

# EXPOSURE RISK ASSESSMENT OF POLYCYCLIC AROMATIC HYDROCARBONS ASSOCIATED WITH THE INDOOR AIR PARTICULATE MATTERS IN SELECTED INDOOR ENVIRONMENTS IN KANDY, SRI LANKA

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**Abstract:** The present work investigates the health risks associated with the inhalation exposure of particulate bound polyaromatic hydrocarbons in selected indoor environment in Kandy city. Particulate samples from five different types of indoor environments were collected for chemical analysis of 16PAH priority pollutants listed by USEPA. Categorization of locations was based on the degree of urbanization, type of fuel used for cooking and proximity to road from the location. Key meteorological parameters such as wind speed, wind direction, ambient temperature and relative humidity were also measured. The collected samples were analysed using a high performance liquid chromatograph (HPLC), after the necessary extraction and cleaning up of the sample. The total concentrations of particulate PAHs are in the range of 0.386 ng/m<sup>3</sup> to 14.65 ng/m<sup>3</sup>. The comparison of PAH levels at different categories of environment indicate that the degree of urbanization, fuel type and proximity to road influences the total concentrations of particulate PAHs. The dominant particulates of PAHs measured at the selected environments are naphthalene, acenaphthylene, acenaphthene. Then benzo[a]pyrene [B(a)P] indicating particulate PAHs are contributed by a mixture of both diesel and petrol engine type of vehicles, and biomass combustion. The total BaP<sub>eq</sub> concentrations at different environmental categories ranged from 0.06 to 3.08 ng/m<sup>3</sup>. The total BaP equivalency results showed the potential health risk to cancer due to inhalation exposure is of concern for residents living in high urban area with usage of wood as fuel. Since the total BaP<sub>eq</sub> concentrations for this category was higher. Very close to, or slightly exceeded the maximum permissible risk level of 1 ng/m<sup>3</sup> of benzo(a)pyrene in other categories also.

**Keywords:** Polycyclic Aromatic Hydrocarbons, Indoor air quality, Carcinogenic risk

## 1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of widespread environmental pollutants containing two or more fused benzene rings. The environmental occurrence of PAHs has been associated with adverse effects on public health, as they are considered to be the largest group of carcinogens present in the environment (Zhu, et al., 2008). These compounds are typically formed during the incomplete combustion of organic materials such as coal, oil, gas, and wood. The natural sources of PAHs include volcanic activity and forest fires. The lighter PAH (2–3 rings), which are generally not carcinogenic, are mostly found in the gas phase, while the heavier ones are mainly associated with airborne particles such as soot. Indoor emission sources of PAHs include smoking, cooking, and heating. Significantly, 50–75% of the particulate matter containing PAHs generated due to combustion of various cooking fuels are in respirable size range.

Hill capital of Sri Lanka, Kandy, is a valley surrounded by hills and air pollution due to vehicular emission in this city is becoming a major health concern. Further, a considerable proportion of houses use firewood as domestic fuel source, a situation which can lead to increased levels of air pollutants such as PAHs in these indoor environments.

The present study aims at quantifying PAHs levels associated with particulate matter in selected indoor environments in Kandy city and to investigate the influence of firewood burning and vehicular emission on PAHs levels. Further, the carcinogenic risks associated with the exposure to detected levels at different categories of indoor environments are estimated.

## 2 Methodology

### 2.1 Sampling sites

Samples were collected from 14 sites in and around Kandy city which included 12 houses and two office buildings. The 12 houses were selected according to three main criteria; degree of urbanization, type of domestic fuel that they use for cooking, and proximity to roads. Other two sites were selected from offices around the Kandy city. The locations are shown in Figure 1. Categorization of the sampling sites, as shown in Table 1, was based on the degree of urbanization, influence of different types of domestic fuels, and proximity to roads.

Table 1 Description of different categories of.

Category type	Description	Sites included
1	High urban area + Gas users	L1, L3, L9, L11
2	Less urban area + Gas users	L2, L6, L8,
3	High urban area + wood uses	L4, L7, L10,
4	Less urban area+ wood uses	L5, L12
5	Office premises	L13, L14

#### Site Selection

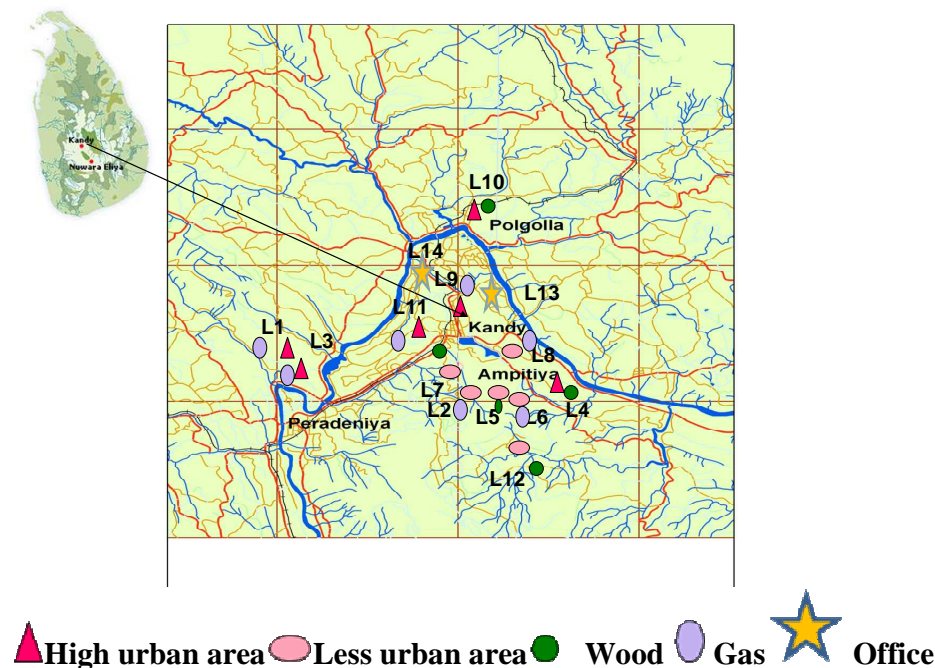


Figure.1 : *Locations of PAHs sampling points*

## 2.2 Sample Collection

Particulate matters in selected indoor air were collected onto 55 mm diameter glass micro fibre filters using personal air samplers. Sampling was done for 24 hrs in the selected houses and in offices only 8 hours because offices are mainly occupied during these hours. In case of office buildings, two samplers were used to collect a reasonable volume of air. The personal air samplers were chosen for this study because they were portable, less noisy, battery-operated and easy to mount inside the buildings. The flow rates were checked regularly to obtain the total flow rate throughout the sampling period. Air sampling was conducted from May 2009 to August 2009 under identical weather conditions. Indoor air samples were collected in the kitchen or in close proximity to the kitchen. This sampling location was selected because, most homes in Kandy, the kitchen has more or is closer to potential PAH sources than other rooms and is often where most family activities occur.

## 2.3 Sample preparation and analysis

The filter with collected particulate matters was extracted for 7 hours in a 250 ml soxhlet apparatus with dichloromethane. The extract was concentrated by solvent evaporation under reduced pressure in a rotary evaporator (Yamato scientific co.ltd, RE-46). The temperature of the water bath was set at 40 °C. The remaining residues in the evaporated flask were dissolved in 1 ml of acetonitrile as required by the subsequent analytical process.

Identification and quantification of PAHs in the samples were carried out using a HPLC equipped with a ZORBAX Eclipse XDB- C8 column, (250 mm× 4.6 mm× 5 µm), under gradient elution of deionised water and acetonitrile at a flow rate of 0.8 ml/min. Calibration data developed by Wickramasinghe et.al (2008), was used. The readings were corrected to observed recovery efficiencies of the standard samples analysed.

## 2.4 Hazard and Risk assessment

The Toxic Equivalent Factor (TEF) approach has been extensively used in risk assessment of different classes of PAH mixtures. The overall toxicity of a PAH mixture is defined by the concentration of individual PAH compounds ( $C_i$ ) in a mixture times their relative TEF.  $BaP_{eq} = (C_i) TEF$ . The TEF is an estimate of the relative toxicity compared to BaP for which a great deal of data exists. For this study the TEF values proposed by Nisbet and LaGoy (1992) were used. The  $BaP_{eq}$  values can be subsequently used for cancer risk assessment.

## 3 Results and discussion

The observed PAH concentrations are given in Table 2. Accordingly, the total concentration of PAH measured in this study ranged from 0.38 to 14.65 ng/m<sup>3</sup>. Highest concentration of total PAHs was present in location 13 which is located in high urban area. Lowest total PAHs was present in location 6 located in less urban area. Naphthalene (Nap) in this study was found to be the most abundant of the 16 PAHs studied. This pattern is observed in most of the reported PAHs levels in literature too. The main reason for

this could be due to the fact that Nap dominates in most of the PAH emissions ( Zhu, et al., 2008, Li et al., 2005 , Kalaiarasan, et al., 2009, Mumford , et al., 1991). The Nap concentration varied from 0.10 to 7.39 ng/m<sup>3</sup> with an average of 2.813 ng/ m<sup>3</sup> and accounted for 43.49 % of the total PAHs. In addition, a previous study found that Nap emissions in indoors environments were highly correlated with the indoor activities such as the use of mothballs in china (Zhu, et al., 2008).

Table .2: PAH concentrations at different sampling locations

Compound	Concentrations of PAHs at individual sampling locations (ng/m <sup>3</sup> )													
	L1	L2	L3	L4	L5	L6	L7	L8	L9	L10	L11	L12	L13	L14
Nap	2.83	2.13	1.31	4.69	0.22	0.1	0.88	0.48	7.39	4.12	0.97	3.14	4.15	6.98
Ace	-	-	-	3.29	-	-	2.28	0.18	4.05	1.17	2.58	1.22	2.05	-
Acy	-	-	-	1.78	0.02	0.11	-	0.37	1.51	0.46	1.07	1.06	6.03	0.04
Flu	-	-	0.01	-	-	-	-	0.01	0.13	0.26	0.18	0.28	0.17	-
Phe	-	-	-	0.06	-	-	-	0.002	0.1	0.06	0.04	0.03	0.04	0.03
Ant	0.37	-	-	0.01	0.19	0.08	0.23	-	0.19	0.23	0.34	0.18	0.02	-
Flt	-	-	-	0.34	-	-	-	-	0.13	0.45	0.51	0.38	0.1	-
Pyr	-	-	0.16	0.28	0.08	0.06	-	0.09	0.7	0.16	0.35	0.16	0.11	0.06
B(a)A	-	0.12	0.7	0.07	-	-	0.05	0.003	0.14	0.12	0.15	0.08	0.06	-
Chr	0.04	-	0.08	-	-	-	0.06	-	0.12	0.14	0.14	0.08	0.01	-
B(b)F	-	-	-	0.46	-	-	-	-	-	-	1.16	-	0.06	-
B(k)F	-	0.006	-	0	-	-	-	-	-	-	-	-	0.05	-
B(a)P	-	0.06	1.26	0.46	0.02	0.03	1.02	0.013	0.08	0.91	0.64	1.22	0.27	0.18
DBA	-	-	0.39	0.51	-	-	1.12	0.016	0.08	0.43	0.26	-	0.01	-
BPe	-	0.01	0.23	-	0.01	-	0.44	-	0.03	0.66	0.22	-	0.02	-
Ind	-	-	-	-	-	-	-	-	-	-	-	-	0.02	-
Total	3.24	2.326	4.14	11.95	0.54	0.38	6.08	1.164	14.65	9.17	8.61	7.86	13.17	7.29

**Nap**-naphthalene, **Ace**- acenaphthene, **Acy** -acenaphthylene, **Flu** -fluorene, **Phe** -phenanthrene, **Ant** –anthracene, **Flt** -fluoranthene, **Pyr** -pyrene, **B(a)A**- benz(a)anthracene, **Chr** -chrysene, **B(b)F**- benzo(b)fluoranthene, **B(k)F**- benzo(k)fluoranthene, **B(a)P**- benzo(a)pyrene, **DBA**-dibenzo(a,h)anthracene, **BPe**- (Benzo(g,h,i)perylene, **Ind**-ideno(1,2,3,-c,d)pyrene

According to this study, naphthalene concentration did not vary with the degree of urbanization because low concentrations of naphthalene was observed in some high urbanised locations, while in some less urban locations high concentrations of naphthalene were found. According to the results, the Nap concentration was not dependent upon the fuel type as it was found in domestic as well as office environments. Li, et al., 2005 reported that in Chicago Nap had the highest mean and median concentrations among the 16 PAHs. The medians are 177 and 168 ngm<sup>3</sup>, and the maxima are 2340 and 1867 ngm<sup>3</sup>, for indoor air and outdoor air, respectively. Compared with two other indoor PAH studies in the US conducted in 1986 and in 1994, indoor naphthalene concentrations found in this study are lower. They found that indoor naphthalene emission was largely associated with mothball usage, while suggesting that another combustion sources may also have been responsible for high naphthalene concentrations such as combustion of kerosene, and other fuel vapours.

Acenaphthalene (Ace) is the second highest PAH present at these 14 locations ranging 0.18 - 4.05 ng/ m<sup>3</sup>. Acenaphthene found in concentrations ranging from 0.02 to 6.03 ng/ m<sup>3</sup>. Then Benzo[a]pyrene (B[a]P) concentration ranging from 0.03 to 0.7 ng/ m<sup>3</sup>. All PAHs with molecular weight between 128 and 202 had average concentration 5.22ng/ m<sup>3</sup>, whereas PAHs with molecular weight 228–278 was found in much lower concentrations with an average of 1.03 ng/m<sup>3</sup>. All PAHs with molecular mass 152–202 had average concentrations higher than 1 ng/ m<sup>3</sup> and their medians ranged from 0.5 to 19 ng /m<sup>3</sup>. By comparison, PAHs with molecular mass 228 or higher exist in air in significantly lower concentrations. No median or average for these heavy PAHs is above 0.25 ng/ m<sup>3</sup> for indoor samples or 0.5 ng/ m<sup>3</sup> for outdoor samples.

As shown in Table 3, the quantitative results mean varied significantly depending on the degree of urbanization, and type of fuel used for cooking. Among five categories, as described above, the highest concentrations of total PAHs found are in houses that use wood as their cooking purposes and located in high urban area. In addition, those locations are close to road so vehicular emission may be the major source. This is due the fact that the indoor air quality in these houses is affected by both emissions from vehicles as well as from biomass combustion.

The second highest concentration was observed in office environments. Proximity to roads may be the major cause of this observed high PAHs concentration. In addition, a recent study reported the amount of dust released during operation of computers to be between 4.0 and 6.3 mg dust per day, and suggested that a significant potential for exposure to chemicals such as PAHs that are associated with re-suspended dust particles (Destailats, et al., 2008). Some samples analysed by them had significantly higher levels of B(b)F, Chr, Flu, and Phe than domestic dust. Since these compounds are present in outdoor air and can be emitted by various indoor sources, this study could not estimate the contribution of computer emissions, but it is suggested that a fraction of measured PAHs might have been emitted from the heated plastic materials, chips and other computer components.

Table :3 *Variation of the average individual PAH compound in different categories in (ng/m<sup>3</sup>).*

Compound	Category 1	Category 2	Category 3	Category 4	Category 5
Nap	3.125	0.900	4.400	1.410	5.560
Ace	1.650	0.060	2.230	1.160	1.025
Acy	0.640	0.160	1.120	0.360	3.035
Flu	0.080	0.003	0.130	0.090	0.080
Phe	0.030	0.001	0.060	0.010	0.035
Ant	0.220	0.020	0.120	0.200	0.010
Flt	0.160	-	0.390	0.120	0.050
Pyr	0.300	0.050	0.220	0.080	0.080
B(a)A	0.240	0.041	0.090	0.045	0.030
Chr	0.090	-	0.070	0.046	0.005

B(b)F	0.290	-	0.230	-	0.030
B(k)F	-	0.002	-	-	0.025
B(a)P	0.440	0.030	0.685	0.750	0.225
DBA	0.180	0.005	0.470	0.370	0.005
BPe	0.120	0.003	0.330	0.150	0.010
Ind	-	-	-	-	0.010
Total	7.415	1.274	10.560	4.820	10.215

Based on the values given in Table 3 and the TEF values proposed by Nisbet and Lagoy (1992), the calculated B(a)P<sub>eq</sub> values for the five categories of locations are given in Table 4. It is evident from Tables 3 and 4 that even though PAH concentration in category 4 is much lower compared to category 5, the total toxicity at category 4 is higher than at category 5. This is due to the emissions from biomass burning include more carcinogenic PAHs than in emissions from vehicles.

Table 4. TEF values for different categories of indoor environment

Category	B(a)P <sub>eq</sub> value
1	1.24
2	0.06
3	3.08
4	2.63
5	2.35

Increased risk of lung cancer associated with life time exposure to indoor particulate PAHs was calculated B(A)P at five categories according to EPA estimated potency factor of  $8.7 \times 10^{-5}$  for life time exposure to a mixture with  $1 \text{ ng/m}^3$  B(a)P. The estimated risk values are given in Table 5. As can be seen the risk value is higher than the acceptable level of 1 per 100000 in all categories except for category 2 which uses gas as fuel and located in less urban areas.

Table 5: Unit risk assessment

Category	Unit Risk value(100000)
1	6.3
2	0.3
3	15.6
4	13.4
5	12.0

## 4 Conclusion

This study reveals that the highest mean concentration of indoor particulate PAHs are in houses which use firewood as fuel and located in the highly urbanised areas, followed by office environments which are situated in high urban areas. Third high concentrations were at urban houses that uses gas and the least concentrations were observed in less urban houses that use gas as fuel.

The total B(a)P equivalency results showed the potential health risk to cancer is of concern for residents living in urban areas with wood use as their domestic fuel, gas uses and office environments. The total B(a)P<sub>eq</sub> concentrations in high urban houses with wood use as domestic fuel, in office environments, and in high urban areas with gas uses exceed the maximum permissible risk level of 1ng/m<sup>3</sup> of B(a)P. The carcinogenic risk is beyond the acceptable level in all categories other than the category which are located in less urban areas and use gas as fuel.

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