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Predication of the sources of particulate polycyclic aromatic hydrocarbons in China with distinctive characteristics based on multivariate analysis

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ABSTRACT

Particulate polycyclic aromatic hydrocarbons (PAHs) have the characteristics of high carcinogenicity, degradation resistance, and long-distance transport. It is necessary to determine the critical sources that contribute to PAHs in atmospheric particulate matter (PM), soils, and sediments. In the present study, particulate PAHs were collected from Chinese cooking, biomass combustion, waste incineration, coal combustion, and petroleum combustion in different regions of the Chinese mainland. Through linear regression analysis, the relationships have been investigated between different measurement units, including the mass of particulate PAHs per mass of particle matter, mass of particulate PAHs per volume of air, and emission factors of particulate PAHs. This information helps to effectively evaluate the contamination level of PAHs among different studies. On the scale of the entire Chinese mainland, the top three sources of particulate PAHs are industrial coal combustion (11,244 t/year, 44.45%) > domestic coal combustion (7,797 t/year, 30.82%) > straw combustion (6,015 t/year, 23.78%). Efforts should be made to develop clean coal utilization. Although cooking contributes to a small amount of PAHs in China, it is extremely harmful for cooks [benzo(a)pyrene: 1.64 $\mu g/m^3$]. Principal component analysis (PCA) is an effective method to identify various pollution sources. Using PCA, Chinese cooking, biomass combustion, waste incineration, domestic coal combustion, coking coal combustion, coal combustion for power generation, and diesel/gasoline engines that possess distinct PAH compositions are effectively discriminated from one another. Moreover, the characteristics of particulate PAHs emitted from human activities in China are not totally consistent with those in other countries. It is critical for pollution control to determine the distinctive characteristics of particulate PAHs emitted from typical production processes and life in China.

1. Introduction

Particulate polycyclic aromatic hydrocarbons (PAHs) that are linked to skin, lung, bladder, liver, and stomach cancers have been identified as priority substances (ATSDR, 1995; CONTAM, 2008; Keith, 2015). PAHs are ubiquitous in environmental media and can be formed from either natural or human-activity-related sources. These hydrocarbons are primarily contributed by incomplete combustion (Zhang et al., 2013, 2015; Zhang and Fan, 2016). Anthropogenic sources, including fossil fuel (coal and petroleum products) combustion, biomass fuel (straw residues and wood) combustion, incineration of municipal waste and hospital waste,

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and oil frying, significantly enhance the contamination of PAHs in environmental media (Lee et al., 2005; Li et al., 2003; See et al., 2006; See and Balasubramanian, 2008; Shi et al., 2009). PAHs emitted from these anthropogenic sources have drawn the attention of many environmentalists and regulatory agencies, as well as our research group (Zhang et al., 2013, 2015; Zhang and Fan, 2016).

In general, there are two types of studies on emissions of PAHs. A fraction of studies collect both gaseous and particulate PAHs using gas traps or sorbents to evaluate the total PAHs in the air. In contrast, the others solely collect particulate PAHs using only filters (Li et al., 2003). High-molecular-weight PAHs (HPAHs) are preferentially associated with particle matter (PM). HPAHs pose higher carcinogenic potential to humans by inhalation than do the low-molecular-weight PAHs (LPAHs) associated with the gaseous phase (Iordanidis et al., 2008; Kong et al., 2011; Orecchio and Papuzza, 2009). Furthermore, emitter of HPAHs are critical sources of contamination by PAHs in atmospheric PM, soils, and sediments because of their degradation resistance and long-distance transport (Iordanidis et al., 2008; Wang et al., 2012). Therefore, it is of great importance to study the concentrations and characteristics of particulate PAHs emitted from various anthropogenic sources, especially on the Chinese mainland, where atmospheric particulate pollution is one of the serious environmental problems in recent years.

The isomer ratios (e.g., LPAHs to HPAHs and anthracene to the sum of phenanthrene and anthracene) have been widely used to characterize and distinguish petrogenic PAHs from pyrogenic PAHs (Zhang et al., 2013, 2015). However, the ratios fail to identify specific sources, such as cooking, biomass combustion, waste incineration, coal combustion, and petroleum-product-based combustion. Compared with the isomer ratio method, the results from receptor models based on multivariate analysis are more robust and can provide more information on contamination sources by statistically analyzing the entire dataset (Li et al., 2003; Zhang et al., 2013). The CANOCO 4.5 software bundled with CanoDraw for Windows (Microcomputer Power, Ithaca, USA) can be used to perform multivariate analysis and to describe the structure in a single data set (Lepš; Šmilauer, 2003; Ter Braak and Smilauer, 2002). There is a lack of comprehensive studies on the contamination level and characteristics of PAHs emitted from various sources relating to typical production processes and life in China.

Therefore, the objective of this study is to collect published data, to build the relationships between different measurement units of particulate PAHs, and to effectively evaluate the contamination level of particulate PAHs from various combustion sources in different regions of the Chinese mainland. The study also aims to distinguish PAHs emitted from eight specific anthropogenic sources using multivariate analysis. This is a greater number of sources than in the previous studies. Simultaneously, we aim to determine the characteristics of particulate PAHs emitted from these sources. Effective pollution control is primarily based on precise source analysis, which necessitates knowing the distinctive composition of PAHs emitted from various sources and more-robust source-analysis methods, such as multivariate analysis.

2. Materials and methods

2.1. Data collection

Published data on composition of USEPA's sixteen PAH priority pollutants emitted from various anthropogenic sources on mainland China were collected. These 16 pollutants were naphthalene (Nap), acenaphthylene (Any), acenaphthene (Ane), fluorene (Fle), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenzo(a,h)anthracene (DaA), benzo(g,h,i)perylene (BgP), and Indeno(1,2,3-c,d)pyrene (I1P). The anthropogenic sources and the references for them included in this study are given in the Supplementary Material.

The P% values are defined as the percentage of the particulate PAHs in the total particulate and gaseous PAHs. These values were used to calculate the concentrations of particulate PAHs when the concentrations of total PAHs were given. The P% summarized by Li et al. (2003) was used to measure particulate PAHs from Chinese cooking due to the lack of data on P% for Chinese cooking. For biomass combustion, the average P% values were calculated based on Lu et al. (2009), Shen et al. (2011, 2012a), and Zhang et al. (2011). The P% values determined by Hu et al. (2015) were used to calculate the concentrations of particulate PAHs from various engines in Hangzhou without the corresponding values (Zhu et al., 2003).

2.2. Linear regression analysis

In previous studies on PAHs, various methods and measurement units were used. Therefore, the relationships between the mass of particulate PAHs per mass of particle matter (C_{PM}), mass of particulate PAHs per volume of air (C_{air}), and emission factors for particulate PAHs (EF_{PAHs}) were built through linear regression analysis using Excel and were based on data from the previous studies in order to compare emissions of PAHs from different sampling sites and different anthropogenic sources. The linear regression equations and the sources of data are presented in Table 1. PAHs emitted from Chinese cooking in Singapore (See et al., 2006; See and Balasubramanian, 2008) were used to build the relationship between $C_{PM\text{-}cooking}$ and $C_{air\text{-}cooking}$ (E1 in Table 1) due to the limited data available from mainland China. For the same reasons, PAHs from industrial coal combustion in Taiwan (Yang et al., 1998, 2002) were used to build the relationships between $EF_{PAHs\text{-}industrial\ coal}$ and $C_{PM\text{-}industrial\ coal}$ (E5 in Table 1). The relationship (E6, E7 and E8 in Table 1) between $C_{air\text{-}engine}$ and both $EF_{PAHs\text{-}engine}$ and $C_{PM\text{-}engine}$ were also developed based on PAHs from engines collected from both the Chinese mainland (Cui et al., 2017; He et al., 2010; Li et al., 2013; Lu et al., 2012; Wang et al., 2016) and Taiwan (Lin et al., 2006, 2013; Mi et al., 1996, 2001; Wu et al., 2010). The $EF_{PAHs\text{-}tunnel}$ and $C_{air\text{-}tunnel}$ (Ho et al., 2009) in Hong Kong were also used to build the relationships between them (E10 in Table 1) in this study. PAHs emitted from other countries were not used because the cooking style and the quality of fuels may affect the relationships between different measurement units.

Generally, the national emissions of particulate PAHs from different sources were calculated based on the average EF_{PAHs} values and fuel consumption in 2015 (Table S1). According to Yao et al. (2015), the EF_{PAHs} from frying was computed (79.44 μg/kg cooking oil). For straw combustion, the average EF_{PAHs} (60.15 mg/kg) from combustion in rural stoves in Beijing (Shen et al., 2011) and Nantong (Wei et al., 2014) was used. Assuming that ash production is 30% of the incinerated wastes, the amounts of ash from municipal waste incinerators (MWI) and hospital waste incinerators (HWI) are 1.86×10^7 t and 2.1×10^4 t in 2015, respectively (Lynn et al., 2017). The average concentrations of PAHs in MWI and HWI ash were used to calculate the national emissions of particulate PAHs from waste incinerators.

2.3. Principal component analysis

Before PCA, the concentrations of the 16 priority PAHs of the USEPA were normalized to that of BaP for all samples collected from Chinese cooking, biomass combustion, waste incineration,

Table 1
Relationship between PAH concentrations in two forms and emission factors.

Equations		References
$C_{PM\text{-}cooking} = 4.093C_{air\text{-}cooking}$ (E1)	$R^2 = 0.988$, n = 7	He et al., 2004; See et al., 2006, See and Balasubramanian, 2008
$C_{PM\text{-}biomass} = 121.6EF_{PAHs\text{-}biomass}$ (E2)	$R^2 = 0.619$, n = 32	Shen et al., 2010c, 2011, 2012a, 2012b; Wei et al., 2014
$C_{PM\text{-}domestic\ coal} = 76.38EF_{PAHs\text{-}domestic\ coal}$ (E3)	$R^2 = 0.849$, n = 19	Bi et al., 2008; Chen et al., 2005; Huang et al., 2014
$C_{PM\text{-}industrial\ coal} = 0.270C_{air\text{-}industrial\ coal}$ (E4)	$R^2 = 0.914$, n = 11	Mu et al., 2014; Wang et al., 2015
$EF_{PAHs\text{-}industrial\ coal} = 0.153C_{air\text{-}industrial\ coal}$ (E5)	$R^2 = 0.883$, n = 5	Mu et al., 2013a; Yang et al., 1998, 2002
$EF_{PAHs\text{-}engine} = 10.987C_{air\text{-}engine}$ (E6)	$R^2 = 0.778$, n = 25	He et al., 2010; Lin et al., 2006; Mi et al., 1996, 2001; Wang et al., 2016; Wu et al., 2010
$C_{PM\text{-}diesel\ engine} = 9.533C_{air\text{-}diesel\ engine}$ (E7)	$R^2 = 0.866$, n = 12	Cui et al., 2017; Li et al., 2013; Lin et al., 2006; Lu et al., 2012; Wu et al., 2010
$C_{PM\text{-}gasoline\ engine} = 1005C_{air\text{-}gasoline\ engine}$ (E8)	$R^2 = 0.782$, n = 10	Li et al., 2013; Lin et al., 2013; Wang et al., 2016
$C_{PM\text{-}tunnel} = 1.477C_{air\text{-}tunnel}$ (E9)	$R^2 = 0.999$, n = 8	Chen et al., 2013b; Dai et al., 2015; He et al., 2006, 2008; Liu et al., 2015; Wang et al., 2007
$EF_{PAHs\text{-}tunnel} = 0.918C_{air\text{-}tunnel}$ (E10)	$R^2 = 0.969$, n = 10	Chen et al., 2013b; Cui et al., 2016; Dai et al., 2015; He et al., 2008; Ho et al., 2009; Liu et al., 2015; Wang et al., 2007

$C_{PM\text{-}cooking}$, $C_{PM\text{-}biomass}$, $C_{PM\text{-}domestic\ coal}$, $C_{PM\text{-}industrial\ coal}$, $C_{PM\text{-}diesel\ engine}$, $C_{PM\text{-}gasoline\ engine}$, and $C_{PM\text{-}tunnel}$, mass of PAHs per mass of particle matter from Chinese cooking, biomass combustion, domestic coal combustion, industrial coal combustion, diesel engines and gasoline engines, and traffic tunnels, mg/kg; $C_{air\text{-}cooking}$, $C_{air\text{-}industrial\ coal}$, and $C_{air\text{-}tunnel}$, mass of PAHs per volume of air from cooking, industrial coal combustion, and traffic tunnels, ng/m^3; $C_{air\text{-}diesel\ engine}$ and $C_{air\text{-}gasoline\ engine}$, mass of PAHs per volume of air from diesel engines and gasoline engines, μg/m^3; $EF_{PAHs\text{-}biomass}$ and $EF_{PAHs\text{-}domestic\ coal}$, emission factors of particulate PAHs from biomass combustion and domestic coal combustion, mg/kg; $EF_{PAHs\text{-}industrial\ coal}$ and $EF_{PAHs\text{-}engine}$, emission factors of particulate PAHs from industrial coal combustion and diesel/gasoline engines, μg/kg; $EF_{PAHs\text{-}tunnel}$, emission factors of particulate PAHs from traffic tunnels, μg/vehicle/km.

domestic coal combustion, coking coal combustion, coal combustion for power generation, diesel engines, and gasoline engines. BaP is detected in all samples and normalization of PAHs to BaP prior to PCA analysis is necessary because the types and analytical methods for PAHs from various sources vary (Li et al., 2003). The normalized PAHs in these samples were subjected to PCA and are presented in Table S2. PCA was performed using CANOCO 4.5 software bundled with CanoDraw for Windows (Microcomputer Power, USA) to explore the similarities and differences in the composition of particulate PAHs in samples collected from the eight different sources (Lepš; Šmilauer, 2003; TerBraak and Šmilauer, 2002). In PCA, logarithmic transformation of data was performed to cause the data to distribute in a nearly normal fashion.

3. Results and discussion

3.1. PAHs emitted from Chinese cooking, biomass combustion, and waste incineration

In the four studied cities (Fig. 1a), the concentrations of particulate PAHs from charcoal broiling in the laboratory are lowest (8.98 mg/kg PM, 2.16 ng/m^3), while those from oil frying in the laboratory are highest (43,500.14 mg/kg PM, 10.48 μg/m^3). The national emission of particulate PAHs from oil frying is approximately 2.5 t/year. The concentrations of particulate PAHs from cooking in Hangzhou are higher than Guangzhou and Shenzhen due to the prevalence of frying meat in Hangzhou. The concentrations of particulate PAHs from household cooking with a decreased amount of cooking oil are lower than those from restaurant cooking. The concentrations of particulate PAHs from Hunan and Dongbei cooking are lower than those from Cantonese and Sichuan cooking. For the Cantonese style, famous for its lightly seasoned cooking, methods of frying with peanut oil and blended oil, sautéing, and stewing are frequently employed. Sichuan style, famous for its numbing and spicy flavor, also mainly employs frying with blended oil, sautéing and stewing. For the Hunan style, famous for its spicy and sour flavor, methods of steaming, simmering and stir-frying with canola oil and peanut oil are mainly used. The Dongbei style is similar to home cooking and is famous for pot-roast, and this style mainly employs peanut oil and methods of simmering, marinating, flavor-potting and roasting (Zhao et al., 2007). The emissions of PAHs from cooking are affected by the consumption of cooking oil, oil temperature, and types of cooking oils (Yao et al., 2015). The particulate BaP emitted from frying (1.64 μg/m^3) is 4 orders of magnitude greater than the population-average concentration of BaP (9.1×10^{-5} μg/m^3) in indoor air in U.S. residences, which poses serious health risks for cooks in poorly ventilated rooms (Logue et al., 2012).

For biomass combustion, the EF_{PAHs} in the seven studied cities or provinces (Fig. 1d) range from 0.82 mg/kg (burned biomass) to 85.76 mg/kg (burned biomass). The values of C_{PM} vary from 100.32 mg/kg PM to 10,090.68 mg/kg PM (Fig. 1b). There are many influencing factors on EF_{PAHs}, such as fuel properties (e.g., moisture and bulk density), the combustion facility and its conditions (e.g., temperature, oxygen and air supply) (Lu et al., 2009; Shen et al., 2011, 2012a). In general, the EF_{PAHs} values from straw combustion are higher than EF_{PAHs} from wood combustion. Burning activities measured in the field often yield higher emissions of pollutants than those measured in the laboratory due to the limitation of air circulation (Johnson et al., 2008, 2010; Roden et al., 2006, 2009; Shen et al., 2011). Therefore, the EF_{PAHs} values from straw combustion that were measured in the laboratory in Hangzhou (Lu et al., 2009), Shanghai (Zhang et al., 2011), and Hebei Province (Zhang et al., 2011) are lower. The C_{PM} values from biomass combustion are higher than those from Chinese cooking, except for frying. However, the EF_{PAHs} values from biomass combustion (0.82–85.76 mg/kg) are considerably higher than the EF_{PAHs} from frying (79.44 μg/kg). The national emission of particulate PAHs from straw combustion is up to 6,015 t/year.

The concentrations of PAHs in the ash of MWI and HWI range from 0.63 mg/kg to 12,601.2 mg/kg (Fig. 1c), and they are not determined by the daily capacity of the incinerators. The designs of the incinerators and the types of wastes have important effects on the production of PAHs. The concentrations of PAHs produced by fluidized-bed incinerators are lower than for grate-furnace incinerators due to higher combustion efficiency (Shen et al., 2010a; Van Caneghem et al., 2012). Notably high concentrations of PAHs from medium HWI around Beijing and HWI with daily capacities of 1.2 t in Wenchuan are attributed to low combustion efficiency. Moreover, the concentrations of PAHs in the ash from MWI (0.63–4.93 mg/kg, mean: 2.34 mg/kg) are lower than those in the ash from HWI (5.49–12,601.2 mg/kg, mean: 2,142.52 mg/kg) due to the high plastic content of HWI and high moisture content of MWI in China (Chen et al., 2013a; Dong et al., 2016). Moisture clearly reduces the reaction temperature and inhibits the pyrosynthesis of PAHs. Other impact factors include combustion temperature, excess air coefficient, residence time, additives, etc. (Chen et al., 2013a). The amounts of PAHs in the ash from MWI and HWI are 44 t/year and 45 t/year in China, respectively (Lynn et al., 2017). The PAHs from electronic shredder waste are 17 t/year (Ma et al., 2009).

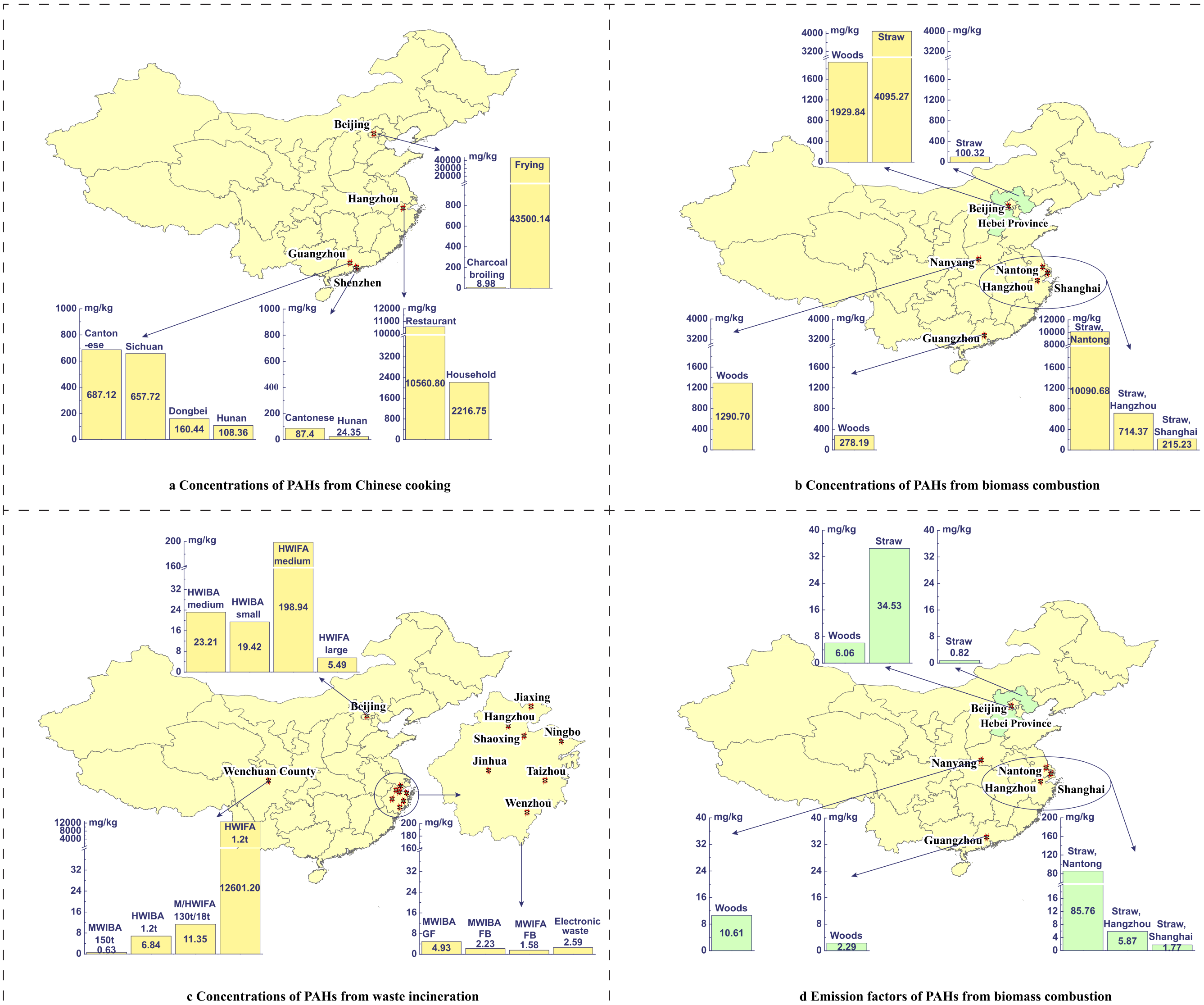

Fig. 1. Concentrations of particulate PAHs (per mass of particle matter) and their emission factors emitted from Chinese cooking, biomass combustion, and waste incineration on mainland China. HWI, hospital waste incinerators; MWI, municipal waste incinerators; BA, bottom ash; FA, fly ash; FB, fluidized-bed incinerators; and GF, grate-furnace incinerators.

3.2. PAHs emitted from domestic and industrial coal combustion

The EF_{PAHs} from domestic coal combustion in the nine studied provinces and Beijing (Fig. 2c) vary from 0.01 mg/kg burned coal (Guangzhou) to 662.95 mg/kg burned coal (Beijing) with an average value of 83.84 mg/kg. Honeycomb briquette, as a cleaner and more efficient source of heating and cooking, produces fewer particulate PAHs (0.01–111.37 mg/kg burned coal) than coal chunks (2.45–662.95 mg/kg burned coal). Although the honeycomb briquettes are incentivized by the Chinese government, the coal chunks are still widely used in the rural or poor areas of North China due to their low cost (Bi et al., 2008; Chen et al., 2004, 2005; Liu et al., 2009). Bituminous coals, as low-rank coals, have characteristics of high volatile content and high coal-tar yield, which results in higher emissions of PM and pyrosynthesized PAHs (Chen et al., 2005; Mitra et al., 1987). Moreover, the EF_{PAHs} values from anthracite coal combustion (0.01–13.55 mg/kg) are lower than EF_{PAHs} from biomass combustion (0.82–85.76 mg/kg), while the values from bituminous coal combustion (0.16–662.95 mg/kg) are higher than EF_{PAHs} from biomass combustion. In addition to the types of coal, variations of EF_{PAHs} are also dependent on stove/coal combination, combustion conditions and operation technologies (Liu et al., 2009; Shen et al., 2010b). The national emission of particulate PAHs from domestic coal combustion are up to 7,797 t/year. The C_{PM} from domestic coal combustion range from 0.44 mg/kg in Guangzhou to 50,636.12 mg/kg in Beijing (Fig. 2a).

The distributions of C_{PM} from coal combustion by various industries, such as coke plants, smelting plants, power plants, and industrial boilers, in five provinces/cities are presented in Fig. 2b. The C_{PM} values (280.80–3719.95 mg/kg) from coking coal combustion are higher than those from other industrial coal combustion. Moreover, the C_{PM} values from coke plants in Shanxi are higher than in other regions. The enrichment of PAHs within finer particles results in a higher concentration of PAHs within PM10 (Arditsoglou et al., 2004; Wang et al., 2012). The fine particles (PM10) pose high risks to the respiratory system of humans and possess characteristics of long-distance transport (Iordanidis et al., 2008; Kong et al., 2011; Wang et al., 2012). Although the EF_{PAHs}

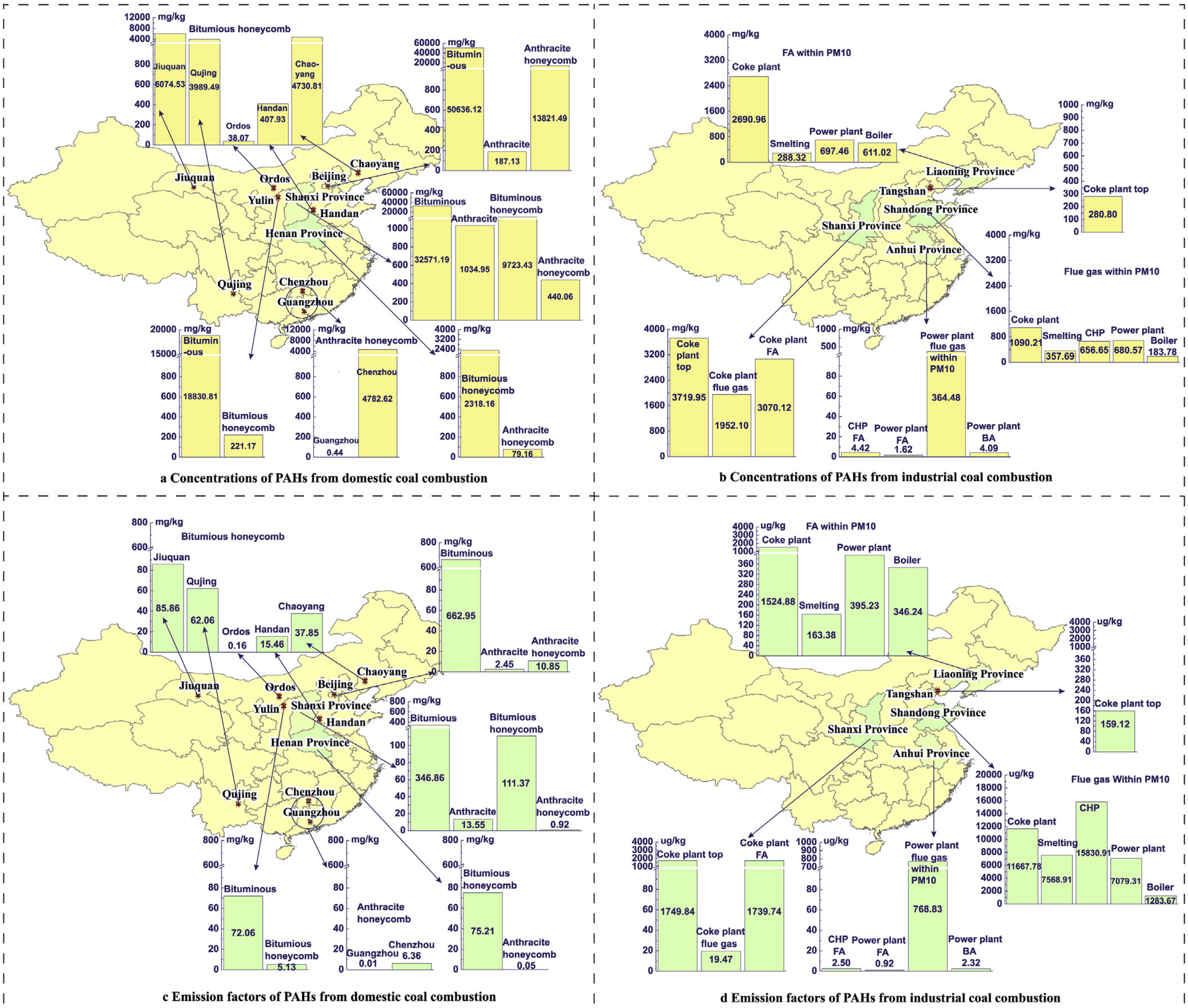

Fig. 2. Concentrations of particulate PAHs (per mass of particle matter) and their emission factors emitted from industrial and domestic coal combustion. BA, bottom ash; FA, fly ash; FB, fluidized-bed incinerators; and CHP, combined heat and power plants.

values in Shandong are considerably higher (1283.67–15830.91 μg/kg), the C_{PM} values in Shandong are lower than in Shanxi and Liaoning (Fig. 2b and d). This finding can be explained by the high emission of PM10 (45.65–157.57 mg/m^3) from industrial coal combustion in Shandong (Kong et al., 2011; 2013). The EF_{PAHs} values from industrial coal combustion range from 0.92 μg/kg to 15830.91 μg/kg with an average of 2959.00 μg/kg. Their variations are attributed to types of coals, plant design, operating conditions, sampling methods (Li et al., 2016). The national emission of particulate PAHs from industrial coal combustion reach 11,244 t/year. EF_{PAHs} from coke plants and power plants are higher than those from smelting plants and industrial boilers.

3.3. PAHs emitted from petroleum product-related sources

The distributions of EF_{PAHs} from engines in six cities on the Chinese mainland are presented in Fig. 3c. The EF_{PAHs} values that were measured in engine factories are lower than the values for the exhaust under actual operating conditions (Duan et al., 2016; Li et al., 2013). The EF_{PAHs} values are not determined by the speed and load of engines (He et al., 2010; Lu et al., 2012). However, the EF_{PAHs} values under uniform speed are lowest compared with acceleration, deceleration, and idling (Hu et al., 2015). For diesel engines, the EF_{PAHs} values range from 203.35 μg/kg fuel in Beijing (FAW-WDEW 4CK, China) to 1,160.41 μg/kg fuel in Nanjing (Iveco A40 under acceleration) with an average value of 554.31 μg/kg fuel. These values are higher than the EF_{PAHs} of gasoline engines [minimum: 160.51 μg/kg fuel (Chang'an SC6372 in Nanjing), maximum: 591.25 μg/kg fuel (Santana in Hangzhou), and mean: 332.18 μg/kg fuel]. Therefore, the differences in the values of EF_{PAHs} primarily result from the diversity of testing methods, driving modes and engine types. The national emissions of particulate PAHs from diesel and gasoline engines are 94 t and 37 t, respectively. For diesel engines, the C_{PM} values (Fig. 3a) vary from 73.66 mg/kg (Beijing) to 17,123.48 mg/kg (Luoyang). These values are lower than the C_{PM} from gasoline engines (1,368.64 mg/kg in Beijing to 54141.46 mg/kg

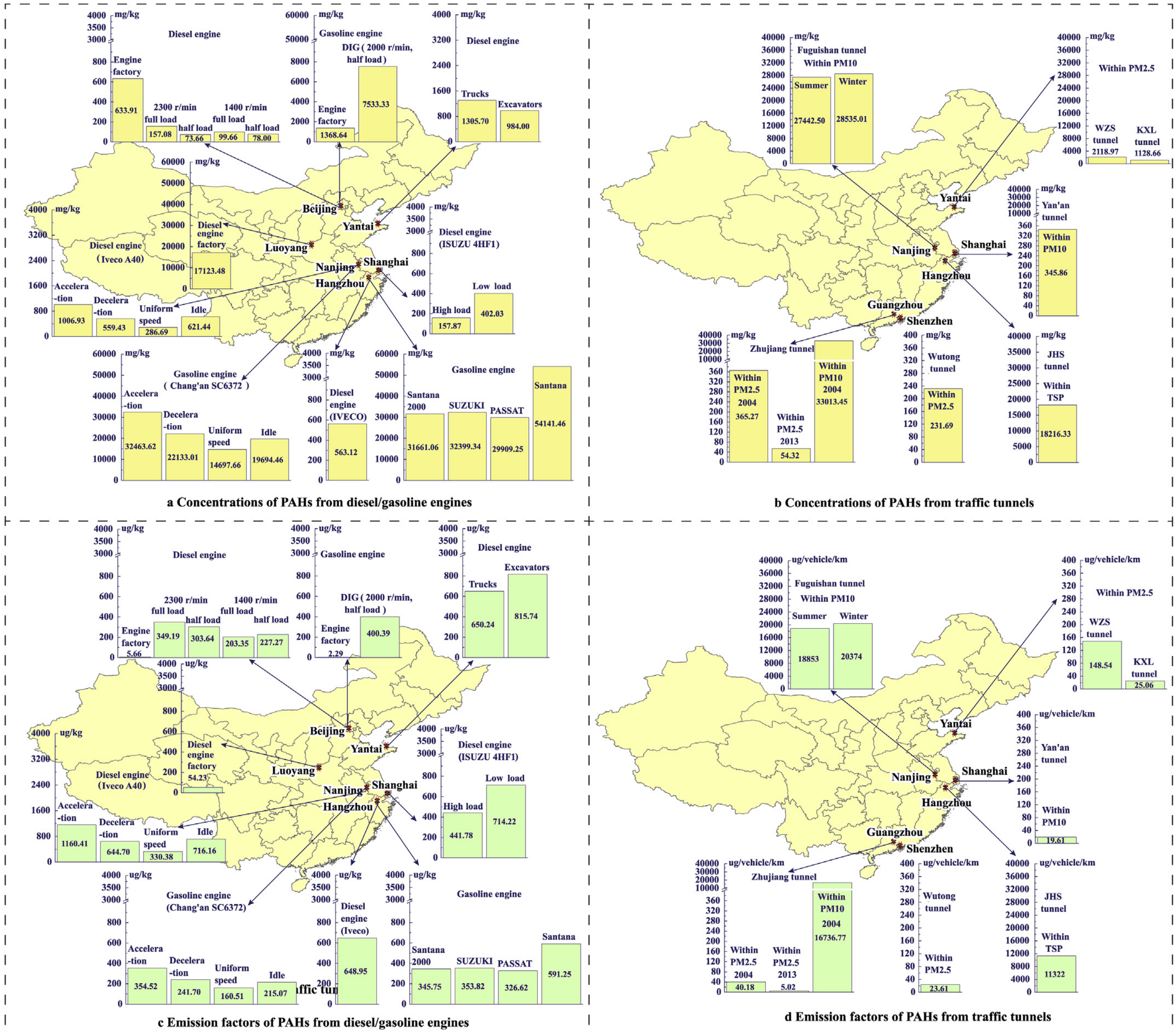

Fig. 3. Concentrations of particulate PAHs (per mass of particle matters) and their emission factors emitted from engines and traffic tunnels on mainland China. TSP, total suspended particulates.

in Hangzhou) because of the high emissions of PM from diesel engines. Therefore, two different regression equations were used to transform C_{air} to C_{PM} for diesel engines and gasoline engines (Table 1).

For traffic tunnels, the C_{PM} and EF_{PAHs} in six cities on the Chinese mainland are presented in Fig. 3b and d, respectively. Both C_{PM} and EF_{PAHs} within PM10 (C_{PM}: 345.86–33,013.45 mg/kg; EF_{PAHs}: 19.61–20,374 μg/vehicle/km) are considerably higher than the C_{PM} and EF_{PAHs} within PM2.5 (C_{PM}: 54.32–2,118.97 mg/kg; EF_{PAHs}: 5.02–148.54 μg/vehicle/km). For the Fuguishan tunnel in Nanjing, both C_{PM} and EF_{PAHs} in winter are slightly higher than those in summer owing to a greater number of diesel vehicles in winter (Chen et al., 2013b). Generally, EF_{PAHs} from diesel engines are higher than EF_{PAHs} from gasoline engines, while C_{PM} from diesel engines are lower due to the high PM emissions from diesel engines (see the above paragraph). Thanks to implementation of the vehicle emission control policies since 2004, both C_{PM} and EF_{PAHs} in the Zhujiang tunnel in 2013 are considerably lower than the values measured in the same tunnel in 2004 and in other studied tunnels (Dai et al., 2015).

Based on the above results, the national emissions of particulate PAHs from various sources are presented in Fig. 4. The top three sources of particulate PAHs are industrial coal combustion > domestic coal combustion > straw combustion. On the scale of the entire Chinese mainland, particulate PAHs from these three sources account for 99% of the total particulate PAHs. The results from Anderson-Darling Test using Oracle Crystal Ball software indicated that the emissions of 16 PAHs from the top three sources are log-normally distributed. Through Monte Carlo simulation, the differences between the 75th and 25th percentiles varied from 35% (straw combustion) to 61% (domestic coal combustion) of the average values. There were large uncertainties due to the many influencing factors discussed in this study. Slight differences were detected between the directly calculated values and the Monte

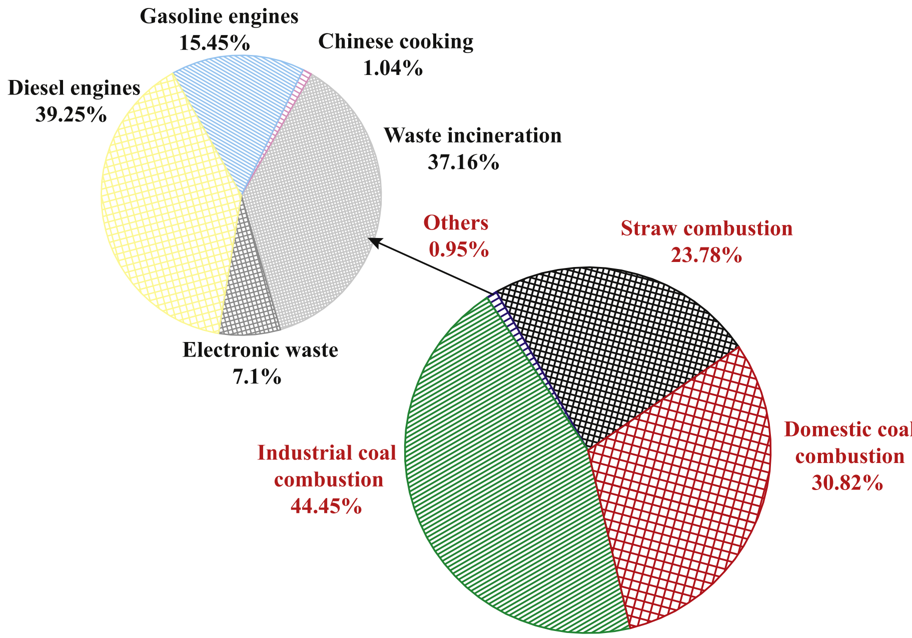

Fig. 4. Source contribution rates to national emissions of particulate polycyclic aromatic hydrocarbons.

Carlo mean values. In 2003, emissions of PAHs from straw combustion in China were higher than those from domestic coal combustion and industrial coal combustion due to the difference in the energy structure between 2003 and 2015 (Xu et al., 2006).

3.4. Linkage of PAH compositions to various sources by multivariate analysis

Using PCA, the variances of particulate PAHs in samples collected from eight sources (Chinese cooking, biomass combustion, waste incineration, domestic coal combustion, coking coal combustion, coal combustion for power generation, and both diesel and gasoline engines) explained by the first to fourth axis (factor) are 28.4%, 17.0%, 11.2%, and 9.5%, respectively (sum: 66.1%). For Any, Phe, Ant, Fla, Chr, B(b/k)F, BaP, DaA, I1P, and BgP, more variances are explained by the first axis than the other axes. For Nap, Ane, and Pyr, more variances are mainly explained by the second axis. In Fig. 5a, the coordinate system is defined by the first and second axes, which contain more information on the compositions of PAHs in the collected samples than the other coordinate systems (Fig. 5b–f). In the scatter plot results from PCA (Fig. 5a–f), the samples that are grouped together have similar compositions of PAHs, whereas the separated samples have different compositions of PAHs. In Fig. 5a, the samples collected from the same sources marked with the same symbols are grouped together, whereas the samples collected from different sources marked with different symbols are separated from each other. Therefore, various sources with different PAH compositions can be effectively identified via multivariate analysis. The particulate PAHs collected from traffic tunnels interfere with those of engines, pavement tar and other factors; thus, they do not have distinct characteristics.

After normalization to the BaP, the average concentrations of the 16 PAHs in the particulate phase from various anthropogenic sources are presented in Table 2. Among eight sources, the three highest concentration PAHs are different from each other. For Chinese cooking, the three highest concentrations of PAHs are Pyr > Fla > BgP. For biomass combustion, the three highest concentration PAHs are Phe > Fla > Pyr. These findings are consistent with those from the same source summarized based on Jenkins et al. (1996), Lee et al. (2005) and Schauer et al. (2001). For waste incineration, they are Phe > Nap > BaA. For domestic/coking/power coal combustion, the three highest concentrations of PAHs are B(b/k)F > Chr > Phe, B(b/k)F > Chr > BaA and Phe > Nap > Fla, respectively. These findings are different from those obtained by Li et al. (2003) (domestic coal combustion: Phe > Fla > B(b/k)F, coking coal combustion: Nap > B(b/k)F > Chr, and coal combustion for power generation: Chr > Fla > Phe). For diesel engines, the three highest PAH concentrations are Pyr > Phe > Fla, which are similar to those (Fla > Phe > Pyr) summarized by Li et al. (2003). However, the three PAHs with the highest concentrations from gasoline engines in China are BaA > BgP > I1P, which are different from those (Pyr > Fla > BgP) summarized by Li et al. (2003). Therefore, it is critical to determine the characteristics of particulate PAHs emitted from various anthropogenic sources in China.

3.5. Implications/suggestions for policy making

Both the results from Xu et al. (2006) and this study indicated that the combustion of biomass and coal are main sources of particulate PAHs in China. Controlling emissions of PM from coal and straw combustion is an urgent problem. The order of the values of EF_{PAHs} is bituminous coal chunk combustion > bituminous honeycomb briquette combustion > biomass combustion > anthracite coal chunk combustion > anthracite honeycomb briquette combustion. The reserves of anthracite coal are considerably lower than those of bituminous coals and the price of anthracite coal is higher. First, implementation of financial subsidies for the use of anthracite coals/honeycomb briquettes is necessary to reduce emissions of PM and their associated PAHs. At the same time, the harmful effects on residential health of PM and their associated PAHs emitted from bituminous coal combustion should be made known across rural regions of the Chinese mainland.

The EF_{PAHs} values from industrial coal combustion (0.92–15,830.91 μg/kg) are considerably lower than those from domestic coal combustion (0.01–662.95 mg/kg). However, industrial coal consumption (3.8×10^9 t) is considerably higher than

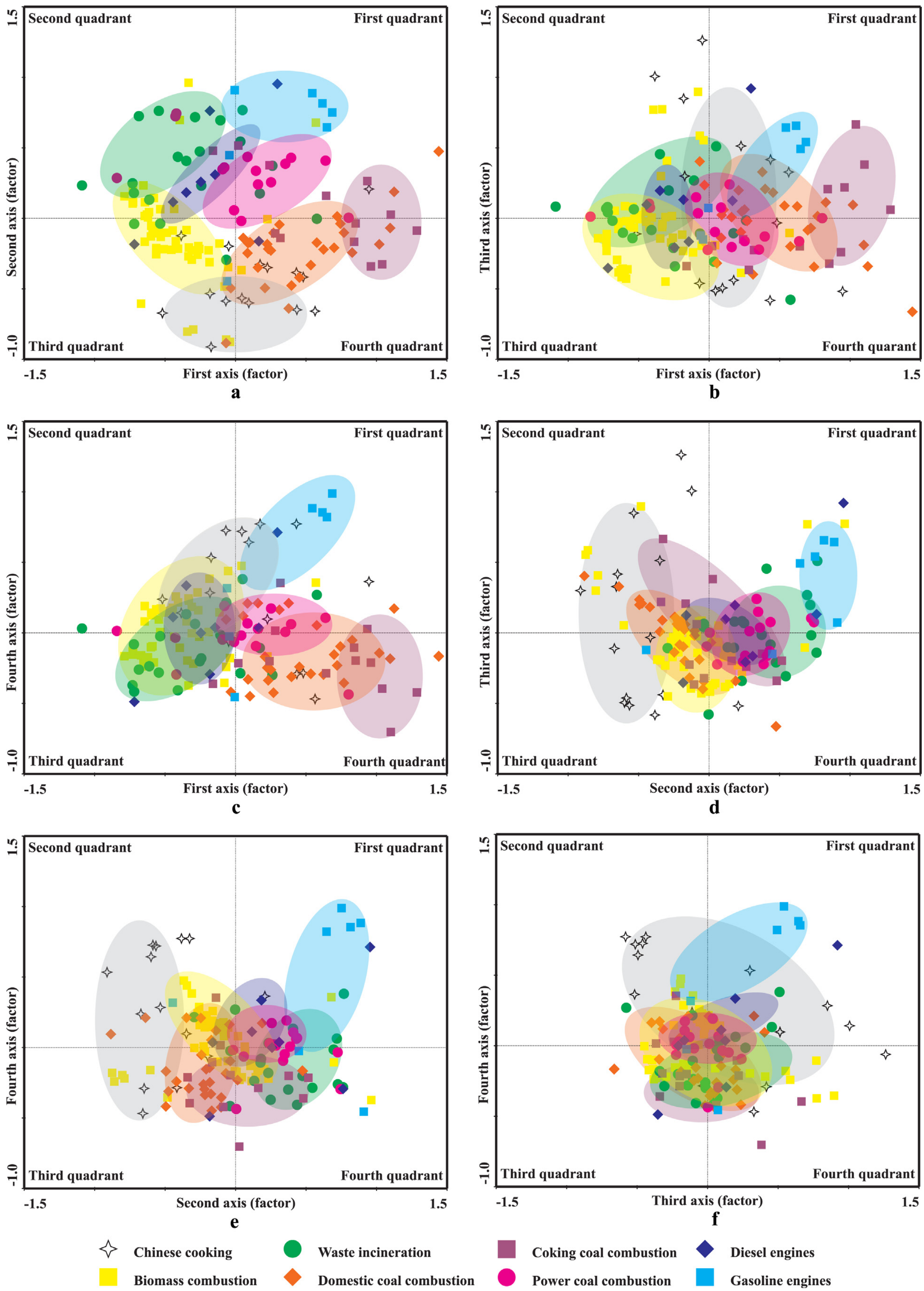

Fig. 5. Scatter plots of samples from various sources in different coordinate systems resulting from principal component analysis.

Table 2
Average concentrations (after normalization) of the 16 priority PAHs of the USEPA in particulate phase from eight sources.

	Chinese cooking	Biomass combustion	Waste incineration	Domestic coal	Coking coal	Power coal	Diesel engines	Gasoline engines
naphthalene	0.30 ± 0.14 n = 8	1.62 ± 0.46 n = 45	68.62 ± 36.96 H n = 20	0.20 ± 0.08 n = 5	0.75 ± 0.34 n = 13	7.77 ± 3.49 H n = 15	19.89 ± 11.90 n = 8	4.07 ± 1.05 n = 7
acenaphthene	0.90 ± 0.30 n = 8	0.87 ± 0.27 n = 45	7.14 ± 3.96 n = 20	0.02 ± 0.01 n = 15	0.16 ± 0.05 n = 13	1.08 ± 0.24 n = 15	4.19 ± 2.00 n = 8	0.97 ± 0.42 n = 7
acenaphthylene	0.00 n = 1	2.88 ± 0.52 n = 45	9.82 ± 6.77 n = 20	0.04 ± 0.02 n = 15	0.19 ± 0.09 n = 13	1.31 ± 0.34 n = 15	7.91 ± 4.22 n = 7	1.54 ± 1.16 n = 3
fluorine	0.05 ± 0.01 n = 8	2.65 ± 0.59 n = 51	13.21 ± 7.00 n = 20	0.15 ± 0.04 n = 29	0.37 ± 0.17 n = 13	3.44 ± 1.84 n = 15	7.55 ± 2.19 n = 8	2.12 ± 0.78 n = 7
phenanthrene	2.10 ± 0.90 n = 14	9.31 ± 1.07 H n = 51	142.44 ± 100.40 H n = 20	2.47 ± 0.53 H n = 29	1.03 ± 0.28 n = 13	7.84 ± 4.54 H n = 15	271.32 ± 261.25 H n = 8	3.99 ± 2.08 n = 7
anthracene	0.32 ± 0.10 n = 10	1.50 ± 0.13 n = 51	10.07 ± 6.07 n = 20	0.71 ± 0.11 n = 29	0.67 ± 0.26 n = 13	1.55 ± 0.51 n = 15	14.03 ± 12.29 n = 8	1.09 ± 0.72 n = 7
fluoranthene	22.57 ± 15.51 H n = 15	6.86 ± 0.66 H n = 51	27.67 ± 14.19 n = 20	1.99 ± 0.22 n = 29	1.10 ± 0.18 n = 13	4.86 ± 1.54 H n = 15	137.11 ± 123.49 H n = 8	3.35 ± 0.74 n = 7
pyrene	41.00 ± 30.47 H n = 15	6.22 ± 0.54 H n = 51	13.50 ± 5.48 n = 20	1.70 ± 0.20 n = 29	0.98 ± 0.18 n = 13	4.69 ± 2.15 n = 15	1112.6 ± 1098.23 H n = 8	2.06 ± 0.60 n = 7
benzo(a)anthracene	3.82 ± 1.55 n = 11	2.58 ± 0.85 n = 51	35.16 ± 21.29 H n = 20	1.72 ± 0.17 n = 29	1.13 ± 0.12 H n = 13	1.89 ± 0.26 n = 15	30.01 ± 16.45 n = 8	11.51 ± 3.87 H n = 7
chrysene	5.58 ± 1.81 n = 15	1.87 ± 0.17 n = 51	12.82 ± 7.87 n = 20	3.72 ± 0.56 H n = 29	1.24 ± 0.13 H n = 13	1.72 ± 0.22 n = 15	30.57 ± 24.31 n = 8	1.87 ± 0.64 n = 7
benzo[b/k]fluoranthene	3.59 ± 1.12 n = 15	2.20 ± 0.11 n = 45	7.47 ± 3.27 n = 20	4.27 ± 0.87 H n = 29	3.48 ± 0.55 H n = 13	3.39 ± 0.47 n = 15	20.09 ± 15.04 n = 7	4.60 ± 1.09 n = 3
benzo(a)pyrene	1.00 ± 0.00 n = 15	1.00 ± 0.00 n = 51	1.00 ± 0.00 n = 20	1.00 ± 0.00 n = 29	1.00 ± 0.00 n = 13	1.00 ± 0.00 n = 15	1.00 ± 0.00 n = 8	1.00 ± 0.00 n = 7
dibenz(a,h)anthracene	0.08 ± 0.08 n = 2	0.35 ± 0.06 n = 45	1.77 ± 1.07 n = 20	0.77 ± 0.19 n = 29	0.39 ± 0.08 n = 13	0.75 ± 0.11 n = 15	17.36 ± 16.53 n = 6	1.34 ± 0.87 n = 3
indeno(l,2,3-cd)pyrene	1.35 ± 0.47 n = 8	0.61 ± 0.04 n = 45	8.60 ± 5.76 n = 20	1.21 ± 0.27 n = 29	0.84 ± 0.13 n = 13	1.05 ± 0.13 n = 15	4.61 ± 3.22 n = 8	4.63 ± 1.26 H n = 7
benzo(g,h,i)perylene	6.71 ± 3.06 H n = 8	0.94 ± 0.36 n = 45	1.97 ± 0.60 n = 20	1.81 ± 0.43 n = 29	0.78 ± 0.09 n = 13	1.13 ± 0.21 n = 15	54.03 ± 49.48 n = 8	5.36 ± 1.28 H n = 7

The three highest concentration PAHs were marked with superscript H.

domestic coal consumption (9.3×10^7 t) and more than half of the industrial coal consumption is attributed to power plants. Therefore, measures should be taken by the Chinese government to support research and development of other power technologies such as solar power instead of coal-fired power.

On the scale of the entire Chinese mainland, diesel/gasoline engines and Chinese cooking are not major sources of particulate PAHs. However, PAHs from engines and Chinese cooking pose serious health risks for the workers or cooks, which necessitate labor-protection measures. There were large uncertainties for PAHs from the same types of sources due to the many influencing factors discussed above. Therefore, environmental forensics for a specific event should be performed on the basis of the distinctive composition of PAHs emitted from specific sources using robust source analysis methods, such as multivariate analysis.

4. Conclusions

In this study, linear regression equations on the relationship between the mass of particulate PAHs per mass of particle matter, the mass of particulate PAHs per volume of air, and emission factors for particulate PAHs have been built to compare emissions of PAHs from different sampling sites and different sources. For all of mainland China, up to 99.05% of total particulate PAHs are from industrial coal combustion (44.45%), domestic coal combustion (30.82%), and straw combustion (23.78%). Although Chinese cooking is not a main source of particulate PAHs (0.1%), the particulate BaP emitted from frying (1.64 $\mu g/m^3$) poses serious health risks for cooks. Eight different anthropogenic sources (Chinese cooking, biomass combustion, waste incineration, domestic coal combustion, coking coal combustion, coal combustion for power generation, diesel engines and gasoline engines) with distinct PAH compositions can be effectively identified via PCA. Moreover, the characteristics of particulate PAHs emitted from human activities in China are not totally consistent with those in other countries. The reasons for the diversity of PAH emissions from these specific pollution sources have also been discussed.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2018.03.091.

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