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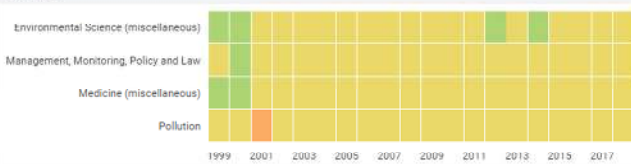
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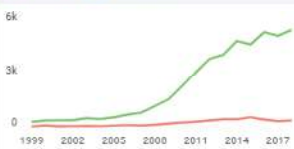
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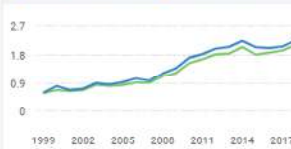
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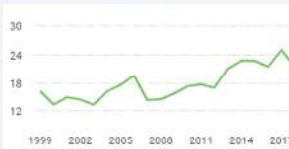
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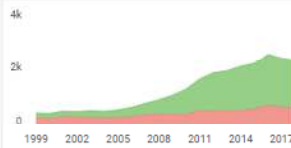
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Assessment of polycyclic aromatic hydrocarbons (PAHs) contamination in surface soil of coal stockpile sites in South Kalimantan, Indonesia

by Andy Mizwar

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Assessment of polycyclic aromatic hydrocarbons (PAHs) contamination in surface soil of coal stockpile sites in South Kalimantan, Indonesia

Andy Mizwar  · Bambang Joko Priatmadi ·
Chairul Abdi · Yulinah Trihadiningrum

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Abstract Concentrations, spatial distribution, and sources of 16 polycyclic aromatic hydrocarbons (PAHs), listed as priority pollutants by the United States Environmental Protection Agency (USEPA), were investigated in surface soils of three different coal stockpile, agricultural, and residential sites in South Kalimantan Province, Indonesia. Total PAHs concentration ranged from 4.69 to 22.67 mg kg⁻¹-dw. PAHs concentrations in soil of coal stockpile sites were higher than those in agricultural and residential soil. A complex of petrogenic origin and pyrolytic sources was found within the study area, as suggested by the isomeric ratios of PAHs. The results of principal component analysis and multiple linear regressions (PCA/MLR) showed that three sources contributed to the PAHs in the study area, including biomass and coal combustion (48.46 %), raw coal (35.49 %), and vehicular emission (16.05 %). The high value of total benzo[a]pyrene equivalent concentration (B[a]P_{eq}) suggests that local residents are exposed to a high carcinogenic potential.

Keywords Coal · PAHs · Petrogenic · Pyrolytic · South Kalimantan · Stockpile

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of environmental organic pollutants consisting of two or more fused aromatic rings in a variety of structural configurations (Haritash and Kaushik 2009; Antizar-Ladislao et al. 2004). They are widely known for their strong toxic, carcinogenic, and mutagenic impact to human health (Ray et al. 2008; Wang et al. 2010a). From several hundred different PAHs, 16 compounds have been listed as priority pollutants by the United States Environmental Protection Agency (16 USEPA-PAHs) (USA-EPA 2015; Achten and Hofmann 2009). Seven of these pollutants have been classified as probable human carcinogens (7 CPAHs) by the International Agency for Research on Cancer (Lee and Vu 2010).

Various sources of PAHs associated with fossil fuels have been reported in the literature. Incomplete combustion of fossil fuels (pyrolytic origin) is considered as the major anthropogenic source of PAHs in the environment, whereas the natural source of PAHs (petrogenic origin) is related to native PAHs in fossil fuels (Achten and Hofmann 2009). However, the petrogenic PAHs in the environment can also be derived from anthropogenic activities, such as coal mining and oil spills (Ahrens and Morrisey 2005; Short et al. 2007). Focusing on coal-mining activities, the coal native PAHs can be released to the environment during coal mining, processing,

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hauling, and stockpiling operations (Pies et al. 2007; Ahrens and Morrissey 2005). The level and pattern of PAHs in coals and its combustion products depend on the coal rank, coal origin, and combustion conditions (Ribeiro et al. 2012; Stout and Emsbo-Mattingly 2008; Wang and Liu 2015).

Some earlier publications have reported the PAHs contamination in soil of coal mining (Liu et al. 2012; Wang et al. 2010a) and coal waste dump areas (Sun et al. 2009; Ribeiro et al. 2012). However, the levels of PAHs contamination in the environment surrounding coal stockpile sites have not been studied in many coal-mining districts. Therefore, the objective of the present study was to determine the concentration, distribution, and composition of 16 USEPA-PAHs in surface soils of coal stockpile and its surrounding areas (agricultural and residential). In addition, the primary sources and risk assessment of PAHs are discussed.

Materials and methods

Study area and samples collection

South Kalimantan, one of the five Indonesian provinces in Borneo Island, is the most important coal-producing bases in Indonesia. The coals in South Kalimantan are mostly sub-bituminous in rank with bituminous coal in some areas (Nas and Hidartan 2010; Riswandi 2008), which are mainly used for power generation and for industrial boilers. As a consequence of the intensive coal-mining activities in the last 25 years, there are 813 coal stockpile sites in South Kalimantan (ESDM 2012), and some of them have been abandoned without adequate environmental treatment. These areas potentially have significant PAHs contamination.

Three different coal stockpile sites in South Kalimantan Province, Indonesia (Fig. 1) were selected for this study. Site-1 (Sp-1) is located very close to the coal haul road in Tapin coal-mining district and near (<500 m from) a residential area (Rs-1) and agricultural land (Ag-1). This site occupies an area of about 2 ha, and it has been abandoned without any reclamation for longer than 5 years. Site-2 (Sp-2) is located in the Kintap coal-mining field in Tanah Laut regency with an area of approximately 3.8 ha. This site has been abandoned for more than 3 years with land revegetation performed sporadically in some parts. An agricultural (Ag-2) and residential (Rs-2) area lies approximately 500 and

750 m from the coal stockpile site, respectively. Site-3 (Sp-3) is located in the Sungai Danau coal-mining field in Tanah Bumbu regency. This active coal stockpile site is very small if compared with the other coal stockpile sites (it occupies approximately 0.5 ha). This site is located quite far (>1000 m) from residential (Rs-3) and agricultural (Ag-3) areas.

Surface soil samples (5–20 cm) were collected in March 2015. Eight to ten soil sub-samples were collected at each sampling point by grab sampling. Especially in coal stockpile, the soil samples were collected in soil surface around coal stockpile sites, not in coal piles. After removal of stones and large particles, those soil sub-samples were mixed to make nine composite samples. The composite samples (weighing 2 kg each) were placed in polyethylene bags and transported to the laboratory. The soil samples were stored at 0–4 °C until the chemical analysis was done.

Samples extraction and cleanup

Before analysis, the soil samples were air-dried and homogenized by passing through a 2-mm sieve. Approximately 40 g of soil samples were extracted by the Soxhlet extraction method with 120 mL dichloromethane (Merck, Germany) for 8 h. Activated copper pieces were added for desulfurization. The extract was transferred to a rotary evaporator and pre-concentrated to a volume of 1 mL.

The concentrated extract was cleaned up using alumina chromatographic column with 4 g alumina oxide (Sigma-Aldrich type WB-5 basic, USA). The cleaned extract was then separated and fractionated using silica chromatographic column with 4 g silica gel (Merck, Germany). The fraction containing PAHs was eluted with 5 mL of methylene dichloride–hexane mixture (1:1 v/v) and then evaporated to 1 mL in a water bath. Prior to analysis by gas chromatography–mass spectrometry (GC-MS), 1 mL of the solution of Semivolatile Internal Standards Mix (naphthalene-d₈, acenaphthalene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂ and perylene-d₁₂) was added to the sample.

Analysis of PAHs and quality assurance/quality control

Samples analysis was performed on a Hewlett-Packard (HP) Series II gas chromatograph coupled with the HP 5989 mass spectrometer (Hewlett-Packard, USA), using electron ionization in the selected ion monitoring mode.

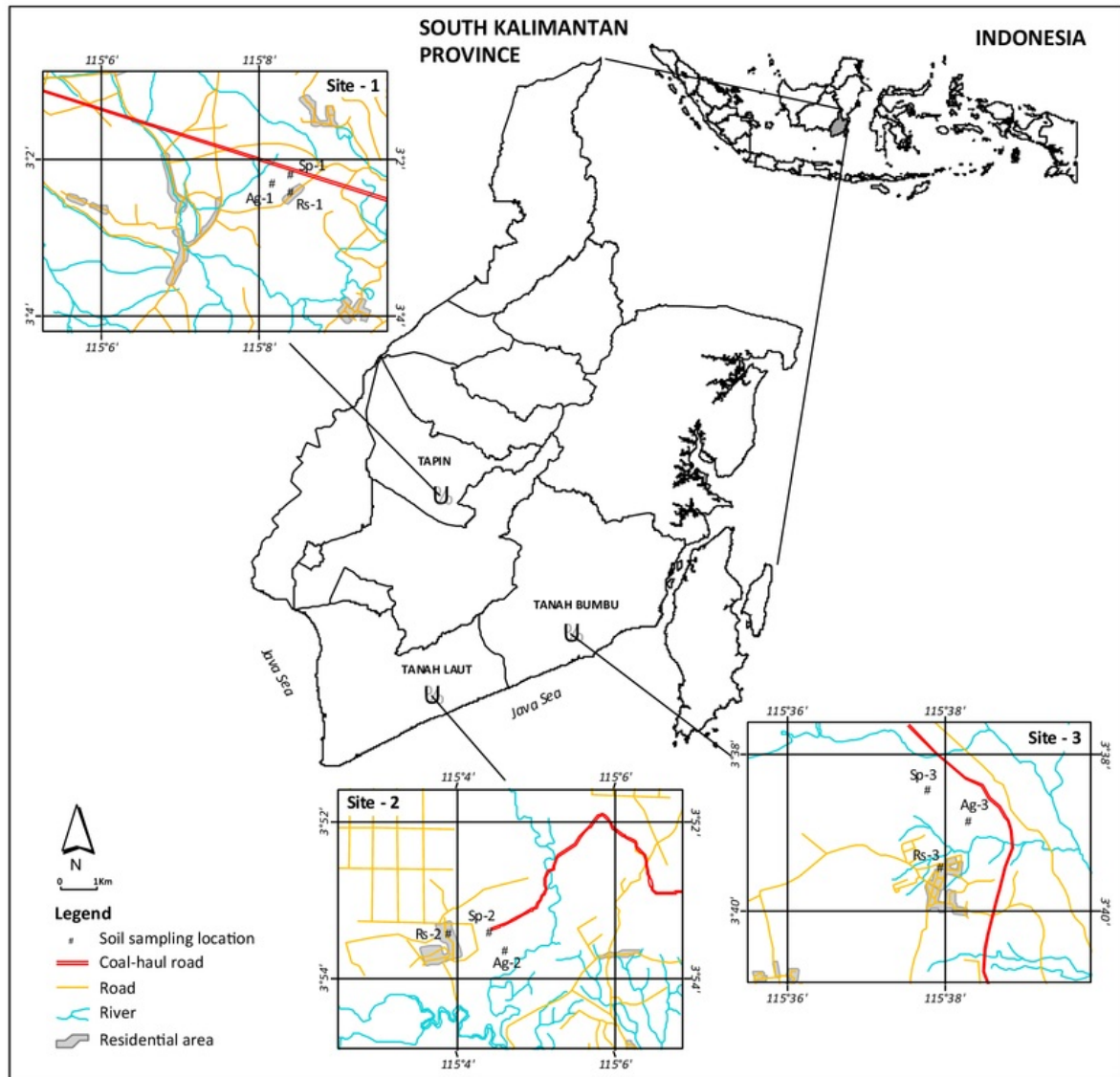


Fig. 1 Sampling location of surface soil (*Sp*—a representative coal stockpile soil; *Ag*—a representative agricultural soil; *Rs*—a representative residential soil)

Information about the analyzed PAHs, along with the quantification and confirmation ions from their mass spectra, is listed in previous work (Mizwar and Trihadiningrum 2015). Chromatographic resolution is achieved with a 30 m × 250 μm DB-5 capillary column which has a 0.25-μm film thickness. The carrier gas was helium at a flow rate of 1 mL/min. The temperature of the injection port was 300 °C, and transfer line was 290 °C. The quantitative analysis was done by internal calibration method, and PAHs identification was performed by comparison of their retention time with standards.

Analytical methods were checked for the precision and accuracy. All samples were analyzed in triplicate to obtain the average level. Replicate analyses gave the relative standard errors less than 15 %. The procedural blank was determined by going through the extraction and cleanup procedures using glass beads instead of soil sample. The results indicated that they were generally low and posed no problem to the analytical quantification. The recovery efficiency was checked by analyzing soil samples spiked with known amount of

PAHs standard. The spiked test showed that the recoveries of 16 USEPA-PAHs were from 80 to 95 %.

Results and discussion

Concentration and distribution of PAHs in surface soils

The levels of the 16 individual PAHs analyzed as well as the sum of these 16 USEPA-PAHs (\sum_{16} PAHs) and 7 CPAHs (\sum_{7} CPAHs) at different sites are listed in Table 1. In general, the \sum_{16} PAHs in the surface soils varied from 4.69 mg kg⁻¹ (Rs-3) to 22.67 mg kg⁻¹ (Ag-1), with an arithmetic mean of 11.72 mg kg⁻¹. Meanwhile, the \sum_{7} CPAHs ranged from 0.72 mg kg⁻¹ (Rs-3) to 10.06 mg kg⁻¹ (Sp-1), with an arithmetic mean

of 4.35 mg kg⁻¹. These measured concentrations greatly exceed the level of natural PAHs concentrations found in soil (0.001–0.010 mg kg⁻¹), which are primarily derived from plant synthesis and natural fires (Edwards 1983). This result indicates that the soils from the area of the present study have been significantly affected by anthropogenic activities.

In coal stockpile soils, the highest \sum_{16} PAHs was found in Sp-1 (17.23 mg kg⁻¹), followed by Sp-3 (15.35 mg kg⁻¹) and Sp-2 (12.52 mg kg⁻¹). The high level of \sum_{16} PAHs at Sp-1, which is near the pivot of the coal haul road, was perhaps caused by the combined effect of intensive coal stockpiling activities in the past and the coal hauling traffic. During coal stockpiling process, coal particles can spread to the soil through stockpiling operation, transport activities, and erosion of coal piles by wind and water, thus releasing coal

Table 1 PAHs concentrations in soil samples

Compound	No. of aromatic rings	Concentrations (mg kg ⁻¹)								
		Site-1			Site-2			Site-3		
		Sp-1	Ag-1	Rs-1	Sp-2	Ag-2	Rs-2	Sp-3	Ag-3	Rs-3
Naphthalene (Naph)	2	1.66	4.22	2.09	1.02	1.11	0.57	0.88	0.89	0.66
Acenaphthylene (Acy)	3	0.03	1.94	1.34	0.12	0.71	0.61	0.19	0.96	0.72
Acenaphthene (Acen)	3	0.01	2.57	2.83	0.02	1.51	0.32	0.06	1.74	1.32
Fluorene (Flu)	3	0.08	0.92	0.56	0.11	0.30	0.37	0.34	0.24	0.18
Phenanthrene (Phen)	3	3.34	1.14	0.96	3.71	0.23	0.22	3.08	0.25	0.18
Anthracene (Anth)	3	0.13	0.11	0.10	0.25	0.20	0.13	0.32	0.23	0.17
Fluoranthene (Flan)	4	0.58	3.11	1.30	0.23	0.69	0.40	1.93	0.71	0.53
Pyrene (Pyr)	4	0.89	1.60	0.80	0.58	0.43	0.40	1.48	0.27	0.20
Benzo[a]anthracene (B[a]A)	4	1.01	2.36	0.26	0.56	0.14	0.44	0.63	0.22	0.16
Chrysene (Chry)	4	1.33	0.85	0.40	0.92	0.21	0.10	0.43	0.13	0.10
Benzo[b]fluoranthene (B[b]F)	5	1.82	1.12	0.74	2.34	0.39	0.19	1.04	0.22	0.16
Benzo[k]fluoranthene (B[k]F)	5	2.81	0.41	0.51	1.85	0.27	0.13	0.45	0.12	0.09
Benzo[a]pyrene (B[a]P)	5	0.76	0.23	0.63	0.54	0.34	0.10	2.31	0.17	0.13
Indeno[1,2,3-cd]pyrene (InP)	5	2.22	0.46	0.95	0.17	0.50	0.49	2.00	0.07	0.03
Dibenzo[a,h]anthracene (DB[ah]A)	6	0.11	0.36	0.75	0.03	0.40	0.26	0.07	0.06	0.04
Benzo[g,h,i]pyrene (B[ghi]P)	6	0.46	1.28	0.20	0.07	0.10	0.02	0.14	0.02	0.01
\sum_{16} PAHs		17.23	22.67	14.42	12.52	7.55	4.76	15.35	6.29	4.69
\sum_{7} CPAHs		10.06	5.79	4.24	6.41	2.26	1.72	6.93	0.99	0.72
\sum B[a]P _{eq}		1.67	1.03	1.63	1.07	0.87	0.49	2.80	0.29	0.22
LMW-PAHs (%)		30.47	23.04	33.75	87.06	53.87	90.51	51.34	35.32	68.86
MMW-PAHs (%)		22.10	10.09	30.99	63.23	19.51	27.94	18.02	21.32	21.12
HMW-PAHs (%)		47.43	22.09	41.69	30.78	26.62	13.86	24.59	18.94	10.02

native PAHs to the environment (Stout and Emsbo-Mattingly 2008). On the other hand, the lower concentration of $\sum_{16}\text{PAHs}$ in soil samples from Sp-2 might be related to adsorption and removal effects by the plants (Nadal et al. 2004; Bakker et al. 2000), especially at the spots of land-revegetated area. In addition, considering that site-3 is a newly operated coal stockpile site, it may be not subjected to long-term contamination, thus causing the level of $\sum_{16}\text{PAHs}$ in Sp-3 to be lower than Sp-1. There are quite a few studies in the literature reporting contamination of PAHs in coal stockpile soils, but the $\sum_{16}\text{PAHs}$ concentrations in the present study were significantly higher than that measured in our previous study (11.79 mg kg^{-1}) (Mizwar and Trihadiningrum 2015).

The highest $\sum_{16}\text{PAHs}$ concentrations from agricultural and residential soils were found in Ag-1 (22.67 mg kg^{-1}) and Rs-1 (14.42 mg kg^{-1}), respectively; the sites were located very close to the coal stockpile and coal haul road. Meanwhile, sites Ag-3 (6.29 mg kg^{-1}) and Rs-3 (4.69 mg kg^{-1}), which were located quite far from the coal stockpile and coal haul road, showed a lower level of PAHs. These results indicate that the agricultural and residential soils of the present study have been affected by coal stockpiling activities. Furthermore, these measured $\sum_{16}\text{PAHs}$ concentrations were significantly higher than those measured in the agricultural and residential soils, which were located around Anhui coal mine, China (mean values of 0.15 and 0.44 mg kg^{-1} for agricultural and residential soil, respectively) (Wang et al. 2010a) and Tiefa coal mine, China (mean value of 0.49 mg kg^{-1} for agricultural soils) (Liu et al. 2012).

The national standards concerning allowable PAHs concentration in soils has not yet been regulated in Indonesia. Malawska and Wiołkomirski (2001) suggested a Polish soil contamination classification system based on 16 USEPA-PAHs as follows: unpolluted soil (natural content) ($<2 \text{ mg kg}^{-1}$), unpolluted soil (increased content) ($0.2\text{--}0.6 \text{ mg kg}^{-1}$), slight polluted soil ($0.6\text{--}1 \text{ mg kg}^{-1}$), polluted soil ($1\text{--}5 \text{ mg kg}^{-1}$), heavily polluted soil ($5\text{--}10 \text{ mg kg}^{-1}$), and very heavily polluted soil ($>10 \text{ mg kg}^{-1}$). According to the criteria, 22 % of the surface soils were polluted including Rs-2 and Rs-3, and 22 % were heavily polluted including Ag-2 and Ag-3. Meanwhile, very heavily polluted accountings for 56 % of total samples were Sp-1, Sp-2, Sp-3, Ag-1, and Rs-1.

Compositional profile of PAHs in soils

The 16 USEPA-PAHs can be divided into three groups according to their molecular weight: low molecular weight PAHs with two and three aromatic rings (LMW-PAHs), medium molecular weight PAHs containing four aromatic rings (MMW-PAHs), and high molecular weight PAHs comprising five and six aromatic rings (HMW-PAHs) (Loick et al. 2009). In the coal stockpile soils, HMW-PAHs (42.20 %) were the dominant compounds, followed by LMW-PAHs (34.63 %) and MMW-PAHs (23.16 %). The high content of HMW-PAHs in the studied samples might be associated with the low-rank coals (Stout and Emsbo-Mattingly 2008; Wang et al. 2010b) produced in South Kalimantan. However, other authors describe that low-rank coals are characterized by a high concentration of LMW PAHs (Achten and Hofmann 2009; Xue et al. 2007). The predominance of HMW-PAHs in coal stockpile soil samples might be also due to the occurrence of self-combustion in coal piles (Emsbo-Mattingly and Stout 2011; Ribeiro et al. 2012). Therefore, the source of PAHs in soils derived from raw coal has hardly been identified due to the lack of information on native coal-bound PAHs in soils (Wang et al. 2010a) and was disturbed by invasion from pyrolytic sources (Liu et al. 2012).

The levels of LMW-PAHs were dominant and formed 56.78 % of $\sum_{16}\text{PAHs}$ in the agricultural zone soils, whereas MMW-PAHs and HMW-PAHs contributed 25.18 and 18.03 % of $\sum_{16}\text{PAHs}$, respectively. Similarly, the dominant compounds in the residential soils were LMW-PAHs (56.74 %), followed by MMW-PAHs (22.84 %) and HMW-PAHs (20.42 %). The abundance of LMW-PAHs in the two kinds of soils implies that this pollution might be caused by incomplete combustion of fossil fuel and the burning of agricultural waste (Kakareka and Kukharchyk 2003).

PAHs source and contribution estimates

The identification of primary PAHs sources is important for controlling pollutant emissions, and several methods have been proposed to apportion these sources. PAHs diagnostic ratio is one of the most widely used as a tool for identifying and assessing PAHs sources. This method involves pairs of PAHs with the same molar mass and similar physicochemical properties to distinguish between petrogenic and pyrolytic sources (Wang et al.

2010c; Shi et al. 2015). According to the PAHs isomer ratios compiled by Yunker et al. (2002), the Anth/(Anth+Phen) ratio <0.1 is taken as an indication of petrogenic origin (crude oil or raw coal), while the ratio >0.1 indicates a pyrolytic source (biomass and coal combustion, and natural fire). The Flan/(Flan+Pyr) ratio of less than 0.4 suggests a petrogenic origin, while the value between 0.4 and 0.5 indicates petroleum (liquid fossil fuel, vehicle, and crude oil) combustion, and values greater than 0.5 indicate combustion of biomass and coal. The ratio of B[a]A/(B[a]A+Chry)<0.2 implies petrogenic sources; 0.2–0.35 indicates biomass and coal burning; and >0.35 indicates petroleum combustion. In addition, the IP/(IP+B[ghi]P) ratio of less than 0.20 likely indicates petrogenic sources, whereas values between 0.2 and 0.5 imply petroleum combustion, and values greater than 0.5 imply the combustion of coal and biomass.

In this study, the values of Anth/(Anth+Phen) were between 0.04 to 0.48, and the values of Flan/(Flan+Pyr) ranged from 0.28 to 0.72. The cross plot of Anth/(Anth+Phen) and Flan/(Flan+Pyr) is depicted in Fig. 2a. It indicates that the sources of PAHs in soils could be classified into three distinct groups. About 22 and 33 % of the sampling sites exhibited the typical characteristics of petrogenic origin (Sp-1 and Sp-2) and the mixture of petrogenic, and biomass and coal combustion sources (Sp-3, Ag-1, and Rs-1), respectively. The remaining sites (Ag-2, Ag-3, Rs-2, and Rs-3) showed the signature of a combination of pyrolytic, and biomass and coal combustion sources. On the other hand, the values of B[a]A/(B[a]A+Chry) and InP/(InP+B[ghi]P) ranged from 0.38 to 0.81 and 0.27 to 0.97, respectively, suggesting a pyrolytic source (Fig. 2b). This indicates that PAHs in

surface soils of coal stockpile and its surrounding area were not from a single source but a mixture of pyrogenic and pyrolytic sources.

Another tool that is usually used to further identify the profiles of PAHs sources is principal components analysis (PCA) followed by multiple linear regression (MLR) (Li et al. 2013; Zuo et al. 2007). PCA/MLR uses PCA to represent the total variability of the original PAHs data in a minimum number of factors. The PCA factor scores for each sample are then used to calculate the contribution percentage of the major sources using MLR (Larsen and Baker 2003). In this study, PCA and MLR analyses were performed using XLSTAT-2014.5.03 in MS Excel 2010 software.

The results of PCA analysis of soil PAHs concentrations are presented in Table 2, with relatively high and moderate factor loadings marked in italics (≥ 0.5). Three principal components (PCs) were extracted with varimax rotation, with the accumulative variances accounted for 87.20 % of the total variability. Among the three, factor 1 accounted for 39.42 % of the total variance, is dominated by Naph (0.95), Flu (0.93), Flan (0.93), B[ghi]P (0.90), B[a]A (0.82), Acy (0.78), and Pyr (0.77), with moderate loadings of Acen (0.62) and DB[ah]A (0.51). Naph and Flu considered as components of low temperature pyrolytic sources (Luo et al. 2006). Meanwhile, several authors have reported Acy, Acen, Flan, Pyr, B[a]A, DB[ah]A, and B[ghi]P as pre-minant biomass and coal combustion profile (Larsen and Baker 2003; Wang et al. 2007; Zuo et al. 2007; Shi et al. 2015). Therefore, factor 1 can represent biomass and coal combustion.

Factor 2 is responsible for 28.90 % of the total variance. This factor is highly weighted by B[k]F

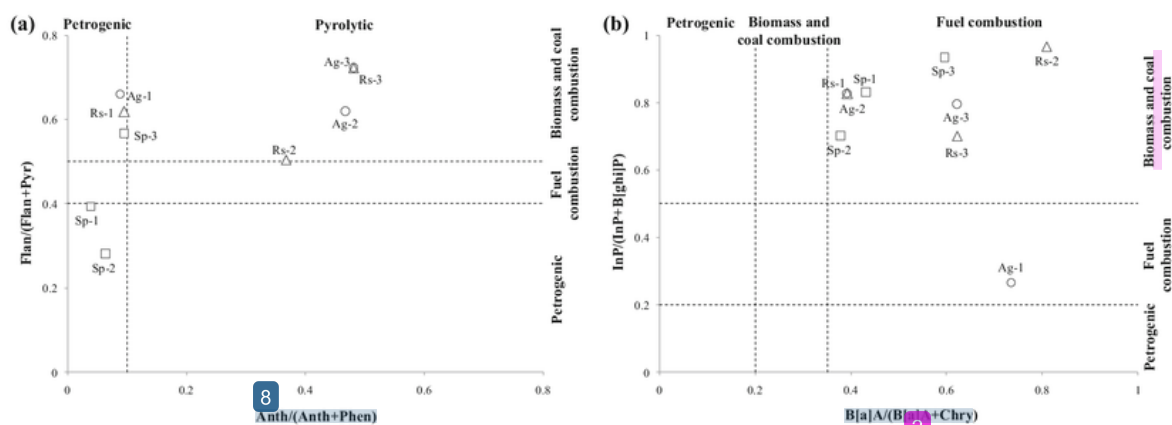


Fig. 2 Isomer ratio plots of **a** Anth/(Anth+Phen) vs Flan/(Flan+Pyr) and **b** B[a]A/(B[a]A+Chry) vs InP/(InP+B[ghi]P)

Table 2 Factor loadings of soil PAHs after varimax rotation

Variable	Principal components (PCs)		
	1	2	3
Naph	0.95	0.23	-0.19
Acy	0.78	-0.44	-0.40
Ben	0.62	-0.49	-0.43
Flu	0.93	-0.28	-0.07
Phen	-0.06	0.83	0.50
Anth	-0.43	-0.07	0.74
Flan	0.93	-0.09	0.31
Pyr	0.77	0.30	0.55
B[a]A	0.82	0.42	0.01
Chry	0.28	0.96	0.04
B[b]F	0.06	0.91	0.18
B[k]F	-0.16	0.97	-0.01
B[a]P	0.04	0.15	0.96
InP	0.11	0.47	0.60
DB[ah]A	0.51	-0.28	-0.30
B[ghi]P	0.90	0.32	-0.10
Variance (%)	39.42	28.90	18.88
Accumulated variance (%)	39.42	68.32	87.20

Rotation method: Varimax with Kaiser Normalization

Values in italics: relatively high and moderate loadings (≥ 0.50)

(0.97), Chry (0.96), B[b]F (0.91), and Phen (0.83). Based on the coal types that were dumped in stockpiling sites and a high proportion of Phen, B[b]F and B[k]F in the coal stockpile soils which was associated with unburned low-rank coals (Stout and Emsbo-Mattingly 2008; Wang et al. 2010b), it is reasonable to assign factor 2 as the source from raw coal. Factor 3 accounted for 18.88 % of the total variability, is predominately weighted by B[a]P (0.96) and Anth (0.74), and moderately weighted in InP (0.60), Pyr (0.55), and Phen (0.50). The source this factor represents appears to be road dust collected from the Shanghai urban area (Ren et al. 2006; Liu et al. 2007). In addition, both InP and B[a]P have been identified as typical tracers of vehicular PAH sources (Fang et al. 2007; Larsen and Baker 2003; Shi et al. 2015). Therefore, this factor is selected to represent the vehicular source of PAHs.

MLR was performed on the PCA factor scores to obtain the mass apportionment of the three sources of the total PAHs burden in each soil sample. Factor score 1–3, which represented biomass and coal combustion, raw coal, and vehicular emission sources, were regressed

against the standard normalized deviate (Z) of the total PAHs concentration. The MLR equation is as follows: $Z = 4.56X_1 + 3.34X_2 + 1.51X_3$ ($R^2 = 0.976$). The quantitative contributions of the three major sources in the surface soil were 48.46 % for factor 1 (biomass and coal combustion), 35.49 % for factor 2 (raw coal), and 16.05 % for factor 3 (vehicular emission).

Risk assessment of PAHs

Toxic equivalent quantity of benzo[a]pyrene (TEQ-B[a]P) was used to assess the ecotoxicological risk at a specific site. The total B[a]P equivalent concentration ($B[a]P_{eq}$) was calculated as the sum of multiplying the concentration of individual PAH by the corresponding toxic equivalent factors (TEFs) (Wang et al. 2013). The TEFs for 7 CPAHs including B[a]A, B[a]P, B[b]F, B[k]F, IP, DB[ah]A, and Chry are 0.1, 1, 0.1, 0.01, 0.1, 1, and 0.001, respectively (Tsai et al. 2004). This method has the main advantage of being relatively easy to apply in the affected human environment. However, underestimation of the risk may occur because only limited PAH compounds are considered (WHO/IPCS 1998).

As listed in Table 1, the values of $\sum B[a]P_{eq}$ from the different sampling sites vary from 0.22 mg kg^{-1} (Rs-3) to 2.80 mg kg^{-1} (Sp-3), with an arithmetic mean of 1.22 mg kg^{-1} . The coal stockpile soils had the highest mean $B[a]P_{eq}$, with a value of 1.85 mg kg^{-1} , compared with agricultural soil (0.73 mg kg^{-1}) and residential soil (0.78 mg kg^{-1}). This is due to the higher concentration of 7 CPAHs in the coal stockpile soils than those in agricultural and residential soils. Moreover, it should be noted that the influence of the high B[a]P concentration was taken into account for the high value of $\sum B[a]P_{eq}$ in Sp-3. These values suggested that the higher carcinogenic potency in the coal stockpile soils, especially in Sp-3, should be given much more attention. In addition, the local residents could be affected by the carcinogenic potency of the high value of $B[a]P_{eq}$ in residential soils, and a certain quantity of $B[a]P_{eq}$ exposure to agricultural soils could create potential risk through enrichment in the food chain.

Conclusion

The total contents of 16 USEPA-PAHs in studied soil samples were higher than those in other studies and the

standards of Poland. The coal stockpiling activities evidently promoted the PAHs contamination level due to the PAHs in the coal stockpile soils being remarkably higher than those in the agricultural and residential soils. Coal stockpiling activities not only contaminated the coal stockpile soils but also affected both the agricultural and residential soils surrounding the coal stockpile sites. The PAHs diagnostic ratio results indicated that PAHs in surface soils were a mixture of pyrogenic and pyrolytic sources. The PCA/MLR results showed that biomass and coal combustion, raw coal, and vehicular emission were potentially the main sources for PAHs contamination in the study area. The high value of B[a]P_{eq} suggests that the soils of the study area are highly contaminated and pose a potential health risk to humans.

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