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Polycyclic and nitro-polycyclic aromatic hydrocarbon pollution characteristics and carcinogenic risk assessment of indoor kitchen air during cooking periods in rural households in North China

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Abstract

Medium-flow atmospheric samplers were used to collect particulate (PM_{2.5}) and gaseous samples from the indoor kitchen of each of 35 randomly selected rural houses in North China while a meal was being cooked. The concentrations of 16 polycyclic aromatic hydrocarbons (PAHs) and 9 nitro-PAHs (NPAHs) in the samples were quantified by high-performance liquid chromatography (HPLC). This study provided the real PAH and NPAH pollution characteristics and carcinogenic risk produced by cooking in rural indoor kitchens in North China. The mean PAH and NPAH concentrations in air in the indoor kitchens during cooking periods were 4049.1 and 1741.6 ng/m³, respectively. The PAH and NPAH concentrations were lower in the particulate phase than the gaseous phase. The mean PAH and NPAH concentrations were much higher for cooking using coal than for cooking using liquefied petroleum gas (LPG) or electricity. The PAH and NPAH benzo[a]pyrene toxic equivalent (TEQ_{BaP}) concentrations for cooking using coal were 1823.3 and 2760.9 ng/m³, respectively. Lower PAH and NPAH concentrations were found in kitchens with than without range hoods. Range hoods decreased the PAH and NPAH TEQ_{BaP} concentrations by 68.8% and 61.9%, respectively. Appropriate fuel and ventilation choice will improve air pollution in indoor kitchens during cooking. The results provide important evidence for changing cooking habits and developing policies for cooking in rural China.

Keywords Indoor kitchen air \cdot Rural households \cdot Cooking pollution \cdot PAHs \cdot NPAHs \cdot Benzo[a]pyrene toxic equivalence (TEQ_{BaP})

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are some of the most prominent carcinogenic pollutants of ambient air. Each PAH contains two or more aromatic hydrocarbon rings fused

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² State Environmental Protection Key Laboratory of Food Chain Pollution Control, Beijing Technology and Business University, Beijing 100048, China together (Hao et al. 2016b; Huang et al. 2015; Li et al. 2018). Nitro-PAHs (NPAHs) are derivatives of PAHs. Most NPAHs are more mutagenic and carcinogenic than the parent PAH (Bandowe and Meusel 2017; Lin et al. 2015). In recent decades, PAHs and NPAHs have attracted much attention because they are toxic to humans (Ali et al. 2017; Chen et al. 2016; Shen et al. 2012; Shen et al. 2017; Zhao et al. 2019b). The main sources of PAHs to the environment are incomplete combustion and pyrolysis of solid fuels, and these sources also supply a proportion of the NPAHs found in the environment (Osano et al. 2020; Thuy et al. 2012; Wang et al. 2016). Various fuels are used for cooking in households in rural areas (Smith et al. 2014), and some are important sources of pollutants in indoor kitchens (Munyeza et al. 2019). However, only 20.4% of households in rural China use a range hood (a fan system for extracting fumes produced during cooking) (National Bureau of Statistics of the People's Republic of China 2018). A typical range hood found in a rural kitchen

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is shown in Fig. S2c. Very high PAH and NPAH concentrations have been found in air in indoor kitchens (Ding et al. 2012; Zhang et al. 2019). High concentrations of pollutants in fumes produced during cooking can pose serious long-term health risks to the people cooking (See and Balasubramanian 2006). In China, cooking is more frequently performed by females than males, and only 4% of females smoke tobacco (Mu et al. 2013). Lung cancer has a high incidence rate in Chinese females (Mu et al. 2013), which may be related to pollution caused by indoor cooking (Chen et al. 2019; Yao et al. 2015). It is therefore important to investigate the risks posed by PAHs and NPAHs released during cooking in rural indoor kitchens.

A number of studies of fumes emitted during cooking have been performed at different sites (Chen et al. 2018; See and Balasubramanian 2006; Zhang et al. 2017; Zhao et al. 2019a). See and Balasubramanian (2006) investigated fine particle and metal concentrations in air during cooking and noncooking periods at a cooked food stall in China. Chen et al. (2018) and Zhao et al. (2019a) performed orthogonal experiments to investigate indoor contamination under different cooking conditions in kitchens in residential buildings. Zhang et al. (2017) collected samples of fine particulate $(PM_{2,5})$ produced while cooking dishes using five commonly used oils in a kitchen in an apartment. However, these and other previous studies were focused on samples collected in commercial buildings or during simulated cooking activities in residential buildings. Studies of pollutants emitted during normal cooking activities in residential buildings are still lacking. Personal cooking habits (Abdullahi et al. 2013), the cooking fuel (Fischer and Koshland 2007), kitchen ventilation (Mu et al. 2013), and other factors will affect pollutant concentrations in indoor kitchens. Pollutant concentration patterns and temporal variations will be different in kitchens in residential buildings and commercial kitchens, and simulated cooking activities will not fully represent normal cooking activities. Investigations of pollutant emissions and the factors affecting pollutant emissions in indoor kitchen air during cooking in real domestic kitchens in China will provide data more appropriate for assessing and controlling pollutant emissions in kitchens than data acquired in previous studies.

Various harmful air pollutants can be produced during cooking, but many studies have been focused on PAH concentrations in particulate matter produced during cooking because of the toxicities of PAHs (Chen et al. 2018; Du et al. 2018a; Saito et al. 2014; Zhang et al. 2017; Zhao et al. 2019a). However, higher PAH concentrations have been found in the gas phase than the particulate phase in some studies (Li et al. 2003; Zhao et al. 2011), and this should be taken into account when studying PAH concentrations in indoor air. Previous studies of NPAHs in cooking fumes have mainly been focused on NPAH concentrations (Wu et al. 1998), but the NPAH characteristics and factors affecting NPAH pollutions

have not vet been studied. Although concentrations of NPAHs are much lower than parent PAH concentrations (Bandowe and Meusel 2017), NPAHs are toxic and pose risks to human health (Abbas et al. 2018; Ali et al. 2017). It is important to improve our understanding of PAH and NPAH emissions during cooking in real domestic kitchens to allow the carcinogenic risks posed to people cooking to be assessed. The aims of the study described here were to determine PAH and NPAH concentrations in air in kitchens in which different cooking fuels and ventilation conditions were used and to assess the carcinogenic risks posed by PAHs and NPAHs in indoor kitchens during cooking. To achieve this, air samples were collected in 35 indoor kitchens in rural areas in Beijing and Shanxi Province. The concentrations of 16 PAHs and 9 NPAHs in the particulate and gaseous phases of indoor air in the kitchens and outdoor air during cooking were determined. The results will improve our understanding of PAH and NPAH emissions during cooking in domestic kitchens in rural China and provide evidence for developing policies to control the risks posed by PAHs and NPAHs to people cooking.

Materials and methods

Sample collection

Samples were collected in rural areas in Yanqing District in Beijing and in Yunzhou District in Datong City, Shanxi Province, both in North China. The sampling areas are shown in Fig. S1. Traditional solid fuels (e.g., coal) and clean fuels (e.g., liquefied petroleum gas (LPG) or electricity) are used for cooking in the study areas. The samples were collected in winter (February 2019, when the average ambient temperature in North China was approximately -10 °C). Each sample was collected while a meal (lunch or dinner) was being cooked. Fuel was used only for cooking, while a sample was being collected, to avoid indoor air pollution caused by heating.

A total of 35 households (19 in Yanging and 16 in Datong) were randomly selected, and the householders gave permission for samples to be collected. Each household had a front vard. Details of the households are provided in Table S1. Particulate and gaseous samples were collected using modified LY-2030 medium-flow atmospheric samplers (LaoYing, Qingdao, China) in the indoor kitchen and outside each household. The sampling flow rate was 100 L/min. Quartz filter (QF, d=90mm, 2500QAT-UP, PALL Corp., USA) and polyurethane foam (PUF, 38 × 80 mm, QDXAO, Qingdao, China) were used to collect each particulate and gaseous sample, respectively. To avoid interference from other sources of pollution, the window was opened for 30 min before sampling in the kitchen, and the kitchen doors and windows were kept closed while the samples were collected. A stationary air sampler was installed in a kitchen approximately 1.5 m from the

ground, fixed with a tripod, while simulating the range of the breathing area of the cook as much as possible, approximately 30 cm from the horizontal position of the cooktop. An outdoor sampler was placed 1.5 m from the ground in the center of the front vard of the house. The indoor sampling time started when cooking commenced and ended 6 h later when no more fumes were detected (Zhang et al. 2017), and the outdoor sampling time was 12 h. The sampling periods were selected to attempt to ensure that the PAH and NPAH concentrations in the samples were able to be detected using the analytical method. The sampling procedure at a sampling site is illustrated in Fig. 1, and pictures during sampling and some typical indoor kitchens in which different cooking fuels were used are shown in Fig. S2. In total, 35 particulate and 35 gaseous samples were collected in indoor kitchens, and 17 particulate and 17 gaseous samples were collected from outdoors. Immediately after sampling, each sample was sealed in clean aluminum foil, transported to the laboratory in a refrigerator, and then stored at - 10 °C to prevent the PAHs and NPAHs on the QF or PUF from volatilizing.

Sample extraction and composition analysis

The sample processing and analysis procedures were similar to procedures used in previous studies (Cao et al. 2017; Hao et al. 2018). Briefly, a QF or PUF sample was cut into pieces and then extracted with solvent with ultrasonication. The extract was rotary evaporated, cleaned up by column chromatography, and then evaporated to a specified volume. The concentrations of 16 PAHs and 9 NPAHs in the extracts were determined by high-performance liquid chromatography (HPLC). The sample processing and analysis methods are described in detail in Supplementary Material.

Quality control

The QFs and PUFs were pretreated to remove organic impurities before being used to collect samples. Each QF was baked at 550 °C in a muffle furnace for 12 h and then weighed using a high-precision balance in a constant temperature and humidity environment. Each PUF was washed three times with dichloromethane and then dried. Each prepared QF and PUF was then sealed in aluminum foil and stored in a desiccator until use.

The experimental procedures, operating methods, and standards were unified for each laboratory staff. The samples were processed in batches, and a whole-process blank was analyzed with each batch of 20 samples. The blank samples were treated in the same way as the samples. Duplicate samples were collected and analyzed to assess differences between the sampling instruments. Duplicate samples and procedural blanks were used to correct the concentrations of PAHs and NPAHs. The correlation coefficient (R^2) for the standard curves for the 16 PAHs and 9 NPAHs were all > 99.6%. The 16 PAH and 9 NPAH recoveries determined by analyzing pre-processed QF and PUF samples spiked with standards, as shown in Table S5. The detection and quantification limits were calculated as the concentrations corresponding to 3 and 10 times the standard deviation of the blank noise, respectively. The detection limits and quantification limits of the 16PAHs and 9NPAHs are shown in Table S5.

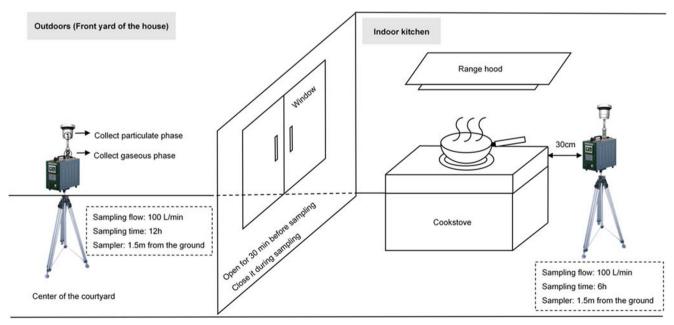


Fig. 1 Diagram illustrating indoor kitchen and outdoor sampling

Risk assessment

The benzo[a]pyrene (BaP) toxic equivalent concentration (TEQ_{BaP}) was used to express the PAH and NPAH concentrations in terms of carcinogenic risk. The TEQ_{BaP}s for the individual species were calculated using the equation (Bandowe et al. 2014):

$$\text{TEQ}_{\text{BaP}} = \sum_{i=1}^{i=n} C_i \times \text{TEFi},$$

where Ci is the actual concentration of PAH or NPAH i and TEFi is the carcinogenic potential of PAH or NPAH i relative to the carcinogenic potential of BaP. The TEFs for the PAHs and NPAHs are shown in Table S4.

Results and discussion

PAH and NPAH concentrations in indoor kitchens during cooking periods

The mean PAH and NPAH concentrations in the gaseous phase, particulate phase, and total (gaseous phase plus particulate phase) in the indoor kitchen air samples (n = 35) during cooking and concentrations found in previous studies are shown in Table 1. The individual PAH and NPAH concentrations are shown in Table S6. To avoid the difference in the concentrations in the indoor samples caused by the distinction in background concentrations between the two sampling areas, the concentrations found in indoor kitchens samples in this study were calculated by removing the concentrations found in outdoor samples.

The mean PAH concentrations in the gaseous phase, particulate phase, and total air indoor kitchen samples during cooking were 3105.6 ± 3881.7 , 943.5 ± 1918.3 , and 4049.1 \pm 5764.7 ng/m³, respectively. The mean NPAH concentrations in the gaseous phase, particulate phase, and total air indoor kitchen samples during cooking were 1041.6 \pm $1737.7, 700.0 \pm 1599.5, \text{ and } 1741.6 \pm 3320.7 \text{ ng/m}^3, \text{ respec-}$ tively. The mean PAH and NPAH concentrations in the particulate phase were lower than the mean concentrations in the gas phase. BaP is the only PAH included in the current Chinese indoor air quality standard. The current maximum BaP concentration allowed in indoor air in China is 1.0 ng/ m³ (GB/T 18883-2002). The mean total BaP concentrations in the indoor kitchens during the cooking periods was $151.8 \pm$ 271.6 ng/m^3 , which was 151.8 times higher than the Chinese limit for indoor air. Lv et al. (2009) found total BaP concentrations in air in rural houses in Yunnan Province of 7.7-380.3 ng/m^3 (mean 95.7 ng/m^3), which were also much higher than the Chinese limit. These results indicated that the residents of rural areas in China are exposed to serious PAH pollution within their homes.

High PAH and NPAH concentrations similar to the concentrations we found have been found in indoor kitchens in rural areas in previous studies. Huang et al. (2018) reported PAH concentrations in the particulate and gaseous phases during cooking in kitchens in rural Tibet of 33 ± 31 ng/m³ and 4420 ± 2280 ng/m³. Wu et al. (1998) found NPAH concentrations in the particulate phase in cooking oil fumes of 900–3400 ng/m³. However, the mean total PAH concentration (2511.6 ng/m³) and NPAH concentration (6.0 ng/m³) found in indoor kitchen air by Ding et al. (2012) were markedly lower than the results of this study, indicating that cooking may increase PAH and NPAH concentrations in

Table 1Mean polycyclic aromatic hydrocarbon (PAH) and nitro-PAH (NPAH) concentrations (ng/m^3) found in indoor kitchen air during cooking inthis study and the conclusions of other studies

Compounds	Location	Time	Mean concentration			References
			Gaseous phase	Particulate phase	Total	
PAHs	Rural indoor kitchen	Cooking period (for one meal)	3105.6 ± 3881.7	943.5 ± 1918.3	4049.1 ± 5765.7	This study
	Rural indoor kitchen	Cooking period (for one meal)	33.0 ± 31.0	4420.0 ± 2280.0	-	Huang et al. (2018)
	Rural indoor kitchen	Daily average concentration in summer	1452.4	1059.1	2511.5	Ding et al. (2012)
	Rural household	In spring	-	-	422.6-4826.1	Lv et al. (2009)
NPAHs	Rural indoor kitchen	Cooking period (for one meal)	1041.6 ± 1737.7	700.0 ± 1599.5	1741.6 ± 3320.7	This study
	Rural indoor kitchen	Daily average concentration in summer	5.2	0.8	6.0	Ding et al. (2012)
	Cooking oil fumes	During oil heating	-	-	900.0-3400.0	Wu et al. (1998)

indoor kitchen air. At present, NPAH pollution during cooking has been studied little and requires further exploration.

The percentages of particulate and gaseous phases found in the PAHs and NPAHs in indoor kitchen air samples are shown in Fig. S3. The proportion of PAHs and NPAHs in the gaseous and particulate phase accounted for 76.7%, 23.3%, and 59.8%, 40.2%, respectively. The PAH and NPAH concentrations in the gaseous phase were 3.3 and 1.5 times, respectively, the concentrations in the particulate phase. Similar conclusions were drawn in previous studies of cooking fumes (Li et al. 2003; Yao et al. 2015) and domestic kitchen air (Ding et al. 2012). Although several scholars found that cooking was one of the most important particle-generating sources in the room (Buonanno et al. 2009; Lai and Ho 2008; Massey et al. 2012; Wan et al. 2011), the PAH or NPAH concentrations in the gaseous phase will further affect gas-particle partitioning (Pankow 1987), which will directly affect PAH and NPAH persistence and fate in the environment. The high gaseous phase PAH and NPAH concentration produced by cooking cannot therefore be ignored.

Composition distribution of PAHs and NPAHs in indoor kitchens during cooking periods

The mean contributions of the PAHs and NPAHs with different molecular weights to the total (gas plus particulate phase) PAH and NPAH concentrations in the indoor kitchen air samples are shown in Fig. 2a. Low molecular weight (LM-) PAHs (two and three ring PAHs), moderate molecular weight (MM-) PAHs (four ring PAHs), and high molecular weight (HM-) PAHs (five-seven rings) in indoor kitchens accounted for 46.0%, 35.4%, and 18.6%, respectively, of the total PAH concentrations. The LM-NPAHs, MM-NPAHs, and HM-NPAHs accounted for 42.8%, 45.3%, and 11.9%, respectively, of the total NPAH concentrations. LM-PAHs and MM-PAHs are the dominant PAHs, and the LM-NPAHs and MM-NPAHs are the dominant NPAHs produced during cooking in indoor kitchens (Du et al. 2018b).

The percentage of PAHs and NPAHs in the gaseous phase and particulate phase in indoor kitchens under different molecular weights is provided in Fig. 2b. The distributions of particulate phase in the LM-PAHs, MM-PAHs, and HM-PAHs were 7.3%, 19.7%, and 69.7%, and exhibited the following trend: as the molecular weight increased, the proportion of particulate phase PAHs increased gradually in total PAHs (Li et al. 2003). Figure 3 shows the contributions of the individual PAHs and NPAHs to the particulate and gaseous phase concentrations in the indoor kitchen. The main PAHs found in the particulate phase were benzo[b]fluoranthene (BbF), BaP, and benzo[k]fluoranthene (BkF), which contributed 17.4%, 11.5%, and 11.2%, respectively, of the total PAHs, and the toxicities of PAHs generally increase as the molecular weight increases (Li et al. 2017), indicating that controlling PAH emissions during cooking should be focused on removing PAHs in the particulate phase.

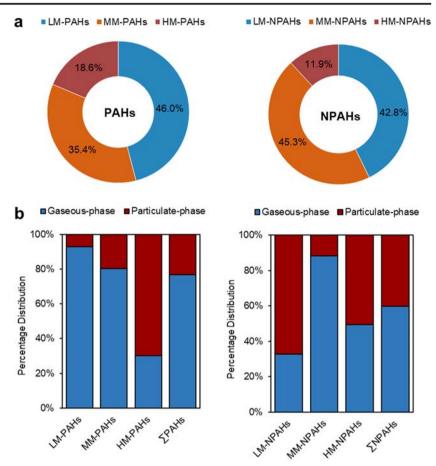
The distribution of particulate phase in the LM-NPAHs, MM-NPAHs, and HM-NPAHs were 32.8%, 88.1%, and 49.4%, respectively. The proportion of gaseous phase in the LM-NPAHs, MM-NPAHs, and HM-NPAHs were 67.2%, 11.9%, and 50.6%, respectively. 3-Nitrophenanthrene (3-Nphe) was the most abundant NPAH in the particulate phase (contributing 61.2%), and 1,3-dinitropyrene (1,3-Npyr) and 3-Nphe were the most abundant in the gaseous phase, contributing 30.9% and 14.5%, respectively. He et al. (2004) and Zhang et al. (2017) found high pyrene (Pyr) and phenanthrene (Phe) concentrations in Chinese air samples collected during cooking. Interestingly, the concentrations of derivatives of Pyr and Phe were also high in our samples.

Factors affecting the pollution characteristics in the indoor kitchen air during cooking periods

Cooking fuel

In areas with relatively constant climate and sociocultural factors (e.g., meteorological conditions, house structures, and stove types), the fuel type will be an important factor affecting indoor pollution (Fischer and Koshland 2007; Shen et al. 2010). The mean total PAH, total NPAH, and individual PAH and NPAH concentrations in the indoor kitchen air samples during cooking using different fuels are shown in Table S7. The average total PAH concentrations when coal, LPG, and electricity were used for cooking were 10750.1 \pm $9244.9, 2139.8 \pm 1761.9$, and 1846.0 ± 1043.3 ng/m³, respectively. The total PAH concentrations were ranked in the following order: coal > LPG > electricity. The MM-PAH and HM-PAH contributions to the total PAH concentrations were higher when coal was used (39.4% and 20.5%, respectively) than when LPG and electricity were used (Fig. S4a). Pyr and BbF were the dominant MM-PAHs and HM-PAHs, respectively. Du et al. (2018b) also found higher HM-PAH concentrations when coal was used than when electricity was used in indoor air. The mean total NPAH concentrations in indoor kitchens were also higher when coal was used (5239.5 \pm 5862.3 ng/m³) than when LPG was used (710.2 \pm 610.0 ng/ m³) and lowest when electricity was used (691.0 \pm 258.5 ng/ m³). Unlike for the PAHs, compared with LPG and electricity, the dominant NPAHs produced using coal were LM-NPAHs, with a proportion of 45.2% (Fig. S4b).

Figure 4 a shows the proportional distributions of the particulate and gaseous phase PAHs and NPAHs when using different fuels. The contributions of PAHs in the particulate phase to the total PAH concentrations when coal, LPG, and electricity were used were 28.9%, 15.3%, and 12.9%, respectively. The contributions of NPAHs in the particulate phase to **Fig. 2** a Proportion of different molecular weights of the total (gaseous plus particulate phase) polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs (NPAHs) in indoor kitchens and **b** distributions of the PAHs and NPAHs in the gaseous and particulate phase in the indoor kitchens under different molecular weights



the total NPAH concentrations when coal, LPG, and electricity were used were 43.7%, 34.2%, and 27.2%, respectively. The contributions of PAHs and NPAHs in the particulate phase to the total PAH and total NPAH concentrations were higher when coal was used than when LPG and electricity were used for cooking. $PM_{2.5}$ may be deposited deep in the alveoli of the lungs and can adversely affect human health (Che et al. 2014), so it is essential to control the use of coal for cooking in rural areas. In recent years, LPG has become the preferred cooking fuel for many rural families because it is convenient to use. However, Ding et al. (2012) found similar PAH concentrations in rural domestic kitchens in which LPG and firewood were used. The mean PAH concentration in our indoor kitchen air samples when LPG was used (2139.8 \pm 1761.9 ng/m³) was higher than the mean PAH concentration of 1400 \pm 490 ng/m³ found by Ding et al. (2012). It is therefore necessary to investigate pollutant emissions during cooking with fuels such

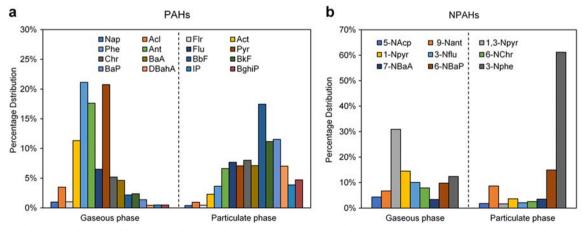
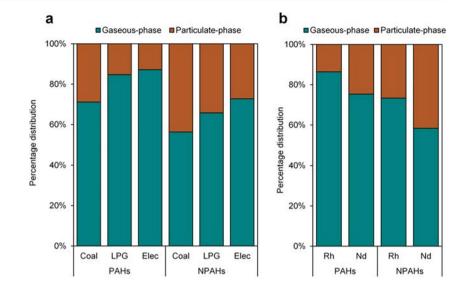


Fig. 3 a Contributions of the individual polycyclic aromatic hydrocarbons (PAHs) to the particulate and gaseous phase concentrations in the indoor kitchen and **b** contributions of the individual nitro-PAHs (NPAHs) to the particulate and gaseous phase concentrations in the indoor kitchen

Fig. 4 Polycyclic aromatic hydrocarbon (PAH) and nitro-PAH (NPAH) distributions in the particulate and gaseous phases under different influencing factors **a** when different fuels were used (LPG liquefied petroleum gas, Elec electricity) and **b** when different ventilation conditions were used (Nd no device, Rh range hood)



as biomass and firewood to determine whether LPG can be used as a "clean" cooking fuel. Like LPG, electricity is a convenient and clean fuel that has gradually become a common cooking fuel in rural areas. Electricity accounted for 58.6% of domestic energy use for heating and cooking in rural areas in China in 2017 (The Office of the Leading Group of the Third National Agricultural Census of the State Council, National Bureau of Statistics of the People's Republic of China 2017). However, the mean PAH and NPAH concentrations were high when electricity was used for cooking in our study, indicating that high-temperature mixing of cooking ingredients or heating of cooking oil will produce a certain concentration of pollutants (Hao et al. 2016a; Wu et al. 1998). Heating food and oil cause PAHs to be transferred from the food and oil to the air (Moret and Conte 2000). The oil temperature, amount of oil used (Abdullahi et al. 2013), and cooking method (Hao et al. 2016a) will strongly affect PAH emissions. The use of clean cooking fuels and cooking habits should be promoted in rural areas to decrease PAH and NPAH concentrations in the indoor kitchen air.

Ventilation condition

The ventilation conditions strongly affect contaminant concentrations in kitchen air during cooking. Mu et al. (2013) found that the housing layout and ventilation feature strongly affect the risks to human health. For an indoor kitchen in particular, the installation of a range hood is also a factor to affect the pollutant concentration. It can be seen from Table S7 that the mean total PAH and NPAH concentrations in the air in kitchens without range hoods during cooking were 4973.3 \pm 6615.3 and 2202.1 \pm 3849.5 ng/m³, respectively. The total PAH and NPAH concentrations in kitchens with range hoods were 1738.8 \pm 735.5 and 590.4 \pm 282.0 ng/m³, respectively, which were 65.0% and 73.2%, respectively, lower than the total PAH and NPAH concentrations in kitchens without range hoods. Good ventilation can minimize the accumulation of pollutants indoors (Kamal et al. 2011). Using a range hood during cooking periods can increase air flow between inside and out and therefore decrease pollutant concentrations in a kitchen. However, we may have overestimated the difference because most households using coal did not have range hoods. We removed the data for the coal-using households and assessed the PAH and NPAH removal efficiencies for when LPG and electricity were used. The mean PAH and NPAH removal rates for range hoods when LPG and electricity were used were only 30.3% and 21.6%, respectively. Range hoods usually have deodorization coefficients of not less than 90%, according to Chinese national standard GB/T 17713-2011. Because of the limitations of the economic levels in rural areas, the brand, service life. and effect of each household range hood were different. Residents may not regularly clean their range hood, which will decrease the exhaust efficiency of the range hood after years of use.

The particulate and gaseous phase PAH and NPAH distributions under different ventilation conditions are shown in Fig. 4b. During cooking periods, the particulate phase PAHs in kitchens with and without range hoods contributed 13.6% and 26.7%, respectively, of the total PAH concentrations and the particulate phase NPAHs in kitchens with and without range hoods contributed 24.7% and 41.6%, respectively, of the total NPAH concentrations. This indicates that using a range hood in an indoor kitchen can remove particulate phase PAHs and NPAHs. Chen et al. (2018) found that range hoods can remove PM_{2.5}. We therefore speculated that compared with the gaseous phase PAHs and NPAHs, the particulate phases were discharged into the outdoors because of adhesion to the particulate matter.

Many factors can affect PAH and NPAH production during cooking. Such as the size of the kitchen may affect air dilution,

the indoor temperature may affect PAH and NPAH gas/ particle partitioning and buoyancy of particles. Besides, differences in the cooking habits of sampling families (such as cooking method, cooking food, cooking time, etc.) may also lead to differences in the concentration of pollutants in the indoor kitchens. Although some of these factors were also considered during sampling (Table S1), we mainly assessed the effects of the cooking fuel and ventilation condition that had a greater impact after comparison in this study. Other factors need to be investigated in future research.

Assessment of carcinogenic risks

The mean TEQ_{BaP}s for the PAHs and NPAHs in the particulate phase, gaseous phase, and total air in the indoor kitchen air samples collected while cooking one meal are shown in Table 2 for all of the sampling points (n = 35). The mean TEQ_{BaP}s for the PAHs in the total air, gaseous phase, and particulate phase were 629.9 ± 1220.3, 150.3 ± 164.6, and 479.7 ± 1081.5 ng/m³, respectively. The TEQ_{BaP}s for the PAHs in the particulate phase were ~ 3.2 times higher than the TEQ_{BaP}s for the PAHs in the gaseous phase. The HM-PAHs (the most toxic PAHs) contributed relatively high proportions of the TEQ_{BaP}s for the particulate phase. In particular, BaP and dibenz[a,h]anthracene (DBahA) contributed 22.7% and 69.0%, respectively, of the TEQ_{BaP}s for the particulate phase. The total NPAH TEQ_{BaP} was 1021.9 \pm 3896.4 ng/m³, which was 1.6 times higher than the total PAH TEQ_{BaP}. This would mainly have been caused by the TEQ_{BaP}s for most PAHs being markedly lower than the TEQ_{BaP} for 6nitrochrysene (6-NChr). The mean TEQ_{BaP}s for the gaseous phase and particulate phase NPAHs were 840.4 \pm 1656.5 and 181.6 \pm 2533.1 ng/m³, respectively. Unlike for the PAHs, the gaseous phase TEQ_{BaP} for the NPAHs was 4.6 times higher than the TEQ_{BaP} for the particulate phase. The TEQ_{BaP}s for only three NPAHs were calculated in this study. More research should be performed to identify unknown NPAHs and determine their TEFs.

The TEQ_{BaP}s under different conditions were also determined, and the results are shown in Table S8. When cooking with coal, LPG, and electricity, the total TEQ_{BaP}s for PAHs were 1823.3 \pm 2185.6, 264.0 \pm 342.3, and 311.6 \pm 177.5 ng/ m³, respectively, and the total TEQ_{BaP}s for NPAHs were 2760.9 \pm 3157.4, 501.6 \pm 586.1, and 521.1 \pm 476.2 ng/m³, respectively. It has been found that a person using solid fuel throughout their life will be at least four times more likely to develop lung cancer than a person using clean energy (Jeong et al. 2019). Coal is often used for cooking and for heating in winter in rural areas in North China (e.g., in Hebei Province and Shanxi Province) (Hebei Provincial Bureau of Statistics

Table 2 Mean benzo[a]pyrene toxic equivalent (TEQ_{BaP}) concentrations (ng/m^3) for polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs (NPAHs) in the indoor kitchens (n = 35)

Group	Compound	Gaseous phase	Particulate phase	Total
РАН	Nap	ND	ND	ND
	Acl	0.1 ± 0.2	ND	0.1 ± 0.2
	Flr	ND	ND	ND
	Act	0.4 ± 0.4	ND	0.4 ± 0.4
	Phe	0.7 ± 0.8	ND	0.7 ± 0.9
	Ant	5.5 ± 7.2	0.6 ± 1.1	6.1 ± 8.2
	Flu	0.2 ± 0.4	0.1 ± 0.2	0.3 ± 0.6
	Pyr	0.6 ± 1.0	0.1 ± 0.1	0.7 ± 1.1
	Chr	1.6 ± 2.4	0.8 ± 1.8	2.4 ± 4.2
	BaA	14.3 ± 23.0	6.7 ± 17.4	21.0 ± 39.8
	BbF	6.7 ± 6.4	16.5 ± 38.1	23.2 ± 43.0
	BkF	7.4 ± 7.7	10.5 ± 21.1	17.9 ± 28.1
	BaP	43.1 ± 61.8	108.8 ± 216.6	151.8 ± 271.6
	DBahA	67.9 ± 87.5	330.8 ± 817.1	398.7 ± 863.6
	IP	0.2 ± 0.4	0.4 ± 0.9	0.5 ± 1.2
	BghiP	1.5 ± 2.6	4.5 ± 10.1	6.0 ± 11.5
	Total	150.3 ± 164.6	479.7 ± 1081.5	629.9 ± 1220.3
NPAH	5-NAcp	1.4 ± 3.4	0.4 ± 1.0	1.7 ± 4.2
	1-Npyr	15.1 ± 31.3	2.6 ± 4.5	17.7 ± 35.2
	6-NChr	823.9 ± 1392.7	178.7 ± 411.8	1002.5 ± 1753.8
	Total	840.4 ± 1656.5	181.6 ± 2533.1	1021.9 ± 3896.4

ND not detected or $< 0.1 \text{ ng/m}^3$

2018; Shanxi Provincial Bureau of Statistics 2018). To prevent loss of heat, doors and windows are closed in winter. This will cause high concentrations of pollutants to remain within the house and increase the risk of lung cancer in the person cooking (Zhu and Wang 2003). It is therefore important to encourage the use of clean energy (e.g., electricity) in rural areas to decrease the carcinogenic risks posed by PAHs and NPAHs released during cooking using other fuels.

During the cooking periods, the total PAH and NPAH TEQ_{BaPs} for the kitchens with range hoods were 244.4 \pm 125.3 and 472.6 \pm 537.9 ng/m³, respectively, which were lower than the TEQ_{BaP}s of 784.2 \pm 1420.3 and 1241.7 \pm 2063.6 ng/m³, respectively, for the kitchens without range hoods. In previous studies it was found that using a range hood can decrease the carcinogenic risks posed by PAHs during cooking (Zhang et al. 2017). Exposure only occurs during cooking periods, but Chinese people tend to cook between two and four times each day. Kamal et al. (2011) found that the exposure time strongly affects the PAH body burden. Zhang and Tao (2009) found that 1.6% of lung cancer cases in China are caused by inhaling PAHs in ambient air. Cooking workers will be exposed to high PAH and NPAH concentrations for long periods. This could be one reason for Chinese women have a high lung cancer incidence rate. Based on these findings, there should be a trade-off in the choice of cooking habits in rural households: Using a range hood or opening windows to allow ventilation during cooking periods could decrease the concentrations of contaminants in an indoor kitchen.

Conclusion

In this study, 35 rural households in Beijing and Shanxi were randomly selected from North China. The concentrations of 16 PAHs and 9 NPAHs produced during cooking periods in indoor kitchens and the outdoor air (background) were tested. High PAH and NPAH concentrations were found in the kitchen air during cooking periods. The cooking fuel and ventilation were the main factors affecting the PAH and NPAH concentrations in the indoor kitchen air. Higher PAH and NPAH, particularly MM-PAH, HM-PAH, and LM-NPAH, concentrations were found when coal was used than when LPG and electricity were used. The mean total TEQ_{BaP}s were also higher when coal was used than when LPG and electricity were used. The mean PAH and NPAH concentrations were 65.0% and 73.2% lower, respectively, for kitchens in which range hoods were used than for kitchens without range hoods, and the TEQ_{BaP}s followed the same trend.

Actual PAH and NPAH concentrations in indoor kitchen air during cooking periods in rural areas in North China were determined, and the results will be very useful for assessing the carcinogenic risks posed by PAHs and NPAHs to people cooking. It is noticed that this study has some limitations: collecting the samples at home while cooking one meal, and other factors that may affect cooking pollution in indoor kitchens are not discussed: such as kitchen size, outdoor air measurements, and cooking habits. Meanwhile, due to the limitation of sampling conditions, there are some unmeasured or uncontrolled conditions in this study, including indoor humidity and sampling artifacts. Some of these conditions may affect the dilution and change of indoor kitchen pollutants. These factors need to be paid attention to in the quantitative results. Large-scale and long-term field tests are required in the future to investigate indoor kitchen pollution and the exposure of cooking workers to PAHs and NPAHs under multifactor variables.

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Data availability All data generated or analyzed during this study are included in this published article (and its supplementary material)

Compliance with ethical standards

Competing interests The authors declare that they have no competing interests.

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