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Monitoring of Polycyclic Aromatic Hydrocarbons in a Rural Area of Jalgaon District, India

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Abstract

The present Study was undertaken to estimate concentrations of polycyclic aromatic hydrocarbons (PAHs) in indoor air from cooking fuels used in home in the rural area of Jalgaon, India. The three major types of cooking fuels used were gas, agrowaste, and wood. A total of 60 rural homes were selected for monitoring and we observed that particle-bound concentrations of PAHs were highest 1222.7μg/m^3 from the combustion of wood. The lowest concentrations of PAHs 14.51μg/m^3 were from when gas was used as the fuel. These results show that PAHs emitted from cooking sources are an important source of indoor air pollution.

Keywords: Cooking fuels, Polycyclic Aromatic Hydrocarbons, Indoor Air Pollution.

Introduction

Polycyclic aromatic hydrocarbons are one of the first identified airborne carcinogenic pollutants and are by-products of incomplete combustion of domestic fossil-fuel burning [1]. A study conducted by Conde *et al.* [2] showed that during biomass combustion the lower molecular weight PAHs, those with 2 to 4 rings, composed 80-89% of the emissions. PAHs adsorb onto dust particles in air and enter the body through the lungs during respiration. PAHs generally occur as complex mixtures (e.g., soot) and not as a single compound. Gevao *et al.* [3] in non-dietary PAHs intake assessment study in Kuwait observed about 16 PAHs in dust and indoor air collected on PUF glass fiber filter. High levels of naphthalene, acenaphthylene, phenanthrene, fluoranthene, and pyrene were detected whereas higher molecular weight PAHs such as benz[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, and dibenz[*a,h*]anthracene are released in low concentration.

PAHs are hydrophobic and relatively insoluble in water. Alcanzare [4] showed that 84 compounds composed of oxygenated PAHs, alkylated PAHs, cyclopenta-fused PAHs and benzologues of pyrene, fluoranthene, and perylene are emitted during wood combustion. Environmental Health Criteria 229, [5] has reported about 65 two-ring to six-ring nitro-, and nitro-oxy derivatives of PAHs during wood combustion. PAHs are likely to have additive and synergistic effects with their substituted species and with other pollutants (U.S. Department of Health and Human Services. 1995) [6]. Toxicological investigations showed different carcinogenic potency for various PAH species and different routes of exposure (oral, dermal, and inhalative) that cannot be predicted quantitatively [7].

The purpose of this study was to determine the exposure of PAHs using different fuels for cooking activity among rural households in the Jalgaon district.

Material and Methods

Study Area

Jalgaon district is located in the north-west region of the state of Maharashtra, India. It is situated between 20^{o} and 21^{o} latitude and 74.55^{o} to 76.28^{o} longitude. The district has a total area of 11,700 square kilometers and 4 million people. Around 72% of the total population of the district resides in rural areas [8]. The representative PAHs monitoring was conducted in 60 rural houses. This includes 20 houses each for gas, agrowaste, and wood users in the rural area. The agrowaste fuels included mainly plant materials, crop residue, grass, trash, twigs, and fallen leaves.

PAHs Monitoring

Monitoring of PAHs was carried out in the selected homes during cooking from November 17th, 2007 to July 2nd, 2009. The sampling was conducted while food was being cooked and ranged from 3 to 4 hours. As there was no major change in the type of fuel used, the indoor air quality monitored in the homes represents the state of the homes over several years. The fuels used for cooking in rural houses were gas, agrowaste, and wood.

Household dust exposures were measured using handheld dust exposure sampler (AS-2, Technovation Analytical Instruments Pvt. Ltd., India) for 3 to 4 hours. The dust (PM_{10}) was collected by filtration of air through a glass fiber filter (Whatman GF/A Grade, 25mm diameter). After sampling, each glass fiber filter was wrapped in aluminum foil then transfer to a dark, black colored bottle and kept at below 10°C for PAH extraction and analysis in the laboratory. PAHs were extracted from the glass fiber filters by Soxhlet extraction and analyzed by gas chromatography.

PAH Analysis

Sample preparation: The PAHs from each glass fiber filter were extracted in acetonitrile by Soxhlet extraction. After weighing each filter, it was cut into small pieces onto glass slides to provide maximum surface area and to facilitate efficient extraction of PAHs. These pieces were transferred into a Soxhlet apparatus using clean forceps. Ten-ml of GC/HPLC grade acetonitrile was then added to the round bottom flask of the Soxhlet assembly. The Soxhlet assembly was kept on ultrasonic bath. PAHs were extracted at a temperature of 60 to 80°C for about 30min. Each extract was filtered through a 0.45-µm membrane filter and stored at 4ºC. These extracts were analyzed on Gas Chromatograph within 24 hours from the sampling [9, 3, and 4].

Conditioning of columns: The capillary column was conditioned for 2 hours before injecting the sample. The operating conditions were: oven temperature of 280ºC; injector temperature of 280ºC, and detector temperature of 300ºC. After conditioning, the solvent was run to clean the column and then the standard mixture was run, followed by samples, and chromatograms were recorded.

PAH analysis: A Chemito 8610 gas chromatograph with flame ionization detector was used for PAH analysis. Separation of PAHs was achieved using a capillary column BPX5 (length-30mm ID-0.53mm film-0.5µm). Nitrogen was used as carrier gas at a constant flow rate of 5mL/min. The oven was programmed at 70ºC for 1 min ramped at 8ºC/min to 140ºC, held for 1 min, and further ramped at 5ºC/min to 290ºC and held for 15 min. The injector temperature was set at 260ºC and the detector temperature was set at 300ºC. The typical chromatogram for the standard was obtained and then sample peaks were confirmed by spiking the sample with known standards. Sigma-Aldrich reference standards were used for calibration of the gas chromatograph.

Concentrations of the 16 PAH species viz. naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, indeno[*1,2,3-cd*]pyrene, dibenzo[*a,h*]anthracene, benzo[*g,h,i*]perylene were determined. The total PAH concentration was estimated as the sum of the concentrations of these 16 PAH compounds for each sample. To assess the homolog distribution for each compounds in such samples Li *et al.* [10] classified these 16 PAH into three categories viz. low molecular weights (LM-PAHs, containing two to three-ringed PAHs), middle molecular weights (MM-PAHs, containing four-ringed PAHs), and high molecular weights (HM-PAHs, containing five to seven-ringed PAHs).

B[*a*]P$_{\textbf{equivalent}}$: In principle, the carcinogenic potency of a given PAH compound can be assessed on the basis of its benzo[*a*]pyrene equivalent concentration (B[*a*]P$_{eq}$). Calculation of the B[*a*]P$_{equivalent}$ concentration for a given PAH compound requires the use of its toxic equivalent factor (TEF), which represents the relative carcinogenic potency of the given PAH compound, using benzo[*a*]pyrene as a reference compound to adjust its original concentration [11].

Results

Table 1 shows the distribution of the 16 priority PAH species in the indoor environment of the houses under study. Higher levels of PAH compounds were observed in the indoor air of the houses using wood and agrowaste as a cooking fuel. The total PAH concentration in the wood-using houses was 1222.7µg/m^3, and the LM-PAHs were higher in concentration compared to MM-PAHs and HM-PAHs. In the houses using agrowaste for fuel, the total PAH concentration was 971.6µg/m^3, and LM-PAHs and HM-PAHs had higher levels compared to MM-PAH. In both wood and agrowaste using house categories, LM and HM-PAH concentrations were high compared to MM-PAHs. In the indoor air of houses using gas for fuel, the total PAH concentration was 14.51µg/m^3. Comparatively low levels of LM-PAH, MM-PAH and HM-PAH were observed in gas-using houses. In the indoor air of houses using gas for fuel, most of the species of PAH were below the detection limit.

Table 1: PAH concentrations (µg/m^3) in the indoor air samples of gas, agrowaste, and wood for fuel.

Compounds Name	Gas (N=20)	Agrowaste (N=20)	Wood (N=20)
Naphthalene	0.19 ± 0.11 (BDL – 0.39)	287.2 ± 211.3 (76.4 – 672.4)	292.9 ± 42.1 (12.5 – 440)
Acenaphthylene	1.32 ± 1.01 (0.4 - 1.5)	0.2 ± 0.1 (BDL – 0.4)	180.3 ± 69.2 (BDL – 182)
Acenaphthene	0.8 ± 0.5 (BDL – 1.4)	73.7 ± 65.8 (9.71 – 143.3)	82.6 ± 63.8 (7.5 – 255.7)
Flourene	6.4 ± 5.8 (0.5 – 12.3)	41.4 ± 38.3 (12.6 – 70.2)	51.4 ± 36.7 (25.4 – 77.4)
Phenanthrene	BDL	24.7 ± 18.3 (BDL – 28.3)	19.2 ± 13.4 (3.88 – 44.8)
Anthracene	BDL	39.5 ± 7.98 (33.8 – 45.2)	80.1 ± 52.1 (7.7 – 211.2)
Pyrene	0.9 ±0.4 (BDL – 0.9)	0.3 ± 0.1 (BDL – 0.5)	35.1 ± 23.4 (BDL – 38.9)
Fluoranthene	BDL	20.6 ± 12.0 (BDL – 24.8)	49.2 ± 23.7 (8.9 – 89.1)
Benz[*a*]anthracene	BDL	0.2 ± 0.1 (BDL – 0.4)	16.7 ± 11.2 (BDL – 19.3)
Chrysene	BDL	68.4 ± 45.2 (BDL – 73.2)	17.1 ± 8.4 (16.1- 19.6)
Benzo[*b*]fluoranthene	4.2 ± 2.5 (BDL – 4.2)	88.2 ± 69.3 (BDL – 95.9)	21.1 ±7.5 (BDL – 28.3)
Benzo[*k*]fluoranthene	BDL	65.8 ± 20.14 (54.4- 95.6)	176.2 ± 84.3 (68.1 – 384.6)

Benzo[*a*]pyrene	0.3 ± 0.1 (BDL - 0.3)	19.5 ± 10.3 (BDL - 28.4)	7.13 ± 3.3 (3.31 – 9.15)
Indeno[*1,2,3-cd*]pyrene	BDL	184.5 ± 137.6 (BDL – 213.6)	68.8 ± 34.2 (BDL – 72.4)
Dibenzo[*a,h*]anthracene	0.4 ± 0.2 (BDL – 0.4)	24.8 ± 24.9 (2.5 – 51.7)	13.2 ± 4.7 (BDL – 15.3)
Benzo[*g,h,i*]perylene	BDL	32.6 ± 21.7 (BDL – 59)	111.7 ± 86.3 (BDL – 123.6)
LM-PAH	8.71 (0.4-12.3)	466.7 (9.71-143.3)	706.5 (3.88-440)
MM-PAH	0.9 (0.3-0.9)	89.5 (0.4-73.2)	118.1 (8.9-89.1)
HM-PAH	4.9 (0.3-4.2)	415.4 (2.5-213.6)	398.13 (3.31-384.6)
Total PAHs	14.51	971.6	1222.7

Note: The values are mean ± SD of the total number of samples. A value in parentheses shows the range of the parameter. BDL: Below Detection Limit.

Table 2 shows the B[*a*]P$_{equiv}$ concentration of the indoor air in houses using different fuels. In the wood- and agrowaste-using category, the B[*a*]P$_{equiv}$ concentration of total PAH was 51.41ng/m^3 and 80.02ng/m^3, respectively. The B[*a*]P$_{equiv}$ concentration of HM-PAHs was higher compared to MM-PAHs and LM-PAHs. In the gas-using category, the B[*a*]P$_{equiv}$ concentrations of total PAH was 1.131ng/m^3 with higher levels of HM-PAHs (1.12ng/m^3).

Table 2: B[*a*]P$_{equiv}$ concentrations (ng/m^3) in indoor air samples of gas, agrowaste, and wood for fuel.

Compounds Name	TEF^a	Gas (N=20)	Agrowaste (N=20)	Wood (N=20)
		B[*a*]P$_{equiv}$ conc.	B[*a*]P$_{equiv}$ conc.	B[*a*]P$_{equiv}$ conc.
Naphthalene	0.001	0.00019	0.2872	0.2929
Acenaphthylene	0.001	0.00132	0.0002	0.1803
Acenaphthene	0.001	0.0008	0.0737	0.0826
Flourene	0.001	0.0064	0.0414	0.0514
Phenanthrene	0.001	BDL	0.0247	0.0192
Anthracene	0.010	BDL	0.395	0.801
Pyrene	0.001	0.0009	0.0003	0.0351
Fluoranthene	0.001	BDL	0.0206	0.0492
Benz[*a*]anthracene	0.100	BDL	0.02	1.67
Chrysene	0.010	BDL	0.684	0.171

Benzo[*b*]fluoranthene	0.100	0.42	8.82	2.11
Benzo[*k*]fluoranthene	0.100	BDL	6.58	17.62
Benzo[*a*]pyrene	1.000	0.3	19.5	7.13
Indeno[*1,2,3-cd*]pyrene	0.100	BDL	18.45	6.88
Dibenzo[*a,h*]anthracene	1.000	0.4	24.8	13.2
Benzo[*g,h,i*]perylene	0.010	BDL	0.326	1.117
LM-PAH		0.00871	0.8222	1.4274
MM-PAH		0.0009	0.7249	1.9253
HM-PAH		1.12	78.476	48.057
Total PAHs		1.131	80.02	51.41

TEF^{a}: Toxicity Equivalence Factor (Nisbet and LaGoy 1992).
BDL: Below Detection Limit.

Discussion

PAH compounds pose the most significant risk to human health in the emissions of residential wood combustion [12]. The study conducted by Ramdahl [13] show that the total concentration of the 16 priority PAHs was 3,000µg/m^3, and the concentrations of 60µg/m^3 for benzo[*a*]pyrene, a known carcinogen, have been measured in flue emissions from small residential stoves. Siegmann and Sattler [14] observed lower PAH concentrations during cooking between 1.08-22.8µg/m^3, using gas for fuel. Similar low concentrations of total PAHs were observed in the houses using gas for fuel in the present study. No studies were located regarding human death following inhalation exposure to any of the 16 PAHs (U.S. Department of Health and Human Services) [6]. Thus, it is impossible to evaluate the contribution of any individual PAH to the total carcinogenicity in humans because of the complexity of the mixtures and the presence of multiple carcinogens.

The higher molecular weight PAHs with 5 or 6 rings cause the most severe carcinogenic effects [2]. The higher molecular weight PAHs associated with dust prevailing during cooking period was observed in rural homes in North India [15]. In the present study, the lower, middle, and higher molecular weights PAHs are reported in the indoor air environment of rural houses. Gevao et al. [3] reported similar PAHs in the indoor air in Kuwait.

The emissions in the consecutive monitoring sessions during the process of cooking in the kitchen are inherently variable. It is difficult, lengthy, and costly to conduct the toxicological studies needed to quantify the disease burden in homes due to the complexity of indoor air pollution.

Conclusion

The results of this study indicate that fuel sources contribute to the particle-bound PAH concentrations in indoor air. The average higher concentration of PAHs in indoor air was observed in the wood as compared to agrowaste and gas fuel users. The

B[*a*]P_{equiv} concentrations were highest in agrowaste, followed by wood and gas fuel users. Therefore, cooking with biomass fuel contributes considerably to PAHs emissions in indoor environment of rural area of Jalgaon district.

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