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Spatiotemporal distribution of atmospheric polycyclic aromatic hydrocarbon emissions during 2013–2017 in mainland China

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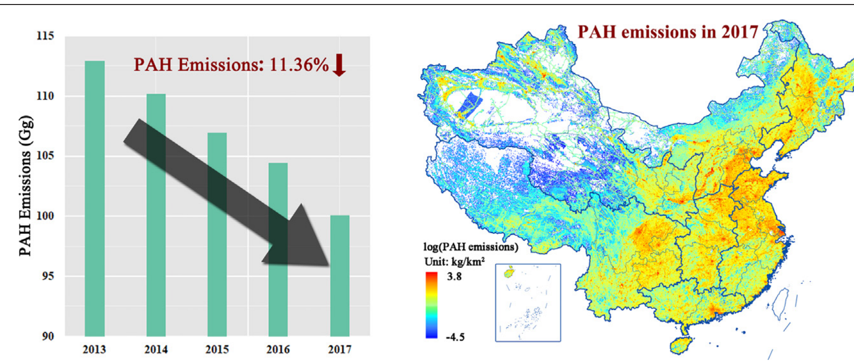
HIGHLIGHTS

- A gridded PAH emission inventory was established for mainland China during 2013–2017.
- The results show that the PAH emissions decreased by 11.36% from 2013 to 2017.
- The PAH emissions in the industrial and residential/commercial sectors declined the fastest.
- PAH emissions have been decreasing, but the total emissions remain significant.

GRAPHICAL ABSTRACT



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ABSTRACT

Following implementation of the most stringent clean air policy in China, the emissions of NOx, SO_2, and fine particles have greatly reduced since 2013. However, the emissions of polycyclic aromatic hydrocarbons (PAHs), which are highly toxic pollutants, and their spatiotemporal changes remain unclear. In this study, a 0.05° × 0.05° gridded PAH emission inventory was developed for mainland China during 2013–2017. The results show that the total PAH emissions have decreased from 112.92 Gg in 2013 to 100.09 Gg in 2017, with the fastest declines in the industrial (17.32%) and residential/commercial (10.58%) sectors. However, the decline in the PAH emissions is smaller than that of the NO_X and SO_2 emissions. The average emission density of PAHs in mainland China in 2017 was 10.43 kg/km^2. North and East China have the largest PAH emissions. The residential/commercial, industrial, and transportation sectors are the major emission sources, accounting for 48.59%, 29.26%, and 17.21%, respectively. Carcinogenic PAH emissions accounted for 7.49% in mainland China, higher than those of developed countries (5.73%) and the global average (6.19%). Differences in the energy structures lead to significant differences in the spatial distribution of PAH emissions in various sectors. From 2013 to 2017, the emissions declined in most Chinese regions. The emission density in East China decreased the most, reaching 3.39 kg/km^2, followed by North China (2.91 kg/km^2). The magnitude of the decline in the PAH emissions and reasons for the decline significantly differ in different regions. Particular attention must be paid to the limited decline (5.22%) in

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Northwest China over the study period. Although China's emission density has been declining, it is still significantly higher than the global average. Therefore, China must further reduce the PAH emissions through technological innovation and reductions of energy consumption and, thus, reduce the regional lung cancer risk.

1. Introduction

Airborne polycyclic aromatic hydrocarbons (PAHs) are typically generated by the incomplete combustion or pyrolysis of fossil and biomass fuels and various industrial processes (Ravindra et al., 2008). Exposure to atmospheric PAHs can cause lung cancer and other diseases (Perera, 1997; Shen et al., 2014; Shrivastava et al., 2017). The United States Environmental Protection Agency (U.S. EPA) has listed sixteen PAH compounds as priority pollutants. Several PAHs, such as benzo(*a*)pyrene (BaP), are highly toxic.

As the largest developing country in the world, China has experienced rapid urbanization and industrialization, which led to a five-fold increase in the energy consumption over the past 30 years (NBS, 2018a). China's atmosphere, soil and water suffered serious pollution and ecological risk (Zhang et al., 2019; Gao et al., 2020; Li et al., 2020). China's PAH emissions are the highest in the world (Shen et al., 2013), especially northern cities, have severe atmospheric PAH pollution. In 2009–2010, the BaP equivalent concentration (BaPeq) in Taiyuan (city in northern China) was ~25 times higher than the level recommended by the WHO (1 ng/m^3; Xia et al., 2013). Xu et al. (2018) reported that the cumulative cases of nonoccupational lung cancer excess associated with BaP from beehive coke ovens were approximately 3500 ($\pm$1500) between 1982 and 2015. This demonstrates that PAH pollution in China seriously threatens both the ecological environment and human health. Based on the implementation of many initiatives, the atmospheric PAH levels have decreased in recent years (Zhuo et al., 2017) but remain severe. For example, in 2012–2013, the national daily BaP_{eq} standard (2.5 ng/m^3) was exceeded at ~71% of the air monitoring sites and ~ 94.8% of the population was exposed to the national BaP_{eq} standard in China (Hong et al., 2016).

The implementation of certain policies has effectively reduced PAH pollution. For example, the implementation speed of the Coal Law in China significantly reduced excess lung cancer cases associated with BaP (Xu et al., 2018). Additionally, replacement of traditional stoves with improved modern stoves was implemented to reduce PAH emissions. Lin et al. (2016) found that the improved stoves can reduce 48%–91% PAHs emissions. In fact, many new policies implemented as part of the "Air Pollution Prevention and Control Action Plan" (introduced in 2013) were closely related to the reduction of the PAH emissions. For example, tighter emission standards along with stricter emission control measures on high emitters (e.g., iron and steel industry (GB 28662–2012)) were applied to the major industry sectors during 2013–2017. The large coal-fired industrial boilers were required to implement the new emission standard (GB 13271–2014), and more than 200,000 small coal boilers were shut down during this period (Zhang et al., 2019). For the transportation sector, the "China 5" emission standard was applied in 2017, and oil quality was also upgraded to be consistent with the emission standards. In addition, replacing the coal/biomass with electricity and natural gas in rural households was an important measure for the residential sector. By the end of 2017, energy consumption in 6 million households in China switched from coal to electricity and natural gas (Zhang et al., 2019).

Establishing a gridded PAH emission inventory is extremely important because it can be used to clarify the spatial distribution of pollutant emissions (Streets et al., 2003; Bond et al., 2004). It also serves as the key input of chemical transport models. Several organizations and researchers have established gridded PAH emission inventories such as the PKU-PAH, USPEA, EMEP, and REAS-POP (Vestreng and Klein, 2002; Inomata et al., 2012; Shen et al., 2013; USEPA, 2014; Muntean et al., 2016; Li et al., 2018). However, some of these PAH emission inventories have a relatively coarse resolution and contain a small number of species. For example, the resolution of REAS-POP is 0.5° × 0.5° and the inventory includes only nine PAH species (Inomata et al., 2012). In China, studies have been carried out to establish a gridded PAH emission inventory. Shen et al. estimated the China's PAH emissions in 2007 to be 106 Gg, accounting for 20.3% of the global emissions (Shen et al., 2013). Several other researchers focused on the PAH emissions at the city or regional level (Jiang et al., 2013; Li et al., 2018; Huang et al., 2019). However, there is a lack of studies of gridded PAH emissions in China in recent years.

Owing to the frequent and severe air pollution, the Chinese government has issued many policies and regulations. Based on the implementation of the "Air Pollution Prevention and Control Action Plan" in China, the $PM_{2.5}$ concentration has decreased by 30%–40% from 2013 to 2017 (Li et al., 2019b; Zhang et al., 2019). However, the PAH emission have been little investigated despite the relatively high toxicity of PAHs. Considering the strict policies and rapid increase of the energy consumption, it is extremely important to clarify the spatiotemporal variation of the PAH emissions during 2013–2017 in China. Therefore, the main objectives of this study were (1) to establish a gridded emission inventory during 2013–2017 for mainland China; (2) to clarify the main PAH emission sources; and (3) to analyze the spatiotemporal variation of the PAH emissions.

2. Methods

The study area was mainland China. Hongkong, Macao, and Taiwan were excluded from this study. The following 16 PAHs were included in this work: naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorine (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz(*a*)anthracene (BaA), chrysene (CHR), benzo(*b*)fluoranthene (BbF), benzo(*k*)fluoranthene (BkF), benzo(*a*)pyrene (BaP), dibenz(*a*,*h*)anthracene (DahA), indeno(l,2,3-cd)pyrene (IcdP), and benzo(*g*,*h*,*i*)perylene (BghiP). Among them, BaA, CHR, BbF, BkF, BaP, IcdP, DahA, and BghiP are carcinogenic PAHs.

2.1. Calculation of provincial PAH emissions

In this study, 49 emission sources were considered, mainly including indoor and open biomass burning, industrial coal consumption, coke production, primary Al production, iron–steel industry, and transportation. They were divided into five sectors: energy production, industry, commercial/residential sources, transportation, and agriculture (Table 1). Natural sources were not considered in this study because of their extremely low proportion (Shen et al., 2013). The detailed four-level source categorization is listed in Table S1. The emissions of 16 PAHs were calculated using Eq. (1):

$$E_{PAH} = \sum_{i,j,k} A_{i,k} \times X_{i,k} \times EF_{i,j,k} \tag{1}$$

where *i*, *k*, and *l* represent the fuel, process, and technology, respectively, and *A* is the activity level. Most of activity data were obtained from statistical yearbooks (NBS, 2014-2018a, b, c, d). The parameter *X* is the fraction of the activity rate of a given technology, which was calculated using the technology split method (Bond et al., 2007, Shen et al., 2013; see details in Section S1). In total, 24 sources were considered for the technology split (Table S1). The parameter *EF* is the

Table 1
PAH emission sources considered in our study.

Energy Production	Industry	Residential & commercial	Transportation	Agriculture
coal	coal	non-organized waste burning	Gasoline	outdoor straw burning
gas/diesel	coke production	bituminous coal	diesel oil	gas/diesel
residue oil	iron sintering	anthracite coal		
solid biomass	crude steel production	liquid petroleum gas		
municipal waste	Hot rolling	dry natural gas		
industrial waste	petroleum refinery	natural gas liquid		
dry natural gas	primary Al production	biogas		
liquefied natural gas	other industrial oil	kerosene		
	dry natural gas	indoor straw burning		
	natural gas liquid	indoor firewood combustion		
		indoor dung cake burning		

The PAH emission sources are listed in detail in Table S1.

emission factor of each PAH species j, which was derived from previous studies (Shen et al., 2011, 2013; Li et al., 2018).

The amount of combusted crop residue is not available in statistical yearbooks after 2007 and thus was estimated using Eq. (2):

$$E_i = P_i \times N_i \times R_i \times F_i \times D_i, \tag{2}$$

where i is the specific crop (corn, wheat, or rice); P is the crop yield; N is the grain-to-straw ratio; R and F are the combustion ratio and combustion efficiency, respectively; and D is the dry matter fraction of each straw type. Crop yield data were derived from statistical yearbooks (NBS, 2014-2018d). The combustion ratios and dry matter fractions were obtained from Zhou et al. (2017).

The livestock manure was calculated using Eq. (3):

$$A = S_i \times Y_i \times C_i \times R_i, \tag{3}$$

where A is the annual mass of burned livestock manure; S represents the number of each livestock type (i), which was derived from statistical yearbooks; C represents the dry matter fraction of livestock manure, which was set to 18%, and Y is the annual fecal output of a single livestock (Mg head^{-1}). The manure output of cattle, horse, and other large animals was set to 10, 7.3, and 8 Mg, respectively, based on previous studies (Tian et al., 2011; Zhou et al., 2017). The parameter R is the proportion of total livestock manure, which is directly combusted, and was set to 20% (Liu and Shen, 2007; Zhou et al., 2017). Livestock manure is only used in pastoral and semi-pastoral areas for biomass burning in China; therefore, Tibet, Inner Mongolia, and the Gansu, Xinjiang, and Qinghai provinces were considered (Tian et al., 2011).

2.2. Spatial allocation methods for different PAH sources

The provincial PAH emission sources were spatially allocated to a $0.05° \times 0.05°$ grid using several proxies or methods (Table 2). The energy production sector was allocated based on the location and plant capacity of the power station, which were obtained from the World Resources Institute (WRI, http://datasets.wri.org/dataset). The industry sector was allocated to the grid using a new subnational fuel data disaggregation method based on point-of-interest data (DPOI). Details about the DPOI allocation method were previously discussed (Li et al., 2019a). In brief, the DPOI reduces the spatial bias caused by the downscaling of emissions. It can be used to determine the real industrial energy consumption by combining the industrial enterprise POI with official energy consumption data. This approach represents a feasible solution for the use of big data and official statistics to facilitate the development of emission inventories in countries, such as China, which lack accurate information regarding locations of industrial point sources. The residential/commercial sector was allocated based on the rural population/population, which was derived from the LandScan dataset (https://landscan.ornl.gov/download; Bright et al., 2017). The transportation sector was allocated based on the annual road density (OpenStreet data, http://www.geofabrik.de). The gasoline/diesel consumption in the agriculture sector was allocated according to the rural population. Open biomass burning in the agriculture sector was allocated based on the gridded fire point numbers in cropland. The gridded fire point numbers were calculated from the MODIS active fire product MCD14ML (http://modis-fire.umd.edu/). The MCD14ML can be used to detect smaller and cooler fires and is less affected by the cloud cover and aerosol conditions (Giglio et al., 2003).

2.3. Uncertainty

The uncertainty of the PAH emissions is mainly caused by the activity level and emission factors. We analyzed the uncertainty of the PAH emissions using a Monte Carlo method based on various inventory studies (Bond et al., 2007; Wu et al., 2016; Zhou et al., 2017). Based on previous studies, we assumed that the uncertainties of the activity data and emission factors are uniformly and lognormally distributed, respectively (Shen et al., 2013). The standard deviations for the activity levels were derived from Shen et al. (2013) and the geometric mean and geometric standard deviation of the emission factors of different sources were obtained from Zhang and Tao (2009), Zhang (2010), and Shen et al. (2013). The PAH emission calculations were replicated 10,000 times, with a random selection of all inputs.

3. Results and discussion

3.1. Total emissions and source contributions in mainland China

The total PAH emissions in China decreased from 2013 to 2017 (Fig. 1a). The annual PAH emissions from 2013 to 2017 were 112.92 Gg (103.61–152.88 Gg, 95% confidence interval based on the Monte Carlo simulation), 110.14 Gg (99.68–146.27 Gg), 106.92 Gg (96.62–140.91

Table 2
Spatial allocation methods and the relevant data sources for PAH emissions from province to grid.

Sector/fuel	The proxy from province to grid	Sources
Energy production	the location and capacity of power stations	World Resources Institute (WRI, http://datasets.wri.org/dataset)
Industry	disaggregation method based on point-of-interest data (DPOI)	(Li et al., 2019a)
Residential and commercial	rural population/population	LandScan dataset (https://landscan.ornl.gov)
Transportation	road network	OpenStreet data (http://www.geofabrik.de)
Gasoline/diesel consumption in agriculture sector	rural population	LandScan dataset
Open biomass burning in agriculture sector	the gridded fire point numbers	MODIS active fire product MCD14ML (http://modis-fire.umd.edu/)

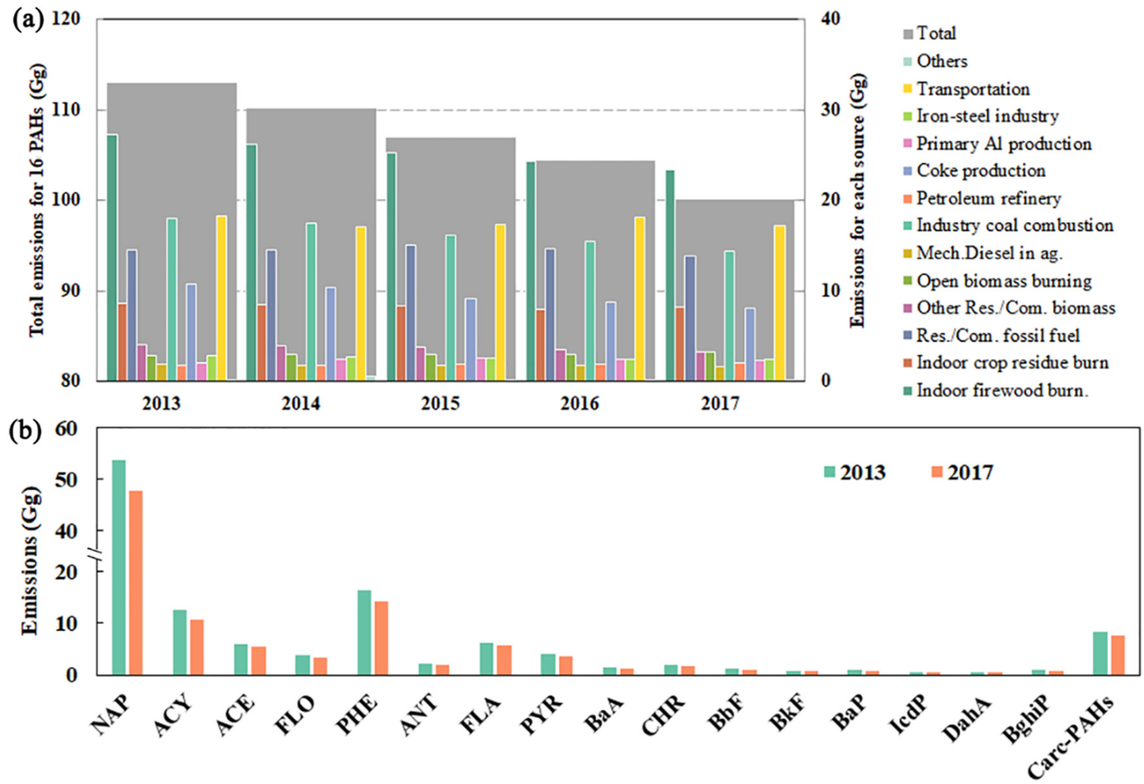

Fig. 1. (a) Anthropogenic emissions of 16 polycyclic aromatic hydrocarbons (PAHs) by sector and year in mainland China. The gray column represents the total annual PAH emissions; (b) Changes in the PAH emission composition profiles between 2013 and 2017. Among the investigated PAHs, BaA, CHR, BbF, BkF, BaP, IcdP, DahA, and BghiP are carcinogenic.

Gg), 104.43 Gg (94.38–137.13 Gg), and 100.09 Gg (90.89–132.26 Gg), respectively. The proportions of the emission sources were similar during these 5 years. Taking 2017 as an example, the residential/commercial sector is the largest emission source of PAHs in China (48.63 Gg), accounting for 48.59% of the total emissions. Indoor firewood combustion contributed the most to the emissions, accounting for 23.35% of the total PAH emissions (up to 23.37 Gg), followed by fossil fuel combustion (13.83%). Compared with 62% in 2007 (Shen et al., 2013), the proportion of the residential/commercial sector significantly decreased, which is closely related to recent technological improvements and the energy transition in rural areas (Tao et al., 2018). The PAH emissions of improved cooking stoves are much lower than those of traditional cooking stoves (Shen et al., 2012), and many households have switched from coal to electricity and natural gas (Zhang et al., 2019), which also reduces the PAH emissions.

Although continuous technology improvements have been made in the industrial sector in recent years, the proportion of industrial PAH emissions has increased from 19.5% in 2007 to 29.26% in 2017 owing to the significant increase in industrial energy consumption. The industrial energy consumption increased from 1.90 billion tce to 2.95 billion tce from 2007 to 2017. Although vehicle ownership in China has increased by 94.06% in the past 10 years (from 159.8 to 3101 million) (NBS, 2018b), the proportion of traffic PAH emissions has only increased from 12.6% in 2007 to 17.21% in 2017 (from 13.36 to 17.23 Gg), which is mainly owing to strengthened vehicle emission standards, retirement of old "yellow-label" vehicles, and improvements of the fuel quality. For example, the "China 5" emission standard was applied to light gasoline and diesel vehicles in 2017 (NDRC, 2016). The agricultural and energy sectors accounted for 0.11% and 4.83% of the PAH emissions, respectively.

The PAH emissions decreased by 11.36% from 2013 to 2017, representing a smaller reduction compared with SO_2 (−59%) and NO_X (−21%; Zheng et al., 2018). The emission changes vary significantly in different emission sectors. In general, the emissions of the industrial and residential/commercial sectors decreased the most, reaching 17.32% and 10.58%, followed by the transportation sector (5.76%). The other two sectors contributed little to the initial PAH emissions, accounting for only 4.6% of the total emissions on average in these years. Industrial coal consumption and coke production contributed the most to the decline of industrial PAH emissions, decreasing by 19.94% and 25.16%, respectively, from 2013 to 2017, which is mainly related to upgrades of industrial boilers and strengthened industrial emission standards. The decrease of emissions in the residential/commercial sector was mainly because of the promotion of clean fuels. A more stringent emission standard was applied to coal-fired power plants. In particular, in 2015, China pledged to further reduce the emissions from coal power by 60% by 2020 using the ultralow emission technique, which led to a significant reduction in SO_2 and NO_X in the power generation sector. In contrast, the proportion of PAH emissions in the power generation sector was very small (0.11%), which explains the larger decline of the SO_2 and NO_X emissions compared with that of the PAH emissions. All of these measures are part of the "Air Pollution Prevention and Control Action Plan" (SCC, 2013), which thus has played a decisive role in reducing the PAH emissions.

The proportions of the PAH emission composition profiles were similar during 2013–2017. In 2017, two- and three-ring PAHs accounted for the majority of the PAH emissions (83.29%). The NAP accounted for 47.83% of the total PAH emissions, followed by PHE (14.18%) and ACY (10.72%). The emission reduction of these three species accounted for 77.14% of the total decline. The carcinogenic PAH emission density decreased by 9.50% over these five years, from 0.86 kg/km^2 in 2013 to 0.78 kg/km^2 in 2017. Note that the emissions of the eight carcinogenic PAHs in mainland China in 2017 were 7.49 Gg, accounting for only 7.49% of the total emissions. However, they were higher than those in developed countries (5.73%) and the global average (6.19%; Shen et al., 2013). This larger contribution might lead to greater ecological and lung cancer risks in China (Wang et al., 2012).

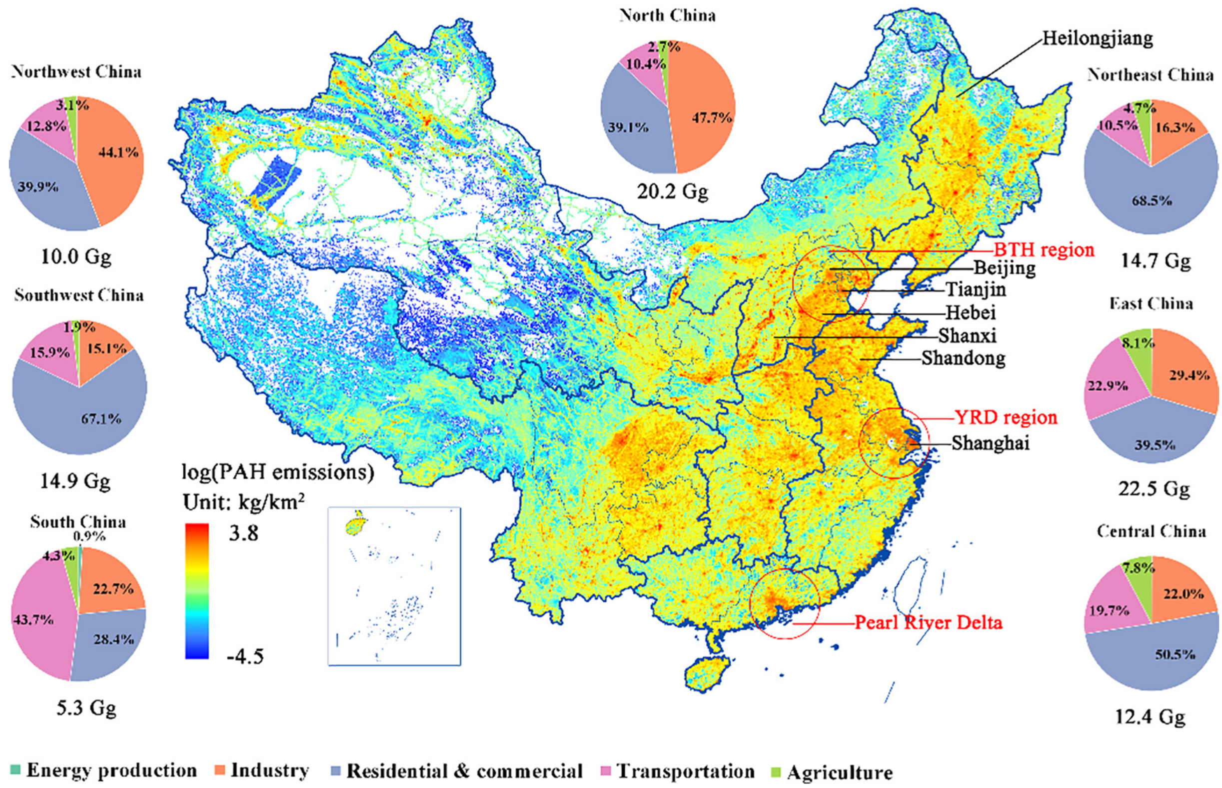

Fig. 2. Geographic distribution of polycyclic aromatic hydrocarbon (PAH) emissions in mainland China in 2017. Relative contributions of the five sectors are shown as pie charts for each region.

3.2. Spatial distribution of PAH emissions in 2017

The average emission density of PAHs in China in 2017 was 10.43 kg/km^2. North and East China are the regions with the largest PAH emissions, with emissions accounting for 42.7% of the total emissions in mainland China (Fig. 2). East China had the highest emission density of 30.05 kg/km^2, followed by Central China (23.31 kg/km^2) and Northeast China (16.09 kg/km^2). Shandong, Shanxi, and Hebei are the three provinces with the highest PAH emissions (8.05, 7.22, and 6.69 Gg, respectively (Fig. 3). These three provinces are in North China and utilize a large amount of coal/biomass for heating. In addition, these three provinces also have relatively developed industries, which explains the high PAH emissions. In contrast, Beijing's PAH emissions (0.51 Gg) are the lowest among all provinces, which is because of very strict control measures. Differences in energy structures lead to the significant differences in the spatial distribution of PAH emissions in various sectors (Fig. 4). In 2017, the residential/commercial, industrial, and transportation sectors contributed 95.1% to the total PAH emissions in mainland China.

The residential/commercial sector was the dominant emission source in the 21 provinces. The emission density of the residential/commercial sector in Northeast, North, and Central China exceeds 10 kg/km^2. These areas are often densely populated and large amounts of biomass and coal are burned for heating. For example, in Heilongjiang, the PAH emissions of the residential/commercial sector reached 5.09 Gg including high proportions of indoor coal (33.29%) and biomass

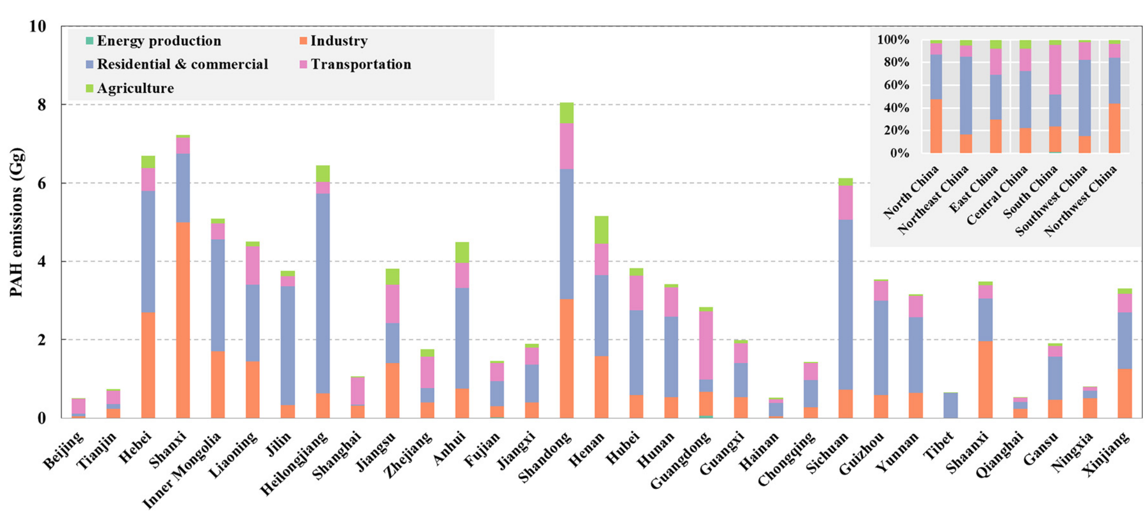

Fig. 3. Polycyclic aromatic hydrocarbon (PAH) emissions of the five sectors for each province in 2017.

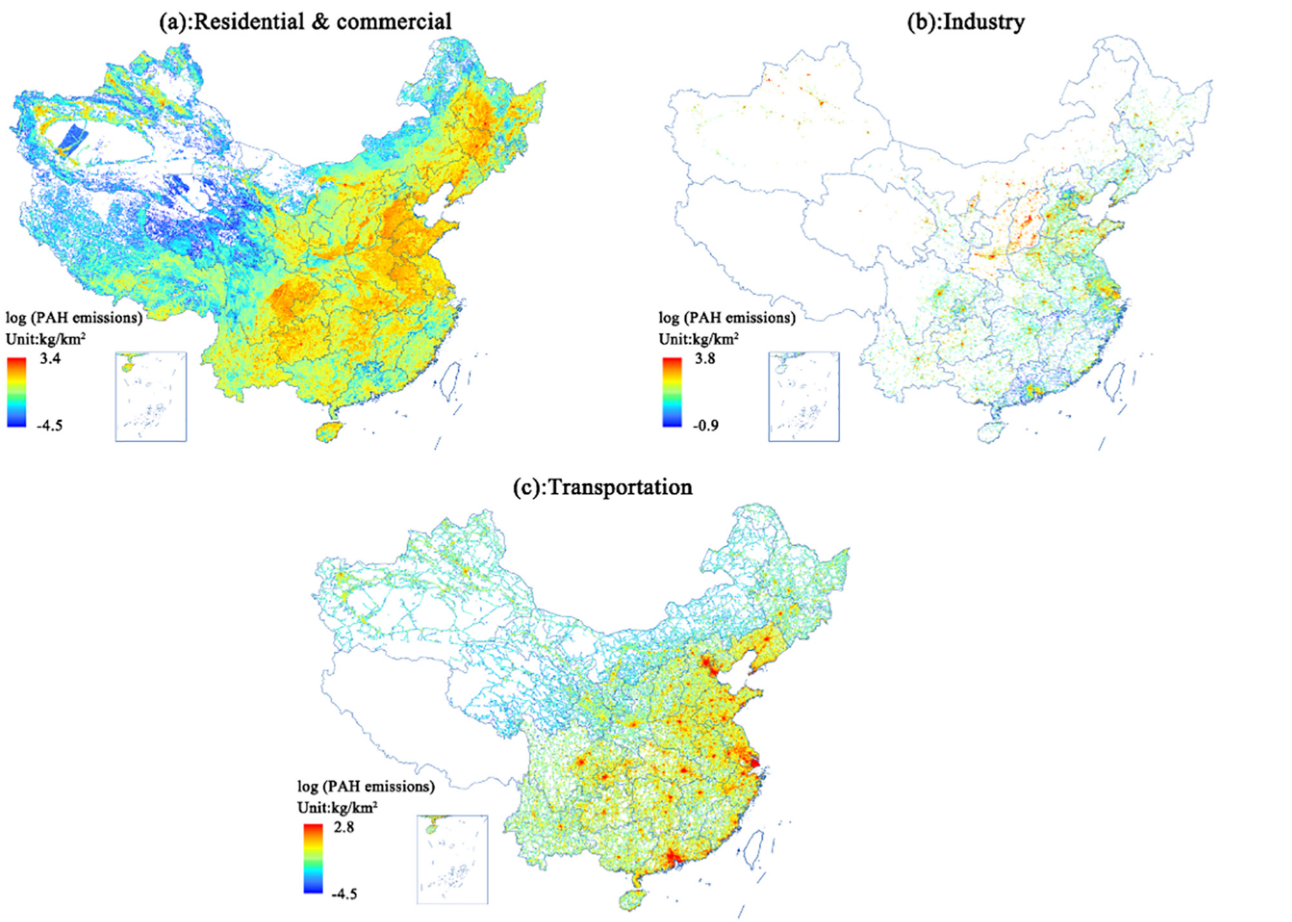

Fig. 4. Geographic distribution of the polycyclic aromatic hydrocarbon (PAH) emissions in the (a) residential/commercial, (b) industrial, and (c) transportation sectors in 2017.

(39.25%) combustion. Furthermore, the PAH emissions in the residential/commercial sector accounted for 78.9% of the total emissions in Heilongjiang; they were higher than the total emissions of both the power generation and agricultural sectors in mainland China (4.94 Gg). South China has the lowest emissions in the residential/commercial sector (1.52 Gg). The emission density (3.72 kg/km^2) was much lower than that of Northeast China (11.02 kg/km^2).

East China has the highest PAH emission density in the industrial sector (8.83 kg/km^2). Fig. 4 shows that the areas with high emission densities are mainly concentrated in Shanxi, Tianjin, Hebei, Yangtze River Delta (YRD), and some other developed areas. The Shanxi Province has the largest industrial PAH emissions (4.99 Gg), with an emission density of 31.23 kg/km^2, which is more than ten times the national industrial emission density. The proportion of industrial sources in the Shanxi Province is the highest among all provinces, reaching 69.05%, which is due to emissions originating from coking and industrial coal combustion. These two processes account for 95.6% of the total industrial PAH emissions in Shanxi. This indicates that the energy structure of Shanxi, which is the top coal producer in China, is still dominated by coal. In addition, the industrial emission density in Beijing–Tianjin–Hebei (BTH) and the YRD (two important urban agglomerations in China) was 13.19 and 10.78 kg/km^2, respectively, that is, much higher than the average level (3.05 kg/km^2) in China. Shanghai, which is the most developed region in China, had the highest industrial emission density (55.49 kg/km^2). Note that Beijing has implemented the most stringent atmospheric control measures in China (SCC, 2013; Zhang et al., 2016), resulting in very low industrial PAH emissions (41.1 t).

In several developed regions, such as Beijing and Shanghai, PAH emissions from industrial and residential/commercial sources sharply decreased because of the development of the economy and strict energy management and control strategies. Transportation has become the dominate source of PAH emissions. The PAH emissions from the transportation sector are concentrated in developed areas, such as BTH, the YRD, and Pearl River Delta. Owing to the different economic development levels of the provinces, the PAH emissions from traffic sources significantly differ. The emission density of Shanghai (117.39 kg/km^2) was the highest in China, followed by Tianjin (28.18 kg/km^2) and Beijing (22.44 kg/km^2). The emission densities in these provinces were much higher than the national average level (1.80 kg/km^2).

3.3. Changes in the PAH emissions in different regions from 2013 to 2017

We further calculated the changes in the PAH emissions in mainland China from 2013 to 2017 (Fig. 5). A decreasing trend can be observed for most regions of China. The PAH emissions decreased by 11.37% in these five years, representing a relatively large improvement. The emission density in East China decreased the most, reaching 3.39 kg/km^2, followed by North China (2.91 kg/km^2). All provinces with emission density reductions of more than 5 kg/km^2, that is, Shanghai (45.27 kg/km^2), Tianjin (12.22 kg/km^2), Hebei (8.92 kg/km^2), Shanxi (8.40 kg/km^2), and Beijing (6.48 kg/km^2), are located in these two regions (Table 3).

The BTH region frequently suffers from severe air pollution (Dang and Liao, 2019). Therefore, the policies are stricter than in other regions (Xiang et al., 2020). The "Air Pollution Prevention and Control Action Plan" mandates a decrease in the $PM_{2.5}$ concentration in the BTH by 25% within 5 years, which is higher than the national average (10%). Hence, several pollution control policies (e.g., phase out of small and polluting factories, phase out of outdated industrial capacity, and upgrades of industrial boilers) were established (SCC, 2013). For example,

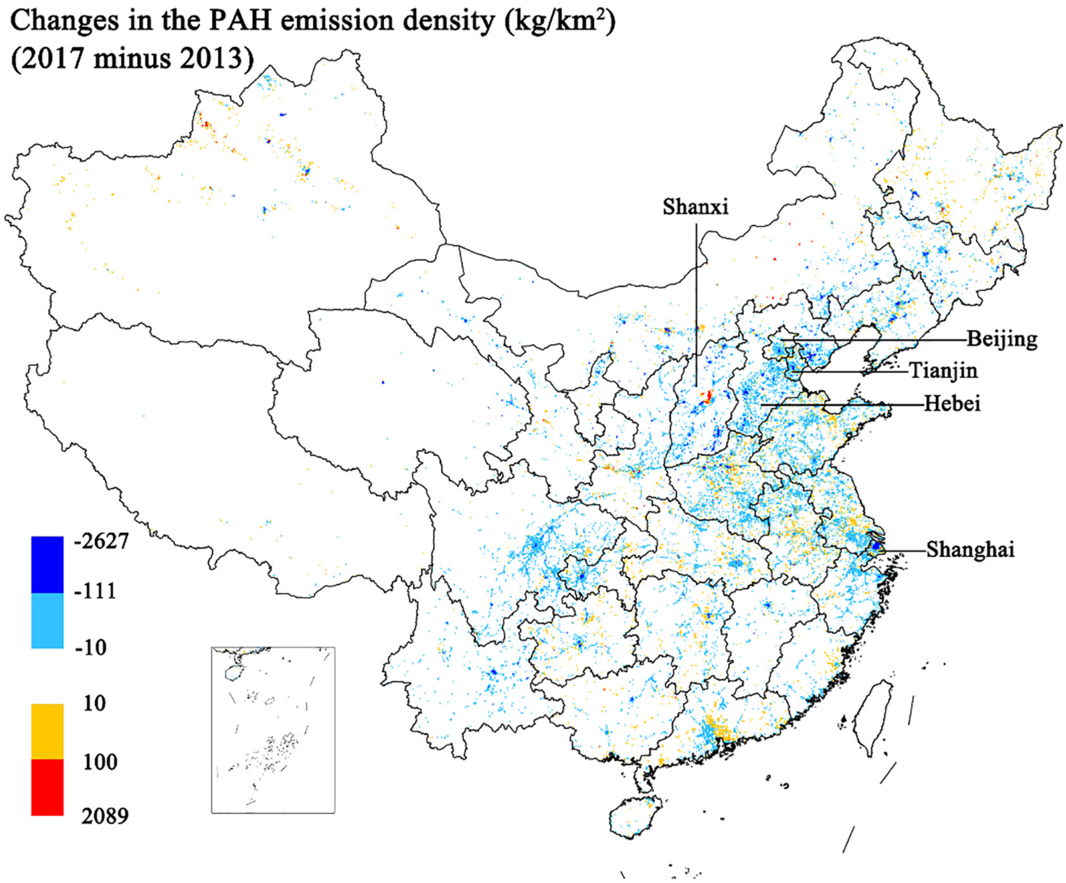

Fig. 5. Changes in the polycyclic aromatic hydrocarbon (PAH) emission density from 2013 to 2017.

Table 3
Changes in the polycyclic aromatic hydrocarbon (PAH) emission density in mainland China by sector from 2013 to 2017 (kg/km^2).

Change (2017 minus 2013)	Sum-PAHs	Each sector				
		Energy production	Industry	Residential & commercial	Transportation	Agriculture
Beijing	−6.48	0.00	−1.69	−0.89	−3.60	−0.29
Tianjin	−12.22	0.11	−10.06	−1.14	−1.09	−0.04
Hebei	−8.92	0.00	−5.85	−2.55	−0.50	−0.02
Shanxi	−8.40	0.00	−6.82	−1.18	−0.41	0.00
Inner Mongolia	−1.18	0.00	−0.35	−0.71	−0.11	0.00
Liaoning	−2.95	−0.01	−1.67	−1.13	0.04	−0.19
Jilin	−3.05	0.00	−0.89	−1.99	−0.19	0.02
Heilongjiang	−0.28	0.00	−0.39	0.24	−0.21	0.08
Shanghai	−45.27	−0.34	−12.91	−18.75	−11.40	−1.87
Jiangsu	−4.67	0.00	−2.39	−1.53	−1.02	0.28
Zhejiang	−3.77	0.00	−0.97	−0.81	−1.67	−0.33
Anhui	−2.38	0.00	−0.79	−2.40	0.26	0.56
Fujian	−1.23	0.04	−0.01	−1.15	−0.25	0.15
Jiangxi	−1.62	0.00	−0.84	−0.71	−0.05	−0.01
Shandong	−4.93	0.01	−1.11	−3.55	−0.56	0.28
Henan	−4.74	0.00	−3.32	−2.05	0.09	0.53
Hubei	−1.10	0.00	−0.79	0.20	−0.26	−0.24
Hunan	−0.91	0.00	−0.56	−0.49	0.13	0.02
Guangdong	−0.41	0.07	0.65	−1.15	0.07	−0.05
Guangxi	−0.46	0.00	0.07	−0.84	0.25	0.06
Hainan	−1.84	0.00	0.23	−1.54	−0.17	−0.36
Chongqing	−3.39	0.00	−1.19	−2.48	0.30	−0.02
Sichuan	−2.84	0.00	−0.82	−1.76	−0.11	−0.15
Guizhou	−0.08	0.00	−1.47	0.58	0.66	0.14
Yunnan	−1.71	0.00	−1.07	−0.44	−0.20	0.00
Tibet	−0.06	0.00	0.00	−0.06	0.00	0.00
Shaanxi	−1.53	0.00	0.98	−1.80	−0.67	−0.04
Gansu	−0.59	0.00	−0.21	−0.24	−0.14	0.00
Qinghai	−0.22	0.00	−0.21	−0.05	0.04	0.00
Ningxia	−1.84	0.00	−1.35	−0.33	−0.09	−0.07
Xinjiang	0.15	0.00	0.01	0.14	0.00	0.00

coal-fired boilers with ≤35 steam tons were eliminated in urban areas (MEP, 2013). Approximately 62,000 small and polluting enterprises in BTH and surrounding regions were phased out or upgraded (Zhang et al., 2019). These policies have played important roles in the reduction of the PAH emissions. The decreases in the PAH emissions from the industrial sectors in Hebei and Tianjin accounted for 65.56% and 82.30% of the total decrease, respectively. The heavy industry of Beijing, the capital of China, accounted for a low proportion. The transportation sector contributed the most to the PAH emission decline in Beijing, accounting for 55.64%. From 2013 to 2017, vehicle ownership in Beijing increased by only 3.7%, far below the national level of 24.04%. In addition, yellow-licensed vehicles were eliminated and the fuel quality was improved. All these measures have led to a sharp decline of the emissions from the transportation sector.

The decline of the PAH emissions in Shanghai (one of the most developed cities in China) was mainly dominated by the residential/commercial sector, which contributed 41.42%. The domestic coal consumption sharply dropped from 117.77×10^4 t to 9.9×10^4 t (NBS, 2014-2018b). The decline in Shanxi was mainly due to the reduced contribution of coking and industrial coal in the industrial sector, which contributed 76.36% to the total decline. However, in several other areas, the PAH emission decline was very limited or the emissions increased. The PAH emissions in Northwest China were only reduced by 5.22%, which was related to the 8% increase in the PAH emissions in Xinjiang. Because of the relatively clean environment and low population density, stringent policies might not have been implemented in Xinjiang. The PAH emission increase was mainly due to the increase in primary Al production and domestic coal combustion (NBS, 2014-2018b). The emission factors of PAHs from these two sources are relatively high (Shen et al., 2013). The increases in the PAH emissions from the two sources offset the decreases related other sources. Therefore, more attention must be paid to the primary Al production and domestic coal combustion in Xinjiang in the future.

In general, the total PAH emissions in mainland China have decreased since 2013, but the decline in the PAH emissions and reasons for the decline significantly differ in different regions. The emission reductions in Beijing and Zhejiang were mainly caused by the transportation sector. The decreases in the emissions in 14 provinces (Tianjin, Hebei, Shanxi, Liaoning, Heilongjiang, Jiangsu, Jiangxi, Henan, Hubei, Hunan, Guizhou, Yunnan, Qinghai, and Ningxia) were mainly because of the industrial sector, while that of other provinces was dominated by the residential/commercial sector. The residential/commercial sector is currently the largest source of PAH emissions in China. Reducing the emissions from this sector (e.g., indoor biomass burning, domestic coal combustion) is extremely important. Promoting the proportion of clean fuel (e.g., switching coal to electricity) and improving the combustion efficiency (e.g., using the improved cooking stoves) can effectively reduce emissions from the residential/commercial sector.

3.4. Uncertainty and comparison with other studies

We determined the emissions uncertainty of each sector during 2013–2017 based on the Monte Carlo simulation. In general, the uncertainty of the emissions inventory was reduced by 5.28% (Fig. S1). Taking 2017 as an example, the energy production sector (−51.60% ~ 153.56%) and transportation sector (−50.29% ~ 61.51%) had the largest emissions uncertainty, followed by the industry (−17.35% ~ 79.53%), agriculture (−32.76% ~ 29.37%) and residential/commercial (−11.69% ~ 32.91%) sector. At 4.94 Gg, the emissions from the energy production and transportation sectors were relatively small; hence, the large uncertainties of these two sectors did not have a significant impact on the total PAH emissions.

We compared the PKU-PAH, REAS-POP, and other emission inventories with that established in our study (Table 4). Our data are consistent with those of Shen et al. (2013) and Zhang and Tao (2009), slightly lower than those of PKU-PAH, and slightly higher than those of Mu (2010). The REAS-POP results are significantly lower than those obtained in this and other studies, which might be owing to the following reasons: (1) The emission factor used for REAS-POP has a relatively high uncertainty, especially regarding domestic biomass burning; (2) REAS-POP excludes several sources (e.g., iron–steel industry, petroleum refineries, primary Al production). Based on our calculations, primary Al production alone was responsible for the emission of 1.92 Gg PAHs in 2014.

Table 4
Comparison of different studies.

Year	Source	Species (number of PAHs)	Emissions (Gg)	References
2007	Shen et. al	16	106.4	(Shen et al., 2013)
2004	Zhang et. al	16	114	(Zhang and Tao, 2009)
2014	PKU-PAH	16	122.7	http://inventory.pku.edu.cn
2004	Zhang et. al	9	26.3	(Zhang and Tao, 2009)
2005	REAS-POP	9	9.6	https://www.nies.go.jp/REAS
2012	Mu	16	81.18	(Mu, 2010)
2014	This study	16	110.14	This study
2014	This study	9	17.86	This study

The emissions data of PKU-PAH were updated to include data from 2014, so we further compared the 2014 emissions of every PAH species recorded by our study and PKU-PAH (Table S2). The normalized mean error (NME) is 11.14%. The PAH emissions of each province based on the two inventories are relatively consistent (Table S3; NME = 21.39%), verifying the PAH emission inventory established in this study.

4. Conclusions

Because the Chinese government has attached great importance to air pollution, especially to the implementation of policies such as the "Air Pollution Prevention and Control Action Plan" since 2013, the emissions of air pollutants, such as $PM_{2.5}$, have declined. However, the emissions of PAHs, and their spatiotemporal changes remain unclear. Therefore, we established a gridded PAH emission inventory with a high resolution of $0.05° \times 0.05°$ for 2013–2017. The results show that the total PAH emissions have decreased from 112.92 Gg in 2013 to 100.09 Gg in 2017. The PAH emissions in the industrial and residential/commercial sectors have reduced the fastest (to 17.32% and 10.58%) owing to strict policies.

The proportions of the PAH emission sources during these 5 years were similar. In 2017, the residential/commercial, industrial, transportation, energy production, and agriculture sectors accounted for 48.59%, 29.26%, 17.21%, 0.11%, and 4.83% of the total emissions, respectively. The NAP accounted for 47.83% of the total PAH emissions, followed by PHE (14.18%) and ACY (10.72%). The proportion of carcinogenic PAHs (7.49%) in mainland China is higher than that of developed countries (5.73%) and the global average (6.19%). The average emission density of PAHs in mainland China in 2017 was 10.43 kg/km^2, with the largest emissions in North China and East China.

From 2013 to 2017, the PAH emissions in mainland China decreased by 11.37%. The largest decline in the emission density occurred in East China (3.39 kg/km^2) and North China (2.91 kg/km^2). Provinces with an emission density reduction of more than 5 kg/km^2 include Shanghai (45.27 kg/km^2), Tianjin (12.22 kg/km^2), Hebei (8.92 kg/km^2), Shanxi (8.40 kg/km^2), and Beijing (6.48 kg/km^2). The decline in the emission density in these provinces was mainly due to the decrease in the energy consumption and technological innovation in the industrial and residential/commercial sectors. Note that the decline in the PAH emissions in Northwest China was very limited, which must be further studied. In addition, we compared the emission inventory established in this study and other inventories, and found our data to be consistent with those of PKU-PAH, Shen et al. (2013) and Zhang and Tao (2009).

In general, PAH emissions in mainland China have been decreasing, but the total emissions remain significant. The decline in the PAH emissions was smaller than that of the NO_X and SO_2 emissions from 2013 to 2017. Through technological innovation and reduction in energy consumption, it is possible to effectively reduce atmospheric PAH pollution, which in turn can help reduce regional lung cancer risk and associated illnesses.

CRediT authorship contribution statement

Teng Wang: Conceptualization, Investigation, Methodology, Writing - Original Draft.
Baojie Li: Conceptualization, Supervision, Resources, Writing - Editing.
Hong Liao: Conceptualization, Supervision, Writing – Editing.
Yan Li: Methodology, Writing - Revised Draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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