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Parent and alkylated polycyclic aromatic hydrocarbon emissions from coal seam fire at Wuda, Inner Mongolia, China: characteristics, spatial distribution, sources, and health risk assessment

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Abstract The Wuda coalfield in Inner Mongolia is a vital coal base in China, and it is the hardest-hit area for coal fires (spontaneous combustion of coal seams and coal gangue). Using gas chromatography–mass spectrometry, this work tested the concentration and analyzed the characteristics, distribution, sources, and health risks of polycyclic aromatic compounds (PACs) in the surface soil of the Wuda District, including the coal mine, coal fire, agricultural, and background areas. The soil of coal mine and coal fire area were heavily polluted with PACs, with mean concentrations of 9107 and 3163 μg kg^{-1}, respectively, considerably higher than those in the agricultural (1232 μg kg^{-1}) and background areas (710 μg kg^{-1}). Alkyl polycyclic aromatic hydrocarbons (APAHs) were the dominant pollutants among these PACs, accounting for 60–81%. Alkyl naphthalenes and alkyl phenanthrenes are the primary pollutants in APAHs, accounting for 80–90% of the total amounts. Additionally, using the positive matrix factorization method, it can be concluded that the primary PAC sources are petrogenic sources, coal and biomass combustion, coal fires, and vehicle emissions. Finally, according to the cancer risk values of 16 PAHs, only the coal mine area showed a potential cancer risk. However, this result lacks a risk assessment of APAHs and underestimates the actual risk. The results of this study improved the understanding of PAC pollution in coal fire and surrounding areas and provided a reference for environmental and health risk investigations.

Keywords Polycyclic aromatic compounds · Soil · Wuda District · Coal fire · Source identification · Carcinogenic risk

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Introduction

Coal fires are natural disasters that spontaneously burn underground coal seams under natural conditions, resulting in mineral loss and environmental damage (Kuenzer et al., 2012). Increased temperatures of sulfur-containing minerals through oxidation lead to spontaneous and even violent combustion of coal, causing underground coal fissures, coal seam outcrops, and coal gangue pile fires, resulting in coal fire disasters (Heffern & Coates, 2004; Stracher, 2004). Coal fires have been reported in China (Kuenzer & Stracher, 2012; Kuenzer et al., 2012; Li et al., 2018), the USA (Stracher, 2004; Strachera & Taylor, 2004), India (Agarwal et al., 2006; Gangopadhyay et al., 2006), South Africa (Bell et al., 2001; Pone et al., 2007), Australia (Glasspool, 2000), and Russia (Sharygin et al., 2009; Stracher, 2004). Coal fires are widely distributed in northern China, resulting in a total loss of more than 200 million tons of coal annually (Kuenzer & Stracher, 2012). The Wuda coal mine area in Inner Mongolia is one of the largest coal fire areas in the world. The Wuda coal fire was first observed in 1961 and had been burning continuously for more than 60 years. Rational development and utilization of coal resources is essential, such as heavy metals and sulfides in coal and its by-products, which have impacts on human health and the global climate (Gopinathan et al., 2022a, 2022b, 2022c; Wu et al., 2022). Moreover, coal fires emit large amounts of inorganic and organic pollutants, including mercury, fluorine, sulfur dioxide, and carbon dioxide (Engle et al., 2013; Hong et al., 2017; Hower et al., 2013; O'Keefe et al., 2010; Zhang et al., 2013). They also release organic pollutants, including CH4, BETX, and polycyclic aromatic hydrocarbons (PAHs) (Garrison et al., 2016; Hower et al., 2013; Liang et al., 2018b), a group of organic pollutants with high biological toxicity and persistence.

The sources of PACs are classified into two types: anthropogenic and natural. Anthropogenic sources include petrogenic (crude oil and petroleum products e.g., kerosene, gasoline, diesel fuel, lubrication oil, and asphalt) and pyrogenic (formed by the incomplete combustion of organic matter such as coal, petroleum, and wood). Natural sources include oil seeps, grass and forest fires, volcanoes, ancient sediment erosion, and early diagenesis (Kannan et al., 2005; Manoli & Samara, 1999; Saha et al., 2012; Tsibart & Gennadiev, 2013).

Our earlier research on mercury, fluorine, sulfate, and 16 PAHs in the Wuda District soil showed that coal fire is the main pollution source (Hong et al., 2018, 2021; Li et al., 2018; Liang et al., 2018a; Qian et al., 2021). However, coal fires are not considered an important source of environmental PACs.

Previous studies have shown that alkyl polycyclic aromatic hydrocarbons (APAHs) were the dominant PACs (Chen et al., 2017; Golzadeh et al., 2021; Huang et al., 2021; Lian et al., 2009; Qian et al., 2022a, 2022b). Additionally, APAHs are more toxic and persistent than their parent compounds (Fallahtafti et al., 2012; Kang et al., 2016; Mu et al., 2014), notably affecting the mortality and malformation rates of biological embryo development (Fallahtafti et al., 2012; Kang et al., 2016; Mu et al., 2014). However, few studies have focused on PACs, particularly APAHs, from coal fire areas. APAH research started relatively late and is still at a relatively early stage internationally, and many researchers are still in the method exploration stage, limiting any large-scale systematic research. Conversely, except for China, countries with strong scientific research abilities most rely on oil as the main fossil energy source and have a very low dependence on coal. Therefore, they do not have the urgency to carry out research on coal-related APAHs.

This paper emphasizes the urgency and necessity of in-depth research into APAHs. Coal fire areas occur globally and analyzing the characteristics of PACs in the soil of these areas is important for promoting an understanding of coal fire disasters and the necessary ecological and environmental protection required in mining areas. This study included PACs (16 PAHs and APAHs) in the soil of the Wuda coalfield and its surrounding areas as the research object and analyzed pollution levels, spatial distribution, sources, and health risks of these PACs. In addition, PAC types in coal mine and coal fire areas and PAC characteristics of coal fire emissions were analyzed. The results can provide a scientific basis and data support for environmental governance and improvements in coal mine and coal fire areas similar with the Wuda coalfield.

Materials and methods

Background and sample collection

Coal fires in China are mainly distributed in the north, with an east–west length of about 5000 km and a north–south width of 700 km (Li et al., 2017a, 2017b) (Fig. 1a). Wuda District is in Wuhai City, in the west of the Inner Mongolia Autonomous Region, with latitude 39° 20′–39° 40′ north and longitude 106° 30′–106° 50′ east (Qian et al., 2021). Functionally, Wuda District is divided into the following areas: coal mine, residential, agricultural, and industrial park areas (Fig. 1b) with a total area approximately 220 km^2 (Li et al., 2018). The coal mine area of Wuda (approximately 35 km^2) is in the northwest bearing of

Wuda District, close to the desert (Qian et al., 2021). Wuda District is an older and larger mining area in China which has coal mining activities and a certain scale of continuous coal fires, causing severe pollution of the surrounding environment (Fig. 1c, d). The agricultural area is in the eastern part of the Wuda District and a small island near the Yellow River to the east of the agricultural area was denoted the background area.

The areas with coal fires in Wuda District were defined as coal fire areas. The sampling points in the coal fire area were located near the smoke vents of coal fire with a total of 14 soil samples being collected and numbered M1–M14. Five samples (M15–M19) were collected from other coal mine areas without coal fires. In addition, six (N1–N6)
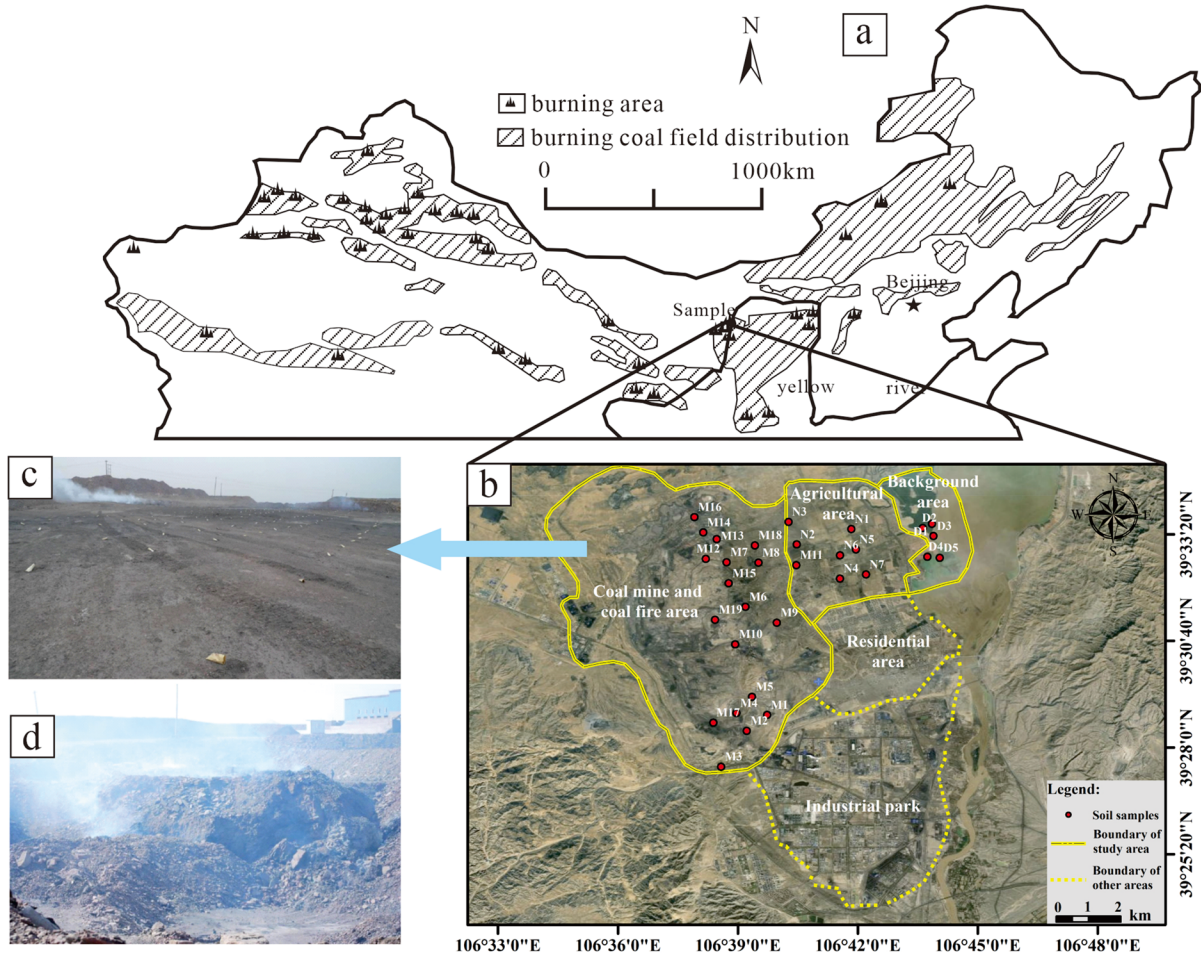
Fig. 1 The geographical location of the research area and distribution of sampling points. **a** Distribution of coal fires in the northern region and the location of Wuda coalfield, **b** distribution of sampling points in Wuda District, **c–d** situation of the coal fire site

and five samples (D1–D5) were collected from the agricultural and background areas, respectively (Fig. 1b). A total amount of 30 soil samples were collected in June 2015. After brushing the dust off, the surface soil samples at a depth of 0–10 cm were collected using a small shovel. Non-natural soil materials such as branches, building materials, and plastics were manually removed and stored in a sealed sample bag. Three parallel samples were collected from the same area and mixed evenly. Geographic coordinates were recorded using a handheld GPS locator. After the soil samples were brought to the laboratory, they were naturally air-dried and ground indoors and then. They were stored in a sealed container away from light for later use.

Pretreatment and analysis of PACs

All samples were extracted and analyzed as previously described by Qian et al. (2022b). The soil and anhydrous sodium sulfate (2 g each) were transferred into a 10-mL glass tube and 5 mL dichloromethane (400 μg/mL NAP-d8) was added under ultrasound for 5 h at 20 °C. Next, the supernatant was transferred into a 2-mL centrifuge tube and vortexed for 1 min and then centrifuge tube centrifuged at 14,000 rpm for 10 min. After centrifugation, the supernatant was passed through a syringe filter (pore size 0.22 μm). Finally, 100 μL filtrate was mixed with 100 μL internal standard (ACE-d10, PHE-d10, CHR-d12, and PER-d12).

The soil PACs were analyzed by gas chromatography–tandem mass spectrometry (Waters, Xevo TQ-GC) with full scan mode. The column model number was DB-5MS (length, 30 m; internal diameter, 0.25 mm; film thickness, 0.25 mm), and the inlet temperature was set at 280 °C. The temperature ramps were as follows: initial temperature was 70 °C for 1 min; 180 °C at a rate of 15 °C/min (hold 2 min); 230 °C at a rate of 10 °C/min (hold 0.5 min); 250 °C at a rate of 5 °C/min (hold 2 min); and 300 °C at a rate of 8 °C/min (hold 5 min). Mass spectrometry was performed using an electron bombardment ionization source (electron impact: 70 eV) at an ion source temperature of 250 °C, interface temperature of 280 °C, and acquisition range of 50–550 aum.

Quality control

Target compounds were analyzed in MassLynx V4.2 (Waters, USA). Qualitative analysis of PACs was completed using reference substance retention time (RT) and fragment ion data, combined with mass spectrograms in the 2017 NIST Mass Spectral Library (NIST 17) and biomarker-related books.

Strict quality control was performed with 5 and 10% of the total samples representing the quality control and blank samples, respectively, and was adapted to confirm the analysis reliably. The carryover showed no detectable PAH contamination, and method verification met the experimental requirements. PAC concentrations of lower than the minimum detection limits (MDLs) were replaced by half of the MDLs (Qian et al., 2022c). The sample recoveries ranged from 91.2 to 125.5%, and the standard deviation of the sample duplicates from 2.1 to 14%.

Statistical analysis

The PAC quantitative analysis was done using by MassLynx V4.2 (Waters, USA), and the concentrations and molecular composition were statistically analyzed using Origin Software version 9.0 (Origin Lab Inc., Northampton, USA) and IBM SPSS (Armonk, New York, USA). The results are presented as means and standard deviation of the mean, and the difference between groups was considered significant at $p < 0.05$. The PAC spatial distributions were mapped using ArcGIS (Ver. 10.2; ESRI, RedLands, USA) with the inverse distance weighted (IDW) spatial interpolation method. The PAC sources were identified using the positive matrix factorization (PMF) and principal component analysis (PCA) methods. The carcinogenic risk was assessed using the cancer risk (CR) method as recommended by the US EPA (supplementary material).

Results and discussion

Concentrations of PACs

Based on the qualitative results of 30 soil samples, in addition to 16 PAHs, abundant APAHs were identified, including alkyl naphthalene (ANAPs), alkyl phenanthrene (APHEs), 2-M-ANT, C1-FLU,

C1-FLA, C1-PYR, and 7-M-Bap. ANAPs and APHEs had the most homologues (Fig. S1). The range of $\sum$PACs (sum of polycyclic aromatic compounds) in the soil samples was from 300 to 17,599 µg kg^{-1} ($n = 30$). The mean concentrations of soil PACs in different functional areas differed as follows: coal mine (9107 µg kg^{-1}) > Coal fire (3163 µg kg^{-1}) > agricultural (1232 µg kg^{-1}) > background areas (710 µg kg^{-1}) (Fig. 2). As shown in Fig. 2, the concentrations of $\sum$APAHs (sum of APAHs) in the soil samples were significantly higher than those of $\sum$16 PAHs (sum of 16 PAHs), accounting for 60–81% of the mean value of PACs. Additionally, alkyl naphthalenes (ANAPs) and alkyl phenanthrenes (APHEs) were the main contributors (80–90%) to APAHs.

The mean concentration of $\sum$APAHs in the coal mine soil was 6236 µg kg^{-1}, 2.2 times that of $\sum$16 PAHs (2871 µg kg^{-1}). In the coal fire areas, the mean concentrations of $\sum$APAHs and $\sum$16 PAHs were 1886 and 1277 µg kg^{-1}, respectively. The coal fire and coal mine areas were relatively more polluted with the 16 PAHs 6.7 times and 14.8 times that of the background area, respectively. The PAC levels in the agricultural area were relatively low with the mean concentrations of $\sum$APAHs and $\sum$16 PAHs being 902 and 477 µg kg^{-1}, respectively. In the background area, the mean concentrations of $\sum$APAHs and $\sum$16 PAHs were 573 and 195 µg kg^{-1}, respectively.

Studies on 16 PAHs in the soil are quite common with study area types mainly including urban (Liu et al., 2019; Thiombane et al., 2019), suburban (Ma et al., 2005; Thiombane et al., 2019), agricultural (Wang et al., 2010; Zheng et al., 2019), and industrial areas (Idowu et al., 2020; Jiang et al., 2016; Li et al., 2017a, 2017b). However, there are few studies on the 16 PAHs in the surface soil of coal mining areas and even fewer studies on PACs in coal fire areas. The mean concentration of the 16 PAHs in the surface soil around the coal mine area in previous studies ranged from 256 to 1886 µg kg^{-1}, lower than that in the Wuda coal mine area (2871 µg kg^{-1}) (Table S1). The concentration of the 16 PAHs in agricultural areas was relatively low, ranging from 178 to 1910 µg kg^{-1}. Among them, China, South Korea, and Poland had the lowest concentration levels of the 16 PAHs, while Delhi, India, had the highest with an average of 1910 µg kg^{-1} (Table S1). However, there remains a lack of research on APAH soil contamination.

Molecular composition of PACs

NAP and PHE, the two most abundant PAHs in the soil samples, accounted for 10–22% and 20–27% of

Fig. 2 Means and standard deviation of the mean of 16 PAHs, ANAPs + APHEs, APAHs, and PACs, as well as the proportion of ANAPs + APHEs and APHs in the soil of different functional areas
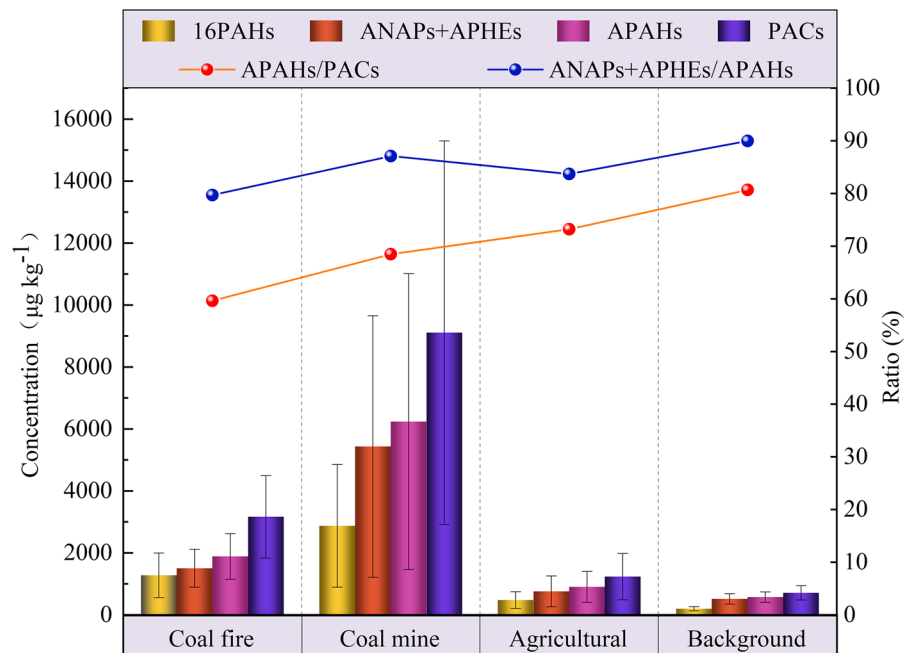
the total, respectively. CHR, FLA, PYR, and FLU were all detected while ACY, ACE, ANT, and DBA had the lowest detection frequencies (Table S2). Only six parent PAHs were detected in the background area soil (NAP, FLU, PHE, FLA, PYR, and CHR) and the high molecular weight 16 PAHs (5 and 6 rings) were lower than the MDLs. Eleven PAHs were detected in the agricultural area, but of these ACY, ACE, ANT, InP, and DBA were all lower than the MDLs. Besides InP, other PAHs were detected in the coal fire areas, whereas all 16 PAHs were detected in the coal mine areas (Table S2). ANAPs and APHEs were predominant in soils of different functional areas. C4-NAP was not detected in coal fire areas, while C4-NAP and C1-CHR were not detected in agricultural areas, and all the components of APAHs were detected in other areas. In addition, C1-NAP, C2-NAP, and C2-PHE had the highest concentration in the coal fire areas, accounting for 19%, 22%, and 16% of $\sum$APAHs, respectively. C2-NAP, C1-PHE, C2-PHE, and C3-PHE accounted for 18%, 9%, 28%, and 9%, respectively, in the coal mine areas. C1- NAP, C2-NAP, C2-PHE, and C3-PHE were dominant in the agricultural and background areas, accounting for 14%, 21%, 19%, and 11% in the agricultural area and 11%, 17%, 33%, and 12%, respectively, in the background area (Table S2).

As shown in Fig. 3a, the 16 PAHs in the soil of the coal mine area are mainly 3 and 4 rings, while the other areas are mainly of low molecular weight (2–3 rings). The proportion of low-ring (2–3 rings) PAHs in the agricultural and background area was slightly

higher at 63% and 69%, respectively. In contrast, this proportion in coal fire and coal mine areas was slightly lower at 53% and 37%, respectively. Additionally, the soils of the Wuda District were dominated by low-ring APAHs, accounting for 86–92% (Fig. 3b). Previous studies have shown that APAHs and the 16 PAHs are prevalent in coal and gangue (Misz-Kennan & Fabiańska, 2010; Stout & Emsbo-Mattingly, 2008). Laumann et al. (2011) showed that ANAPs and APHEs were abundant in coal from different sources and grades. This is consistent with the composition characteristics of APAHs in the surface soil of coal mine and coal fire areas, indicating that coal and coal fires mainly affected soil pollution by PACs.

Soil is the secondary pollution source of low-ring PAHs and the repository of high-ring PAHs (Bozlaker et al., 2008; Wang et al., 2008a; Zhang et al., 2011). Most low-ring PACs have low boiling points and relatively low K_{ow}, and their soil concentrations are easily affected by environmental factors, such as light, rainfall, airflow, and other factors, which can aid in long-distance atmospheric transport (Augusto et al., 2010). Compared with high-ring PACs, low-ring PACs are more likely to enter the atmosphere from coal or coal gangue and return to the soil and water through wet and dry deposition (Liu et al., 2013). The proportion of low-ring PACs in the Wuda District soil was high, indicating that the PACs came from the surrounding activities, such as coal mining and coal gangue stacking (petrogenic sources). Once PACs are discharged from coal and coal gangue, low-ring PACs are more
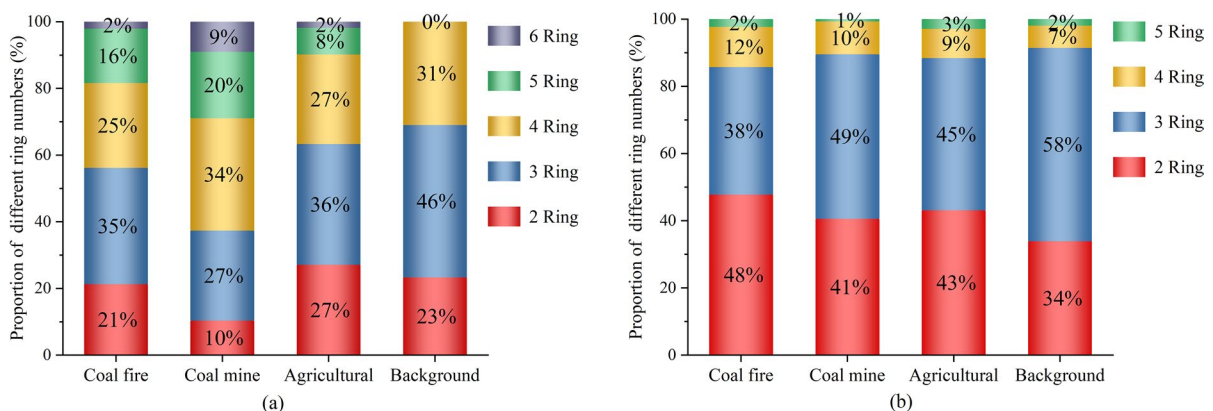
Fig. 3 Percentage distribution with different ring numbers of 16 PAHs and APAHs. **a** The proportion of the 16 PAHs with different ring numbers, **b** the proportion of APAHs with different ring numbers

likely to be enriched in the surface soil at a longer distance compared with high-ring PACs. This could explain why low-ring PACs dominated the PAC pollution in the agricultural and background areas.

Spatial distribution of PACs

The spatial distributions of $\sum$PACs, $\sum$APAHs, and $\sum16$ PAHs were mapped using ArcGIS (ESRI, USA) with the IDW spatial interpolation method (Fig. 4). The spatial distribution patterns of $\sum$PACs, $\sum$APAHs, and $\sum16$ PAHs are essentially the same. The concentration distribution shows that the coal mine and coal fire areas are more polluted, while the agricultural and background areas are less polluted. The pollution level of PACs in the soil near the coal fire point is slightly lower than in the coal mine area without a coal fire. This may be because the low-ring PAHs in the soil from coal fire areas are released into the atmosphere under the influence of high temperatures, thus reducing the content of PACs in the soil. Additionally, there is a perennial northwest wind in the Wuda District with PAC concentrations from northwest to southeast showing a stepwise decreasing trend (Fig. 4). Furthermore, by comparing Fig. 4c, d, the concentrations of APAHs are markedly higher than that of the 16 PAHs. If the concentrations of the conventional 16 PAHs are used as the evaluation standard, the pollution degree of the study area will be underestimated.

The pollution grade evaluation of the 16 PAHs divided soil pollution into four grades: uncontaminated ($\sum16$ PAHs < 200 μg kg^{-1}), weakly contaminated (200 μg kg$^{-1} \leq \sum16$ PAHs < 600 μg kg^{-1}), moderately contaminated (600 μg kg$^{-1} \leq \sum16$ PAHs < 1000 μg kg^{-1}), and heavily contaminated ($\sum16$ PAHs > 1000 μg kg^{-1}) (Maliszewska-Kordy-bach, 1996). The coal fire and coal mine areas have the highest proportion of heavily contaminated points, accounting for 80% and 50% respectively, with no uncontaminated points (Fig. S2). Of the sampling points, 50% and 60% in the agricultural and background areas, respectively, were weakly contaminated; the uncontaminated points accounted for 17% and 40%, respectively; no heavily polluted points (Fig. S2). Previous studies have shown that APAHs are more biotoxic than 16 PAHs (Fallahtafti et al., 2012; Honda & Suzuki, 2020; Krzyszczak & Czech, 2021). Hence, focusing on conventional 16 PAHs is

not a comprehensive environmental pollution assessment. APAHs should also be included in the risk assessment of PACs.

The Wuda industrial park is south of the coal mine area (Fig. 1b). Our previous work has shown that the surface soil of the Wuda Industrial Park was seriously polluted by PACs, with an average value of 10,176 μg kg^{-1} (Xu et al., 2022). This is probably due to the unique geographical location of the industrial park. Wushu Mountain is south of the coal mine area and north of the industrial park (Fig. 1b). On the one hand, Wuhu Mountain blocks part of the northwest wind blowing from the direction of the desert all year round, and the weak "foehn effect" also makes it easier for the gas and solid PACs released from the coal fire and coal mine areas to collect into the soil of the industrial park through atmospheric sedimentation. On the other hand, this could probably be related to soil properties and occurrence conditions. Coal mine and coal fire areas are adjacent to deserts. After field investigation, it was found that part of the surface soil in the mining area has suffered from severe desertification due to the lack of vegetation cover protection and the impact of long-term coal mining. The natural environmental conditions are harsh. The sandy soils are not conducive to PAC accumulation. Although there is serious coal mining and spontaneous combustion of coal fires, the content of PACs in the soil is not as high as in the industrial park.

Identification of PAC sources

The study area has typical coal fires, and the conventional ratio method is unsuitable. Positive matrix factorization (PMF) is a multivariate statistical analysis method with its application trend in soil environment research increasing in recent years (Liang et al., 2019; Wang et al., 2017, 2022; Yusuf et al., 2022). PMF analysis was conducted on 16 PAHs and APAHs in the soil of coal fire, coal mine, and agricultural areas where a total of four factors were extracted for source analysis (Fig. 5). Factor 1 was identified as the petrogenic source with a contribution rate of 37.4%. The low-ring 16 PAHs and most APAHs were predominant, including FLU, PHE, ANT, NAPs, APHEs, C1-FlU, C1-CHR, C1-FLA and C1-PYR. Previous studies have shown that low-ring PAHs come from petrogenic sources (Jordi Dachs et al., 2002; Wang et al., 2008b, 2015b, 2022; Zhang et al., 2019). At

Fig. 4 Spatial distribution of **a** ∑PACs, **b** ∑APAHs, and **c** ∑16 PAHs in the Wuda District soil
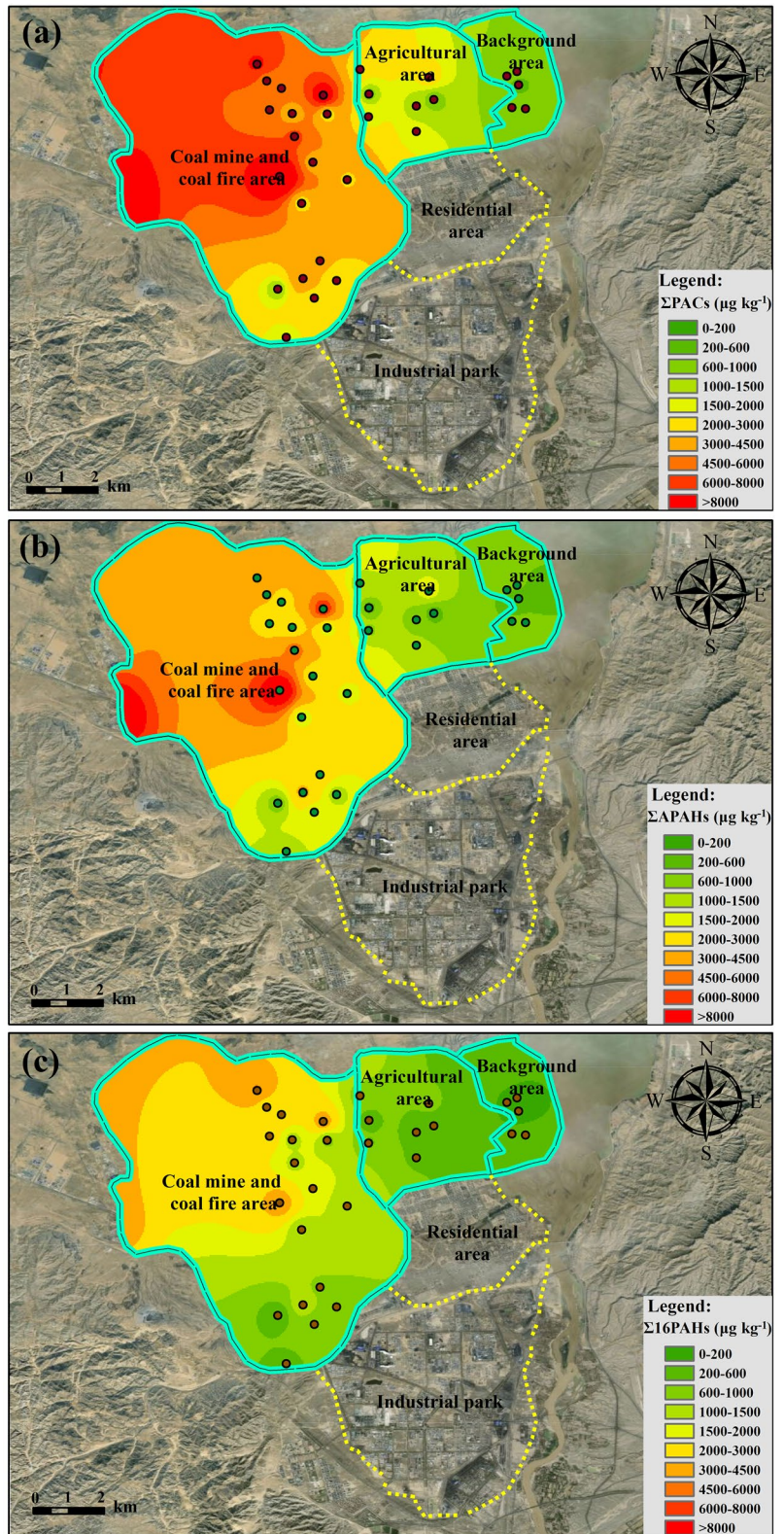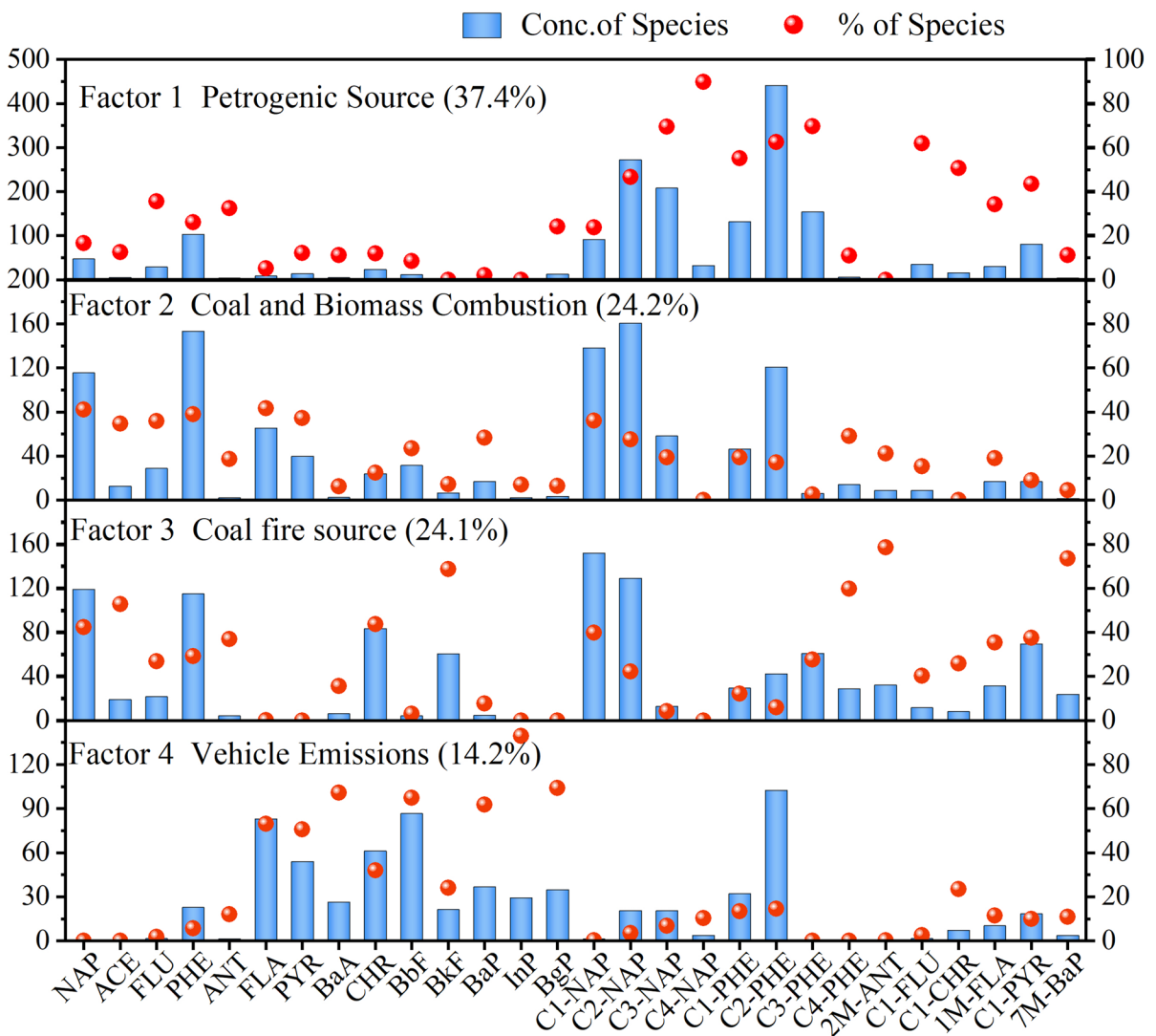
Fig. 5 Source profiles and relative contribution of soil PACs obtained from the PMF model

the same time, 1-M-NAP and 2-M-NAP also contribute to these petrogenic sources (Yunker et al., 2002). Coal and coal gangue of different coal ranks where low-ring PAHs and APAHs dominate (Chen et al., 2022; Laumann et al., 2011). This is consistent with the actual situation in Wuda District. The exposed strata in the Wuda coalfield include Carboniferous (Benxi Formation and Taiyuan Formation), Permian, and Tertiary units with the Taiyuan Formation strata being the main exposed stratum (Li et al., 2018). In addition, ANAPs and APHEs in Factor 1 show a "bell-shaped" distribution, indicating petrogenic sources and inferring that coal or coal gangue

particles are likely to be prevalent in the Wuda District surface soils (Hindersmann & Achten, 2018; Hindersmann et al., 2020).

Factor 2 was identified as coal and biomass combustion, accounting for a contribution rate of 24.2%. NAP, ACE, FLU, PHE, FLA, and PYR had higher loads. Low-ring PACs mainly indicate biomass combustion with PHE, FLU, and ANT usually being released by this method (Wang et al., 2015a; Zhao et al., 2014). NAP is produced during incomplete combustion (Simcik et al., 1999), ACE is a sign of biomass burning, including wood (Biache et al., 2014), and FLA and PYR can be used as tracers for

coal combustion (Callen et al., 2014; Chao et al., 2019; Zhang et al., 2019).

Typical coal fires exist in the Wuda District; however, there is a dearth of research on PAC data in these areas. This study proposes a group of characteristic factors of coal fires using PCA and PMF methods. Meanwhile, this group of characteristic factors should be different from industrial coal burning and coal coking. Factor 3 is presumed to be a typical coal fire source, with a contribution rate of 24.1%. Firstly, PCA was used to analyze the soil samples of the coal fire and coal mine areas, and the two types of samples are well distinguished (Fig. S3a). Additionally, it can be seen from the factor analysis diagram of soil sample PACs that these components are divided into three groups: low-ring 16 PAHs and their alkyl substituents are petrogenic sources; high-ring PAHs (those 4 and above) are pyrolysis sources; and BkF, 2-M-ANT, and 7-M-BaP are characteristic coal fire sources, which are likely to be characteristic factors of coal fire source (Fig. S3b). Combined with the results of the PMF analysis, the main compounds of Factor 3 were BkF, 2-M-ANT, and 7-M-BaP, followed by ACE, CHR, and C4-PHE, while BkF and CHR are typical pyrolysis source products (Callen et al., 2014; Chao et al., 2019; Zhang et al., 2019). Wang et al. (2013) reported ANT as a marker of coal burning with 2-M-ANT being a methylated derivative of ANT, which is generated in the presence of an oxidizing agent. Sulfur and metal elements are oxidized during coal combustion, conducive to ANT methylation (Qian et al., 2022b). By comparison, the high-contribution characteristics of Factor 3 in coal fire areas are completely different from each group in the Wuda Industrial Park soils (Xu et al., 2022). Therefore, it is speculated that the high contribution of these compounds may be related to coal fires, necessitating further research for verification.

Factor 4 was identified as vehicle emissions, contributing 14.2% of the total PAC sources with the main compounds including FLA, PYR, BaA, CHR, BbF, BaP, InP, and BgP. Among these, CHR, BbF, BaP, and InP are usually the indexes of vehicle exhaust emissions (Ho et al., 2009; Wang et al., 2013, 2022); BbF, BgP, and InP represent gasoline vehicle emissions (Agudelo-Castañeda & Teixeira, 2014; Taghvaee et al., 2018); BaA is a diesel engine marker (Wang et al., 2015a; Yang et al., 2013); and CHR and FLA are mainly related to diesel combustion (Kwon

& Choi, 2014). Both BaP and InP have been identified as typical tracers of PAH vehicle emissions (Larsen & Baker, 2003; Taghvaee et al., 2018).

Petrogenic sources accounted for the highest proportion at 37.4%, consistent with the actual situation of mining and transportation activities in coal mining areas. In coal mining, organic pollutants may be released through coal-washing wastewater, coal ash, and coal overflow during vehicle transportation (Huang et al., 2016). In addition, there are large coal gangue mountains in the mining area. As one of the primary solid wastes, coal gangue may also be the main source of PACs in the surface soil of the study area (Sun et al., 2009). After weathering, the PACs of coal and coal gangue will gradually escape into the surrounding soil, resulting in soil PAC pollution (Zhang et al., 2020). In addition, it has been shown that the proportion of low-ring PAHs may be higher than HMW-PAHs in the atmospheric environment, which is more likely to enable long-distance transport (Augusto et al., 2010). Once PACs are discharged from coal and coal gangue, low-ring PACs are more likely to be enriched in the surface soil than high-ring PACs due to their longer transmission distance.

Risk assessment of PAHs in dust

Cancer risk (CR) is commonly used as a potential carcinogenic outcome of exposure to describe the likelihood of cancer. In this study, three exposure methods were used to calculate the carcinogenic risk of 16 PAHs (Table 1). The parameters used for incremental life cycle cancer risk assessment are shown in Table S3. Detailed formulas for cancer risk (CRs) are shown in the attached table. According to the United States Environmental Protection Agency, cancer risk assessment is divided into three grades: negligible risk ($< 10^{-6}$), potential risk (10^{-6}–10^{-4}), and high risk ($> 10^{-4}$).

As shown in Table 1, the CRs of all samples were higher than 10^{-4}, indicating that there was no high carcinogenic risk area in the study area. The CR_{total} of the different areas is shown as follows: coal mine > coal fire > agriculture > background areas. The highest cancer risk (CR_{total}) was 6.43×10^{-6} in the coal mine area. For adults, only the coal mine area had a CR_{total} in the potential risk range with the rest being negligible. The mean values of CRs for children exposed to the three

Table 1 Cancer risks for local children and adults on exposure to $\sum 16$ PAH via various pathways

Area	CR	Child Mean	Max	Min	Adult Mean	Max	Min
Coal fire	CR_{ing} [a]	4.59E−07	8.35E−07	2.89E−07	6.01E−08	1.09E−07	3.79E−08
	CR_{derm} [b]	1.54E−06	2.80E−06	9.70E−07	4.11E−07	7.48E−07	2.59E−07
	CR_{inh} [c]	6.76E−12	1.23E−11	4.26E−12	2.33E−11	4.25E−11	1.47E−11
	CR_{total} [d]	2.00E−06	3.64E−06	1.26E−06	4.71E−07	8.57E−07	2.97E−07
Coal mine	CR_{ing}	1.48E−06	3.05E−06	2.79E−07	1.94E−07	4.00E−07	3.66E−08
	CR_{derm}	4.95E−06	1.02E−05	9.38E−07	1.32E−06	2.73E−06	2.50E−07
	CR_{inh}	2.18E−11	4.50E−11	4.12E−12	7.50E−11	1.55E−10	1.42E−11
	CR_{total}	6.43E−06	1.33E−05	1.22E−06	1.52E−06	3.14E−06	2.87E−07
Agricultural	CR_{ing}	3.29E−07	4.20E−07	2.79E−07	4.31E−08	5.51E−08	3.66E−08
	CR_{derm}	1.10E−06	1.41E−06	9.37E−07	2.94E−07	3.76E−07	2.50E−07
	CR_{inh}	4.85E−12	6.19E−12	4.11E−12	1.67E−11	2.14E−11	1.42E−11
	CR_{total}	1.43E−06	1.83E−06	1.22E−06	3.38E−07	4.32E−07	2.87E−07
Background	CR_{ing}	2.80E−07	2.80E−07	2.79E−07	3.66E−08	3.68E−08	3.65E−08
	CR_{derm}	9.38E−07	9.41E−07	9.35E−07	2.50E−07	2.51E−07	2.50E−07
	CR_{inh}	4.12E−12	4.13E−12	4.11E−12	1.42E−11	1.43E−11	1.42E−11
	CR_{total}	1.22E−06	1.22E−06	1.21E−06	2.87E−07	2.88E−07	2.86E−07

[a] Cancer risk through ingestion of soil particles

[b] Cancer risk from dermal contact

[c] Cancer risk from inhalation

[d] Cancer risk from all three pathways combined

kinds of exposure were generally higher than those of adults with a CR_{total} in the range of 10^{-6}–10^{-4}, indicating that the study area had potential carcinogenic risks for children. The CR for dermal contact (CR_{derm}) was also the highest, while the inhalation pathway (CR_{inh}) was lowest in both children and adults.

Previous studies have shown that APAHs are more toxic and persistent than their parent PAHs (Fallahtafti et al., 2012; Kang et al., 2016; Mu et al., 2014) with the results of this study showing that APAHs in soil are the main contributors to PACs. APAHs were not involved in the CR calculation due to a lack of research on the toxicity equivalent factors (TEFs) of APAHs; therefore, the PAC levels and ecological risks may be underestimated. If future PAC emissions are not controlled at sources, the cancer risk may continue to increase. However, the EPA has not listed APAHs as priority pollutants, primarily due to a lack of reliable and accurate analytical methods for routine identification and quantification of these in samples from complex environments. Moreover, APAHs have a wide structural diversity compared to their parent PAHs, and it isn't easy to obtain all APAHs standards.

Conclusions

There were abundant APAHs and 16 PAHs in the soil of the Wuda District, and the concentration of APAHs was significantly higher than that of the 16 PAHs. The mean concentrations of APAHs accounted for 60–81% of PACs. The mean concentration of PACs in different functional areas differed: coal mine (9107 µg kg^{-1}) > coal fire (3163 µg kg^{-1}) > agricultural (1232 µg kg^{-1}) > background areas (710 µg kg^{-1}). Additionally, there are mainly low-ring PAHs and APAHs in the surface soil of the Wuda District with marked petrogenic characteristics, presumed to be caused by coal mining and the escape of coal gangue after weathering. The sources of PACs in soil were analyzed by PMF combined with PCA with the results showing that the PACs were mainly derived from petrogenic sources, coal and biomass combustion, coal fires, and vehicle emissions. The cancer risk assessment revealed that part of the coal mine area was at a potential cancer risk level. The data collected have highlighted potential carcinogens emanating from coal mines, which could be extrapolated and tested in different coalfields to check if similar patterns are visible. This would help create the right approaches

toward tackling the ecological and health challenges arising from coal mining.

Data availability The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors declared no conflict of interest.

Consent to participate and consent to publish Consent for publication was obtained from all participants.

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