



Seasonal variation of polycyclic aromatic hydrocarbons (PAHs) emissions in China

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The seasonal variation of the PAHs emission in China was examined with a regression model.

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ABSTRACT

A regression model based on the provincial energy consumption data was developed to calculate the monthly proportions of residential energy consumption compared to the total year volume. This model was also validated by comparing with some survey and statistical data. With this model, a PAHs emission inventory with seasonal variation was developed. The seasonal variations of different sources in different regions of China and the spatial distribution of the major sources in different seasons were also achieved. The PAHs emissions were larger in the winter than in the summer, with a difference of about 1.3-folds between the months with the largest and the smallest emissions. Residential solid fuel combustion dominated the pattern of seasonal variation with the winter-time emissions as much as 1.6 times as that in the summer, while the emissions from wild fires and open fire straw burning was mainly concentrated during the spring and summer.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are notorious for their adverse effects on human health (Gaspari et al., 2003; Perera et al., 2002). As the main carcinogenic constituent of ambient aerosols (Straif et al., 2006), PAHs have long been an environmental concern. As the world's largest emitter of PAHs (Zhang and Tao, submitted for publication-b), China suffers severe PAHs contamination. High PAHs concentrations were detected in both the ambient and indoor atmosphere and national standards are often exceeded (Li et al., 2005; Wu et al., 2005).

Significant seasonal variation existed for the ambient atmospheric concentrations of PAHs in China. Zhou et al. (2005) monitored the seasonal variation of atmospheric PAHs concentrations associated with PM₁₀ in Beijing, and found much higher concentrations during the autumn and winter seasons than those in other seasons. Similar results were found in Shenyang, Dalian, Qingdao, Nanjing, Guangzhou and Hong Kong (Tang et al., 2005; Wan et al., 2006; Guo et al., 2003a,b; Wang et al., 2007; Tan et al., 2006). When compared with the summer, factors such as lower temperature, less wet deposition and slower photolysis and radical degradation also contributed to the higher concentrations measured during the winter (Guo et al., 2003b; Zhou et al., 2005; Tan et al., 2006), and

larger emissions during the winter were important causes especially for cities located in northern China (Guo et al., 2003b; Tang et al., 2005; Wan et al., 2006).

Some emission sources of PAHs in China certainly have large seasonal variations, including residential biofuel and coal combustion and open burning of agricultural wastes and wild fires. Due to the need for space heating, the energy consumption in the residential sector is generally higher in the winter than during other seasons. For instance, the total residential energy consumption in Beijing during the colder half of the year was about 5.4 million tce (ton of coal equivalent) in 2006, while only 3.5 million tce was consumed during the other half of the year (Beijing Statistical Bureau). A survey of 100 rural families in the Jilin province also showed that the straw consumption was about 400 and 200 kg/month per family during the winter and summer, respectively (Qin et al., 2007). The seasonal variation of energy consumption in the residential sector has been previously considered in emission inventories for pollutants that are mainly generated from combustion sources. In an emission inventory built for gaseous and primary aerosols in Asia for the year 2000, Streets et al. (2003) assumed there were about 5-folds of variance between the hottest and the coldest months in all of Asia based on a study that was conducted in India. Similarly, in an emission inventory of global carbonaceous aerosol, Lioussé et al. (1996) assumed there was 2.8-folds of variance in the northern hemisphere. Even though, the open burning of agricultural wastes, mainly the straw for crops, happened only during the harvest time of these crops (Cao et al., 2006). The Along

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Track Scanning Radiometer (ATSR) hot spot for the open burning of straw also showed a temporal pattern with concentrated burning in May, June and July throughout China, especially in the plains of northern China in June (Fu et al., 2007). Since wild fires are episodic in nature, the seasonal variation of emission from wild fires is undoubtedly large. For instance, the largest area burned per month in China in 2002 was about 54,000 ha in September, while the smallest was only 180 ha in December (Giglio et al., 2003).

Although a PAHs emission inventory of China with multi-spatial resolutions has been built (Xu et al., 2006; Zhang et al., 2007; Zhang and Tao, submitted for publication-a), and the annual variation of emissions has been addressed from the time period of 1950–2004, the seasonal variation of PAHs emissions has not yet been systematically evaluated. To better understand the magnitude of the seasonal variation of PAHs emissions and evaluate its contribution to the seasonal variation of the ambient concentrations, a PAHs emission inventory including seasonal variation for the year 2003 has been built in this study. With this inventory, the environmental influence of PAHs emitted by China for both local and remote regions in different seasons especially during the winter-time can be addressed. Moreover, this inventory can provide basic information for PAHs control strategies, and emission data for fate and transport modeling with seasonal variation.

2. Methodology

Based on the previously acquired emission inventory, the emission sources of PAHs in China include firewood, straw, domestic coal combustion, coke and aluminum production, traffic and industrial petroleum combustion, industrial coal combustion, open burning of agricultural wastes and wild fires (Zhang and Tao, submitted for publication-a). In this study, the seasonal variation of emissions from biofuel and coal combustion from the residential sector, consumption of coal for residential space heating in centralized heat-generating facilities, and open burning of agricultural wastes and wild fires were considered. Other sources, especially industrial sources were considered to be constant throughout the entire year.

Due to the lack of monthly and seasonal statistical data for energy consumption in China, regression models were developed to distribute the annual energy consumption into each month. In the residential sector, energy is mainly used for cooking which was assumed to be constant during a year, and space heating which is dependent on the local temperature. In rural locations, conventional energy sources such as combustion of firewood, straw, or coal are widely used for both cooking and heating among all the seasons (Edwards et al., 2007). A linear regression model was subsequently built for per-capita residential energy consumption with the annual temperature as an independent parameter. The details on how the model was established and validated are presented in the Supplementary Material. Heat was provided by central heat-generating facilities in most urban districts during the winter, which was well documented by energy statistical yearbooks (NBSC, 2005a). According to government regulations, heat is provided when the ambient temperature falls below 5 °C (<http://www.gov.cn>). For simplicity, the amount of coal consumed by heat-generating facilities was evenly distributed into the time period when heat was provided. In order to further evaluate our model and assumptions, the statistical data for the residential energy consumption for all of the seasons during 2006 in Beijing and the survey data for the monthly straw consumption in Jilin province were used to validate our results (Qin et al., 2007; Beijing Statistical Bureau).

Since open burning of agricultural wastes only occurs during harvest seasons, emission amounts from this source are assumed to be zero during other times of the year. Only burning of wheat, rice, and corn straw was considered in this study because of their large planted areas and output in China (NBSC, 2005b). The harvest time of these crops in different places of China was collected from the literature.

The monthly biomass burned from forest and grassland fires was calculated based on the Global Fire Emission Database (Giglio et al., 2003). This data set has a spatial resolution of $1^\circ \times 1^\circ$, with monthly burned area, fuel loads, combustion completeness, and fire emissions (carbon, CO₂, CO, etc.). The actual combustion biomass for tropical and extra-tropical forest and grassland was calculated from the CO₂, CO and CH₄ emissions and their emission factor matrix.

The monthly emission from all sources was calculated based on the monthly emission activity strengths and emission factors, which were gathered from our previous emission inventories and considered constant throughout the year (Zhang and Tao, submitted for publication-a). In our previous study, the emissions from various sources from the entire country were distributed into a regular grid system using population, GDP, and land cover types data as proxies (Zhang and Tao, submitted for publication-a). The monthly proportions of energy consumption were multiplied with the annual emission densities, and the spatial distribution of emissions for each month was also achieved.

3. Results

3.1. Regression model for energy consumption and temperature

The monthly emission activity strength data were examined with regression models for residential solid energy consumption (including biofuel and coal) in rural districts and for coal consumption for domestic heating in urban districts. The regression model for the residential energy consumption in rural areas and the annual average temperatures for each province in China are shown in Fig. 1. Due to the great variance in agricultural outputs, forest coverage rates, and coal production among these provinces, the energy structures vary drastically among the provinces. Although the rural districts in most provinces largely rely on biofuel, including straw and firewood, the contributions from these two energy sources vary remarkably. For instance, firewood contributed more than 90% of the total residential energy consumption in rural Fujian in 2004, and less than 10% in Jiangsu (NBSC, 2005a). Similarly, rural Hainan consumed nearly no coal, while more than one-third and about one-half of the rural residential energy was supplied by coal for Hebei and Shanxi provinces, respectively (NBSC, 2005a). Therefore, the total residential energy consumption summed from the firewood, straw and coal consumptions by the weight of heat values, instead of each single energy source, was used as the dependent parameter in our model.

As shown in Fig. 1, a linear regression model can be built from the provincial data after deleting several outliers:

$$E_{\text{aver}} = -8.29 \times 10^{-3} T_{\text{aver}} + 0.406, r^2 = 0.32, n = 22 \quad (1)$$

where E_{aver} (ton of coal equivalent per capita) is the total rural residential energy consumption per capita for a province, and T_{aver} (degree Celsius) is the annual average temperatures of corresponding provinces.

3.2. Proportion calculation and validation

The proportion of energy consumed during each month based on the annual energy consumption can be then calculated based on Eq. (1) using the following equation:

$$f_i = \frac{1}{12} \frac{-8.29 \times 10^{-3} T_i + 0.406}{-8.29 \times 10^{-3} T_{\text{aver}} + 0.406} \quad (2)$$

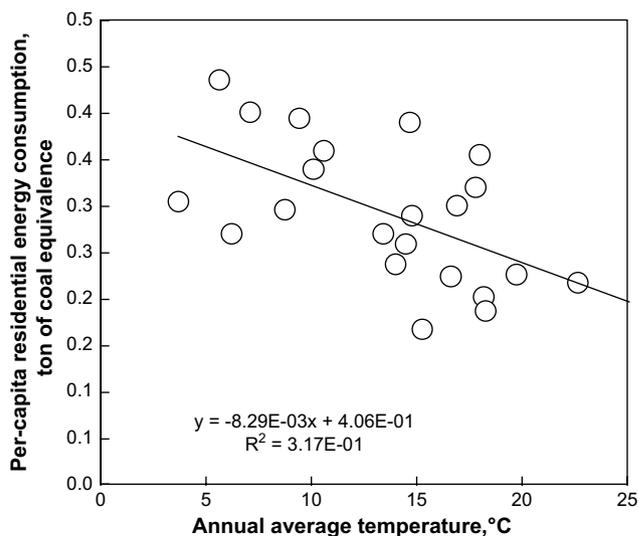


Fig. 1. Relationship between per-capita residential energy consumption and annual average temperature for the provinces of China.

where f_i is the monthly proportion, and T_i is the average monthly temperature. When T_i was larger than 20 °C, a value of 20 °C was assigned. Moreover, due to data limitation, the monthly proportions for all of the three solid fuels were assumed to be the same as calculated by Eq. (2).

Based on Eq. (2), the monthly proportion of residential energy consumption was calculated for all the model grids of China. For different parts of China, the variances of these proportions during the same year changed greatly. In Heilongjiang province, the proportion for January was 13%, which was about 2.4 times higher than that in the summer. A similar degree of seasonal variation was observed in other provinces located in northern China, such as Jilin, Liaoning, Inner Mongolia and Xinjiang. However, almost no seasonal variation was observed in the southern provinces, such as Guangdong and Hainan. In these provinces, the ratio of the coldest month and hottest month is both about 8.3%, very close to the flat average monthly proportion of one-twelfth. The percentages of other provinces are somewhere between these two extremes. For instance, the ratio of the proportions of January and July in Beijing was about 1.8, which was quite typical for northern China. If accounting for the entire country of China, an average variance of 1.6-folds occurred between the highest and lowest values of the residential solid fuel consumption.

Based on a survey conducted by Qin et al. (2007) in Jilin province, the proportion of straw consumption for each month was calculated, and the calculated values were compared with our model values in Fig. 2a. All the data points fall around the 1:1 line in this figure, indicating a good agreement between these two data sets. Our model results were also compared with the statistical data in Beijing (Beijing Statistical Bureau), and the monthly proportion was summed for each season and is shown in Fig. 2b. Our model slightly over- and underestimated the proportions of the first (January–March) and third (June–August) season, respectively, but in general, a good agreement can still be achieved.

When compared with other emission inventories that took into account seasonal variation, it was found that the variance among months in our research was quite close to the estimate of Cao et al. (2006). In their emission inventory of black carbon and organic carbon in China, which was also mainly emitted by residential solid energy consumption, on average a variance of 1.4-folds was reported between the months with the largest and smallest emission. However, these ratios were significantly lower than other results reported in the literature. Streets et al. (2003) assumed about 5-folds of variance between the hottest and coldest months in all of Asia, and Liousse et al. (1996) assumed there was 2.8-folds of variance in the northern hemisphere. Based on the data from

surveys and the statistical yearbook of China, both these emission inventories might have overestimated the seasonal variation of energy consumption in China.

3.3. Seasonal variation of the PAHs emissions in China

The emissions of PAHs and those from various sources for each month in the year 2003 are shown in Fig. 3. As shown in Fig. 3a, higher emissions occurred during the winter season, and the peak value was found in January. Much smaller emissions were found in the warmer seasons, the lowest monthly emission was found in May, June, and September. A smaller peak occurred in the summer season, especially during July and August. In all, the largest monthly PAHs emission was about 0.3 times more than the smallest one in China.

The pattern of overall seasonal variation of PAHs emission was largely caused by residential energy consumption. This includes domestic biofuel, coal, and industrial coal burning for heat supply, as these sources contributed the most to the total emission of PAHs and also had significant seasonal variations (Zhang and Tao, submitted for publication-a).

Another important source of seasonal variation is biomass burning. This includes the burning of agriculture residues on open fields and naturally occurring forest and grassland fires. Agricultural residues' burning mainly occurs during the harvest period in the summer when large amounts of straw are yielded. Furthermore, the planting for the next season happens during the same time period, and a large quantity of straw is subsequently burned in order to save time and labor, especially in the more developed districts (Cao et al., 2006). The harvest time changes drastically depending on the type of straw and regions. Corn straw is usually burned during September and October in northern China, and during July and August in southern China. The harvest time also varies for wheat straw, and continuously changes from May in southern China to August in northern China. For rice straw, the harvest times are more complex, because there are multiple harvest times in a year in the semi-tropical and tropical climate zones. Besides July and August in southern China and August and September in northern China, rice straw is also widely burned in March and April in Guangdong and Hainan provinces of South China (EBCAY, 2005). If the seasonal variation is not considered, the PAHs emission only contributes 2.4% of the total emission in annual average, but the short duration in which agricultural wastes burning occurs makes it an important factor in the seasonal variation patterns of PAHs emissions in China.

The PAHs emissions from wild fires are strongly influenced by local precipitation and wind. In general, the strong precipitation

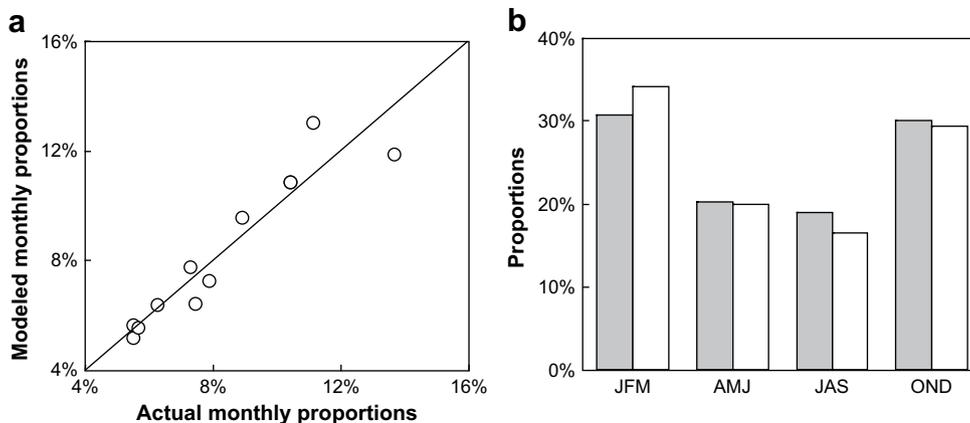


Fig. 2. Comparison of the model predicted and actual monthly proportion of energy consumption, (a) for the survey data of Jilin Province; (b) for the statistical data of Beijing (JFM: January–March; AMJ: April–June; JAS: July–September; OND: October–December).

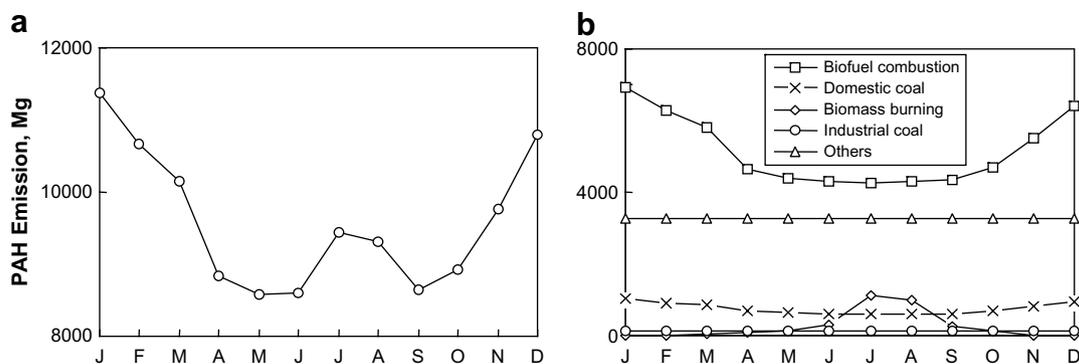


Fig. 3. Monthly emission of PAHs in China (a: total emission; b: individual sources).

and snow cover usually depress the outburst of forest or grassland fires during the summer and winter, respectively (Yuan et al., submitted for publication). The strong wind in the spring and the large amounts of accumulated biomass in the fall create intense wild fires (Sinomaps Press, 2005). Therefore, the peak values for wild fire emissions occur during the spring and fall, especially in the spring. Wild fires contribute much less to the overall seasonal variation than open straw burning, because they had lower annual emissions, but wild fires can greatly influence the environmental condition on a time scale of several days (Yuan et al., submitted for publication).

3.4. Seasonal variation of PAHs emission in different regions of China

Great differences exist among the local climate, land cover, and agricultural activities in different places of China. The climate pattern changes from tropical and semi-tropical in the south to temperate and frigid-temperate in the north, and also from typical monsoon in the east to continental in the west (Sinomaps Press, 2005). The variations of temperature and precipitation within 1 year are also quite unique in different places of China, which also causes different patterns of seasonal variations of PAHs emissions. To evaluate the variation among the different regions, the emissions and source contributions for each month from six zones (Northeast, North, Northwest, South, Southwest and Tibetan plateau). These zones were divided according to their geographical positions and socio-economical conditions, and are shown in the Supplementary Material) were individually calculated and are shown in Fig. 4.

The northeast and northwest areas of China have the highest latitude of all the six regions, and the monthly average temperature in January can be as low as -20°C , which results in a large portion of the residential energy being used for space heating in the winter. Therefore, great seasonal variation of PAHs emissions from residential energy consumption was observed in these regions. Northeastern China has vast areas of forest; the total area of forest in the three provinces in this region is over 35 million hectares (NFBC, 2005). This covers about 37% of the total land area, which is more than twice of the average forest cover ratio of China (NFBC, 2005). The emission of PAHs from forest fires in northeastern China made up a contribution of about 15% of the total emissions of this region during the spring-time, and even more than 25% in May. Xinjiang is an important crop production region of China, especially for cotton and sugar beets, and ranks first in all the provinces of China (EBCAY, 2005). Since this area has a relatively small population, residential energy consumption is lower and makes the emission from agricultural wastes burning more important in this region. A significant peak was observed during July and August in

the northwestern part of China, and similar emission pattern was observed in the Tibetan Plateau region. Unlike other regions, northern China has the largest small-scale coke production (Zhang and Tao, submitted for publication-a), which makes a large contribution to PAHs emissions without seasonal variation, and masks the seasonal variations of other emission sources. In southern and southwestern China, the emission comes mainly from biofuel consumption which has no significant seasonal variation due to the persistent relatively high temperatures throughout the year, and little energy is used for space heating.

3.5. The spatial distribution of PAHs emissions in China during different seasons

The spatial distributions for different seasons of the major emission sources are shown in Fig. 5. For biofuel and domestic coal (the left and middle columns in Fig. 5, respectively), a very similar spatial pattern was observed. Except for the difference caused by the variation of temperature, the spatial distribution of emission density remained the same among different seasons. Biofuel is only burned in the rural areas (Zhang and Tao, submitted for publication-a), but coal is an important energy source in both the urban and rural areas (NBSC, 2005a). During 2003, 36% of the coal consumption for domestic heating for the entire country was in the urban areas (NBSC, 2005a). With urban areas encompassing only 0.15% of the total area of the entire country, a much higher emission density from domestic burning was observed (National Construction Agency of China, <http://www.cin.gov.cn/>). The areas of high emission density in urban areas can be clearly observed in the domestic coal map, but not in the biofuel map.

For biomass burning, the spatial distribution changed drastically among seasons (right column of Fig. 5). Due to significantly higher emission factors of PAHs for wheat straw compared with other kinds of crops, the emission density was much higher in the areas where wheat was planted, mainly the Northern China Plain and the Sichuan Basin. These two regions accounted for about 64% of the wheat yield in China during 2003 (EBCAY, 2005). Although large quantities of rice straw are burned in fields in southern China, the much lower emission factors caused the emission density relatively low (Zhang and Tao, submitted for publication-a). The emission from wild fires was much smaller than that from straw burning, but very concentrated in northeastern China during the spring, especially in Heilongjiang and Inner Mongolia. In southern China during the winter, the emissions from wild fire were also concentrated, resulting in the spatial distribution of emission density from biomass burning during the colder half of the year being significantly different from the other half of the year.

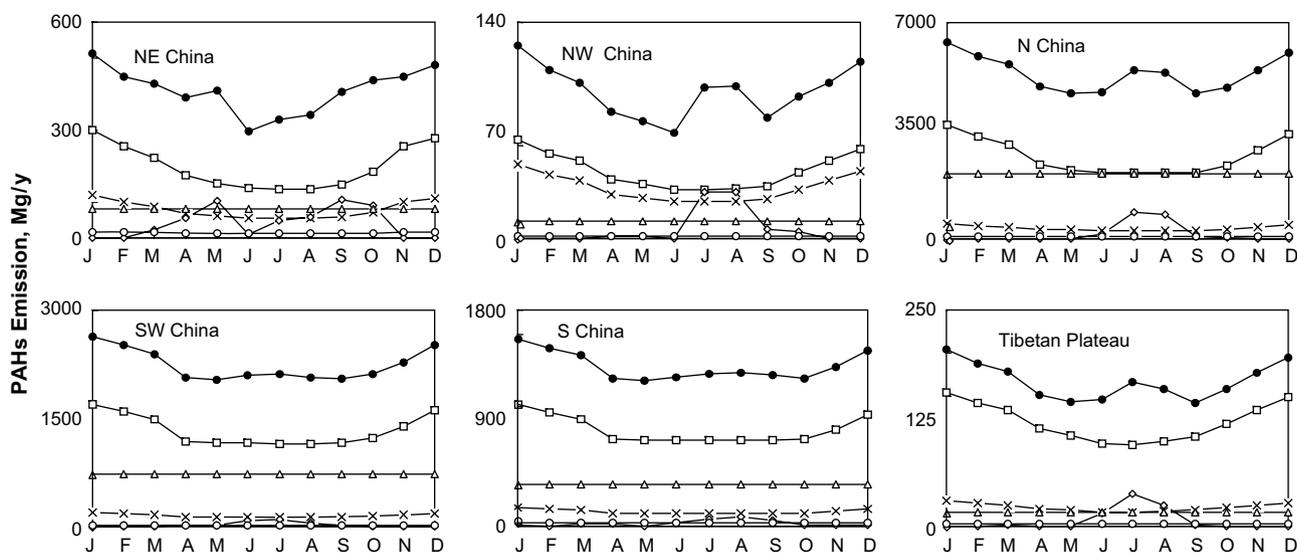


Fig. 4. The monthly PAHs emission rate of several major sources in different regions of China (●: total emission; □: biofuel combustion; ×: domestic coal combustion; ◇: biomass burning; ○: industrial coal; △: others).

4. Discussion

4.1. Uncertainty analysis

Great uncertainties exist for the seasonal variations of the emission of PAHs in China. The first source of uncertainty comes from the annual emission inventory. A variance of about 14, 15, 20 and 30% of the median emissions was estimated for residential straw, firewood, coal combustion and open burning of agriculture wastes, respectively, based on the Monte Carlo simulation that was performed in our previous study (Zhang and Tao, submitted for publication-a). When the annual emissions was subdivided into emission per season or month, this uncertainty increased by about 2- to 4-folds, respectively, since the variance is inversely proportional to the square root of the sample size.

The proportion of emissions from each month compared to the annual volume was calculated based on a regression model in this study by assuming these proportions are independent from all factors except temperature. This assumption does not take into account the different space heating techniques used, non-solid fuel consumption and the living habits of different places in China. For example, the energy utilization efficiency of the newly designed kang (a bed with a fire beneath) is about 2-folds higher than the conventional kangs (Zeng et al., 2007). The energy efficiency of energy-saving stoves that are now widely used in rural China is also about 20% higher than the older stoves (Lin, 1998). These high efficiency space heating instruments cause a lower proportion of energy consumed during the winter.

Another source of uncertainty comes from the different types of energy used at different times during the year, mainly in the urban and semi-urban areas, where coal is used for space heating in the winter, but gaseous energy and electricity are used for cooking in the other seasons. For example, in Xi'an, about 76.4% of the buildings are heated by small-scale boilers or even small coal stoves, especially in low income level families, instead of by a centralized heat-generating facility (Xi'an government, <http://www.xa.gov.cn>). However, based on a survey conducted in January, more than 67.7% of the families in Xi'an at a low income level are using liquefied petroleum gas for cooking (XASB, 2006). This kind of energy switch may result in higher seasonal variations than estimation by the regression model. However, the lack of necessary data prevents further evaluation and modeling. Similar energy switching scenarios also occur

among straw, firewood, coal, and gas fuels in rural areas (Edwards et al., 2007).

Finally, some other sources which were considered constant during a year in this study may also change during different seasons. For example, it was reported that the small-scale coke production was relatively low when the peasants were busy during harvesting or planting time (Kong et al., 2005). Also, the emission factor of traffic oil consumption is higher during the winter than that in the summer because of the increased emission produced from starting cars in cold weather (Tan et al., 2006). However, the seasonal variations of these sources were not considered in our study because of the lack of necessary data.

4.2. Influence of the seasonal variation of emission on the exposure of PAHs

Inhalation from ambient and indoor air is an important pathway for the exposure of pollutants to the human body. Based on an estimation by the World Health Organization, indoor smoke from solid fuels and urban outdoor air pollution are both very important risk factors that carry large disease burdens, and could explain about 3% of the total healthy year lost in the world in 2000 (Smith, 2006). When considering China individually, these two factors ranked 6th and 12th out of all the risk factors and contributed to about 4% to the burden of disease in China in 2000 (Zhang and Smith, 2007). PAHs made a very important contribution in both of the pathways (Zhang and Smith, 2007). Based on a study conducted in Tianjin, inhalation from indoor and ambient air contributed about 15% of the total exposure of PAHs, and made an excess carcinogenic risk of 2.3×10^{-6} during the year 2003 (Tao et al., 2006). Due to the great difference of energy consumption and PAHs emissions that exists between the winter and summer, the exposure of PAHs from inhalation is expected to have significant seasonal variation. Furthermore, the exposure is also influenced by other factors. For indoor pathways, the indoor air concentration of PAHs is greatly influenced by the ventilation condition. Based on a tracer study conducted in Boston, the median air exchange rate is about 2-folds higher during the non-heating season than the heating season (Zota et al., 2005). The poor ventilation greatly increases the risk induced by PAHs in the winter, especially in rural families where residential combustion in the house is a main emission source (Zhang and Tao, submitted for publication-a).

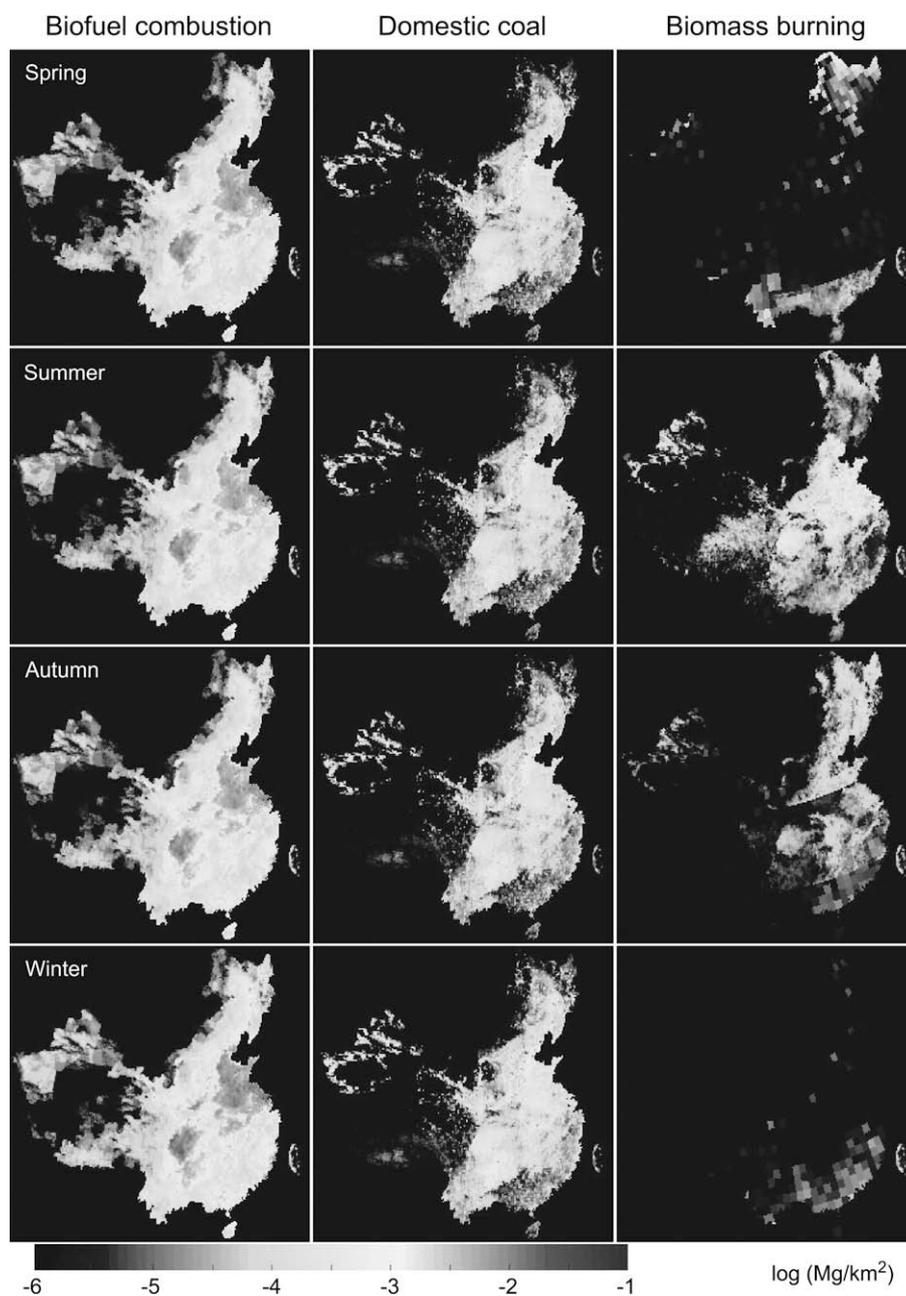


Fig. 5. PAHs emission density maps for the major emission sources in different seasons, the colorful version is available in the Supplementary Material.

Similarly, the ambient concentration is influenced by many other factors, including wind, temperature, stability of the atmosphere, concentration of radicals and aerosols, etc. Guo et al. (2003a,b) monitored the particulate phase concentration of PAHs in Hong Kong, and found that the prevailing northeasterly wind brought large amount of PAHs from the Asian continent during the winter-time, when the emissions were higher, resulting in about a 10 times difference of concentrations between the winter and summer seasons.

4.3. Influence of the seasonal variation of emission on the outflow of PAHs

In addition to the contamination of the local environment, PAHs can undergo long range transport and make contribution to the ambient concentration of PAHs in places located downwind from China. The outflow flux of PAHs from China is greatly influenced by

local emissions in China and meteorological conditions. Lang et al. (2007) has investigated the outflow of atmospheric PAHs from Guangdong province using a forward trajectory statistical method, and found that the outflow direction was quite different depending on the seasons. Briefly, a southerly and northerly transport was observed to dominate during the winter and summer, respectively, while transpacific long range transport occurred mainly under certain meteorological conditions in the winter. Similarly, Liu et al. (2007) also found that 35.4% and 26.3% of the forward trajectories were toward the eastern and southern directions during the winter-time, respectively. Besides the computation of the source regions, observations in some receptor locations also showed a higher probability of the outflow of PAHs from China during the winter-time. A study conducted in Gosan (an island located at the south of South Korea) found that the probability of air parcels that originated from mainland China was about 88% during the cold season, and 35% during the warm season (Lee et al., 2006). This high

probability of outflow from China coincides with the high emissions of China, and makes much larger outflow fluxes during the winter-time. However, the emission from agricultural wastes burning is mainly concentrated in the summer, when southerly winds dominate in the North China plain, and the relatively lower wind speed suppresses the outflow of PAHs (Lang et al., 2007).

5. Conclusions

The magnitude of the seasonal variation of PAHs emission from China in 2003 was estimated to be about 1.3-folds between the months with the largest and the smallest emissions. Residential solid fuel combustion including straw, firewood and coal dominated the pattern of the overall seasonal variation with peak in the winter-time. The seasonal variations contributed by other sources were much smaller. The higher PAHs emission occurring in the winter-time can cause higher exposure of PAHs to human and make larger outflow flux from China in the winter.

This inventory can provide basic information for policy makers for better control the emission of PAHs, and help researchers to better understand the magnitude of the seasonal variation of PAHs emission. More directly, this inventory can be used by fate and transport modelers who take seasonal variation into consideration.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.envpol.2008.06.017.

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