fitted. The chromatogram of 15μg/ml PAHs standard curves are shown in Fig.1.

### Table 1. Analysis parameters of HPLC.

| Project                  | Conditions                     |
|-------------------------|--------------------------------|
| Chromatographic column  | SUPELCOSIL, LC-PAH             |
| Specification           | 5μm, 4.6mm×250mm               |
| Column temperature      | 27℃                            |
| Detector                | Water 2996 photodiode array detector |
| Wavelength              | 254nm                          |
| Mobile phase            | Acetonitrile-water             |
| Flow velocity           | 1.2ml/min                      |
| Sample size             | 10μL                           |

![Chromatogram of a 15μg/ml PAH standards by HPLC](image)

**Figure 1.** Chromatogram of a 15μg/ml PAH standards by HPLC-UV.

4. Analysis of experimental data

4.1. The contents of PAHs in the samples

The contents of 16 PAHs in the samples were calculated by the linear regression equation.

### Table 2. The contents of 16 PAHs in the samples.

| PAHs | ML (1.21%) (ng/g) | TL (1.47%) (ng/g) | XM (1.84%) (ng/g) | DEP (2.05%) (ng/g) | GSY (3.30%) (ng/g) |
|------|------------------|------------------|------------------|------------------|------------------|
| Nap  | 2499.29          | 3062.58          | 676.64           | 467.25           | 100.07           |
| AcPy | 334.30           | 1317.04          | 401.72           | 0.00             | 0.00             |
| AcP  | 71.77            | 0.00             | 0.00             | 0.00             | 9.97             |
| Flu  | 124.54           | 2031.79          | 80.62            | 0.00             | 0.00             |
| PhA  | 414.49           | 4861.37          | 590.31           | 46.50            | 0.00             |
| AnT  | 0.00             | 173.85           | 0.00             | 0.00             | 0.00             |
| FluA | 285.29           | 1303.58          | 259.74           | 0.80             | 0.00             |
| Pyr  | 492.51           | 2420.93          | 521.46           | 6.93             | 0.00             |
| BaA  | 91.39            | 1028.67          | 293.31           | 37.29            | 9.22             |
| Chr  | 601.76           | 9623.17          | 7302.63          | 222.53           | 63.35            |
| BbF  | 0.00             | 684.85           | 0.00             | 0.00             | 0.00             |
| BkF  | 0.00             | 507.87           | 78.59            | 0.00             | 0.00             |
| BaP  | 0.00             | 0.00             | 0.00             | 0.00             | 0.00             |
| DbA  | 0.00             | 391.73           | 65.87            | 0.00             | 0.00             |
| BghiP| 0.00             | 453.58           | 0.00             | 0.00             | 0.00             |
| In [1, 2, 3-cd] P | 0.00             | 0.00             | 0.00             | 0.00             | 0.00             |
4.2. Variation of $\sum^{16}_{\text{PAHs}}$ in the samples

![Figure 2. Changes of total PAHs in five samples.](image)

The total amount of PAHs extracted from coal gangue samples increase and then decrease as the degree of metamorphism increases (Fig 2), having the maximum 27861.01ng/g ($R_0,\text{max}=1.47\%$) and the minimum 182.61ng/g ($R_0,\text{max}=3.30\%$). Niu Zhi-yuan believed that it may be caused by structural changes of aromatic molecules in coal [20]. Functional groups in the aromatics from the original biomolecules are gradually depolymerized and removed when coal rank is low, which lead to an increase in extractable aromatic content in the coal gangue (these aromatic take the form of randomly ordered nuclei). The arrangement of aromatic structures in coal gangue became more and more orderly with continuous increase of coal rank ($R_0,\text{max}>1.47\%$), and they are arranged as approximately parallel sheets in anthracite ($R_0,\text{max}>2.5\%$), resulting in a decrease in the concentration of extractable aromatic substances. Scott A. Stout has obtained similar conclusions by studying coals from different coal ranks [15].

4.3. Changes of concentrations in HPAHs, LPAHs and toxicity equivalent in the samples

![Figure 3. Changes of concentrations in HPAHs and LPAHs in five samples.](image)
The concentrations of HPAHs and LPAHs in coal gangues increase firstly and then decrease (Fig 3). And the concentrations of HPAHs reached the maximum 16414.37ng/g in Tunlan Coal Mine (R₀,max=1.47%), where LPAHs got the maximum 11446.64ng/g as well. HPAHs and LPAHs reached the minimum 9.22ng/g and 173.39ng/g in Gushuyuan Coal Mine (R₀,max=3.30%).

The toxicity and carcinogenicity vary with the changes of PAH ring numbers and types. Toxicity equivalent was used to study the changes of toxicity in the samples. Nisbet et al proposed the concept of toxic equivalent (TEQ) to judge the toxicity of PAHs, where calculation formula TEQ=∑(ε×TEF), in which εrepresents the concentration of PAHs (Table 2), and TEF is a toxicity equivalent factor. The TEF of Bap is defined as 1, and the others are calculated based on it (Table 3) [21]. The table shows that LPAHs are basically non-carcinogenic, and the carcinogenicity and toxicity of PAHs are controled by HPAHs.

As can be seen from table 4, the TEQ value in the five samples are TL>XM>DEP>ML>JC. The toxicity of coal gangues increase firstly and then decreases as the increase of metamorphic degree, with a maximum value 0.7314 at TL (R₀,max=1.47%) and a minimum 0.0017 at GSY (R₀,max=3.30%). The change of toxic equivalent is consistent with the change of HPAHs content.

### Table 3. Chemical properties of 16 PAHs.

| PAHs         | Name   | Ring | Carcinogenicity | TEF  |
|--------------|--------|------|-----------------|------|
| Naphthalene  | Nap    | 2    | —               | 0.001|
| Acenaphthylene| Acpy   | 3    | —               | 0.001|
| Acenaphthene | Acp    | 3    | —               | 0.001|
| Fluorene     | Flu    | 3    | —               | 0.001|
| Phenanthrene | Pha    | 3    | —               | 0.001|
| Anthracene   | AnT    | 3    | —               | 0.01 |
| Fluoranthene | FluA   | 4    | cocarcinogen    | 0.001|
| Pyrene       | Pyr    | 4    | —               | 0.001|
| Benz[a] anthracene | BaA       | 4    | strong          | 0.1  |
| Chrysene     | Chr    | 4    | weak            | 0.01 |
| Benzo[b] fluoranthene | BbF      | 5    | strong          | 0.1  |
| Benzo[k] fluoranthene | BKF      | 5    | strong          | 0.1  |
| Benzo[a] Pyrene | BaP     | 5    | serious         | 1    |
| Dibenzo[a, h] anthra | DBA     | 5    | serious         | 1    |
| Benzo[g, h.i] Perylene | BghiP  | 6    | cocarcinogen    | 0.01 |
| Indeno[1, 2, 3-c, d] Perylene | In [1, 2, 3-cd] P | 6    | serious         | 0.1  |

### Table 4. Toxicity equivalent of the samples.

| Samples     | ML (R₀, max=1.21%) | TL (R₀, max=1.47%) | XM (R₀, max=1.84%) | DEP (R₀, max=2.05%) | GSY (R₀, max=3.30%) |
|-------------|---------------------|---------------------|---------------------|----------------------|---------------------|
| TEQ         | 0.0194              | 0.7314              | 0.1786              | 0.0065               | 0.0017              |
4.4. Characteristic Parameters of PAHs

Table 5. Characteristic values of PAHs in five groups of samples.

| Coal mine R0, max | ML  | TL  | XM  | DEP | JC  | Source of oil | Source of combustion | Low temperature spontaneous combustion sample (ML) 23°C-70°C |
|-------------------|-----|-----|-----|-----|-----|---------------|----------------------|----------------------------------------------------------|
|                   | 1.21% | 1.47% | 1.84% | 2.05% | 3.3% |                |                       |                                                          |
| Total index       | 1.58 | 1.66 | 1.03 | 0.97 | 0.64 | <4            | >4                   | 20.10-21.825                                             |
| Ant/(Ant+Phe)     | 0.00 | 0.03 | 0.00 | 0.00 | /   | <0.1          | >0.1                | 1-1                                                       |
| (3Ring/3Ring)     |     |      |      |      |     |               |                      |                                                          |
| Flua/(Flua+Pyr)   | 0.37 | 0.35 | 0.33 | 0.10 | 0   | <0.4          | >0.4                | 0.30-0.73                                                |
| (4Ring/4Ring)     |     |      |      |      |     |               |                      |                                                          |
| BaA/(BaA+Chr)     | 0.13 | 0.10 | 0.04 | 0.14 | 0.13 | <0.2          | >0.2                | 0.85-1                                                   |
| (4Ring/4Ring)     |     |      |      |      |     |               |                      |                                                          |
| InP/(InP+BghiP)   | /   | 0.00 | /   | /   | /   | <0.2          | >0.2                | 1-1                                                       |
| (6Ring/6Ring)     |     |      |      |      |     |               |                      |                                                          |

The ratios of Ant/(Ant+Phe), Flua/(Flua+Pyr) and BaA/(BaA+Chr) can be used to distinguish between combustion and petroleum sources [22]. Santino Orecchio and Maria Rosaria Mannino proposed to distinguish the sources of PAHs by the total index in order to cope with the situation that the ratio above cannot be used under certain conditions [17, 18].

A ratio of Ant/(Ant+Phe) less than 0.1 is usually an indication to the source of oil, and a ratio greater than 0.1 can be used to indicates the dominance of combustion [15, 26]. The values of Ant/(Ant+Phe) in the five simples are all less than 0.03, which is within the range of oil sources. Therefore, it can be inferred that the range of oil sources can be further divided, and the ratio between 0 to 0.03 can represent the source of raw coal gangue.

Flua/(Flua+Pyr) could indicates the source of combustion when the ratio of it is greater than 0.5, and the ratios less than 0.5 is the characteristic of oil source [26]. Yunker et al further divided the range of this proportion by doing researches [15]. They divided the ratio greater than 0.5 as the combustion source into the ratio greater than 0.5 indicates the combustion of coal and wood, and the ratio between 0.4 and 0.5 indicates the combustion of oil [15]. The ratios of Flua/(Flua+Pyr) vary from 0 to 0.37 in this paper, all of which are within the range of oil sources. Therefore, the ratios between 0 and 0.37 indicate the source of raw coal gangue.

A ratio of BaA/(BaA+Chr) less than 0.20 indicates the source of oil, the ratio between 0.20-0.35 indicate the source of oil or combustion, and the ratio greater than 0.35 indicates the source of combustion [15]. The ratios of BaA/(BaA+Chr) in five samples are 0.04-0.14. Therefore, it can represent the source of raw coal gangue when the ratio of BaA/(BaA+Chr) is in the range of 0.04-0.14.

The value of the total index from the samples increases firstly and then decrease, ranging from 0.64 to 1.66, which are less than 4 and within the range of petroleum sources [17, 18, 28]. Therefore, it represents the source of raw coal gangue when the values of total index ranges from 0.64 to 1.66.

5. Conclusion

The concentration of HPAHs, LPAHs and \( \sum 16 \) PAHs increase first and then decrease with the increase of coal ranks, which all gets the maximum value at \( R_0, \text{max}=1.47\% \) and the minimum value at \( R_0, \text{max}=3.30\% \); Toxicity has similar changes due to the influence of HPAHs contents and types; Neither the concentrations of BaP in the five samples nor the total concentrations of 16 PAHs did not exceed the limit set by the US Environmental Protection Agency.
The proportion of HPAHs increase first and then decrease, while LPAHs are opposite. LPAHs gradually convert to HPAHs due to the reordering of aromatic molecules and side chain attachment with the increase of coal ranks ($R_0,\text{max}=1.21\%-1.47\%$); LPAHs gradually combine into higher-ring PAHs (>6 ring) as continuous increase of coal ranks ($R_0,\text{max}=1.47\%-3.30\%$).

The ratios of Ant/(Ant+Phe) are 0-0.03, the proportions of Flua/(Flua+Pyr) are 0-0.37, the ratios of BaA/(BaA+Chr) are 0.04-0.14, and the range of total index in samples are 0.64-1.66. These ranges are useful in determining the source of raw coal gangue for PAHs in pollutants, but further experimental studies are needed to determine these ranges.

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