

Assessment of Indoor Exposure to Polycyclic Aromatic Hydrocarbons for Urban Poor Using Various Types of Cooking Fuels

C. V. Raiyani, ¹ J. P. Jani, ² N. M. Desai, ¹ S. H. Shah, ¹ P. G. Shah, ¹ and S. K. Kashyap ¹

¹National Institute of Occupational Health, Indian Council of Medical Research, Meghaninagar, Ahmedabad-380 016, India and ²Department of Pharmacology, School of Medicine, University of Pittsburgh, Pittsburgh, Pennsylvania 15261, USA

The use of traditional biomass like wood, cattle dung cake and bushes as a fuel for cooking and house heating purposes is the only source of energy especially for poor families of underdeveloped and developing countries including India, and will continue to meet the need for cooking energy in the coming decades. Though use of fossil fuels such as coal, kerosene and liquid petroleum gas (LPG) are not uncommon, the supply is limited and also restricted to urban centres only. Hence urban poor most of the rural population mainly depend on traditional biomass for their source of cooking energy. Most of the cooking using biomass is done on extremely simple devices like - three rocks; U-shaped hole in a block of clay, mud or bricks; and a pit in the ground. In India all such devices are named 'chulha'. The energy efficiency of such stoves (chulha) is very poor and emit large amounts of air pollutants in the form of smoke. The use of a stove named 'Signi' is a very common device for the fuels - coal and charcoal. Coal is generally fired in 'Sigri' keeping it in an open place like a courtyard, corridor or street until the cessation of visible smoke. Then it is shifted to the kitchen for cooking. Wick or pressure stoves for kerosene or similar type of fuels, and the stoves with two gas burners for LPG fuel are commonly used. Few studies have been carried out characterising levels of benzo(a) pyrene in particulate organic matters (POM) from such houses (Aggarwal et al. 1982; Smith et al. 1983; Patel et al. 1984; Dave 1984; Deshpandey et al. 1984). Indoor levels of priority polycyclic aromatic hydrocarbons (PAHs) in the houses using wood fuel in Kenya and Gambia have been reported by WHO (1987, 1988) but none of them has characterised the levels of PAHs in the indoor air of the houses using various type of cooking fuels. The National Institute of Occupational Health undertook a study to generate baseline data on indoor exposure to PAHs in single room homes using various types of cooking fuels. The study was initiated in 1987. Results of preliminary findings are discussed in this paper.

MATERIALS AND METHODS

At the onset of the programme, sixty households were randomly selected from an urban agglomeration at Ahmedabad, of which each group of 10 households were using either cattle dung, wood, admix of wood and cattle dung, coal, kerosene or LPG as a cooking fuel. Houses using

biomass fuels (cattle dung, wood and admix of both) were made of mudwall and had an iron roof. The houses using coal, kerosene or LPG were pukka (brick) houses. None of the houses had separate kitchens and cooking was carried out within the house all year. Airborne particulate samples were collected on glass microfibre filter paper (EPM-1000, Whatman, 50 mm dia) which were previously dried in an oven at 105°C for three hours, kept in desiccator for 24 hours, numbered, and preweighed on a micro balance accurate to 0.1 mg (Sartorius. Model MP 1608). Air was drawn at a suction rate of 15 lpm for the entire cooking period, which was normally 45 to 60 minutes (duration of one sitting). Samples were collected from the centre of the house and at the height of 80 cm (breathing height in squatting posture). Air suction was started 10 minutes after lighting the fuel in the domestic stove (i.e. chulha, kerosene stove or LPG stove) or shifting of a lighted 'Sigri' to the kitchen. The purpose of this time lapse was to allow build up of pollutants to the equilibrium level. At the end of the sampling period each dust laden filter paper was carefully detached, folded and packed in a PVC bag. The sample filter papers were kept in a desiccator at least for 24 hours before weighing. The difference in weight was evaluated for total suspended particulate (TSP) levels. The samples were kept in a dark and cool place till the analysis. The samples were extracted with spectroscopy grade benzene in a light protected soxhlet extractor for 8 hours. The extract was filtered through G-4 grade sintered glass funnel. One drop of keeper solution (ethylene glycol) was added to the extract and it was then concentrated to \sim 0.5 ml at 60°C using vacuum rotary evaporator. Isolation of the PAHs fraction from the benzene soluble concentrate was accomplished by preparative thin layer chromatography (TLC) and the resulting sample was analysed using HPLC as described by Jani et al (1991). External standard technique was used for quantitation of the samples. Reference standard mixture NBS-1647 (10 ul) was charged on TLC plate along with the samples and used as an external standard for the samples treated on the same plate. Individual PAHs were obtained from Sigma Chemicals, USA: PAH reference standard mixture NBS-1647 was obtained from National Bureau of Standards, USA.

RESULTS AND DISCUSSION

There was no marked difference in room volume of the study houses using different categories of fuels which ranged from 33 to 37 cu m. Family size was in the range of 4 to 8 persons/family.

Mean, median and range for individual PAHs and TSP found in the present study are given in Table 1. Mean levls of TSP in the houses using cattle dung (6.95 mg/cu m), wood (5.73 mg/cu m) or mixture of both (6.22 mg/cu m) were much higher than those in houses using fossil fuels - coal, kerosene and LPG which were 1.16, 0.74 and 0.61 mg/cu m respectively. Amongst the houses using biomass fuels, the levels of total PAHs were relatively much higher where cattle dung was used alone (3.56 µg/cu m) or in combination with wood (3.46 µg/cu m), compared to the levels in houses using only wood (2.01 µg/cu m). The levels of total PAHs in houses using coal, kerosene and LPG were 0.55, 0.25 and 0.13 µg/cu m respectively. It would be worthwhile to mention that outdoor level of TSP was 0.84 ± 0.15 mg/cu m. Hence

Table 1. Levels of PAHs (ng/cu m) during cooking hours in the houses using different type of cooking fuels.

Compound				Тур	Type of cooking fuel	ng fuel			
		Cattle dung	bur	Cat	Cattle dung + wood	poom .		Wood	
	Mean	Median	Range	Mean	Median	Range	Mean	Median	Range
1. Acenaphthylene	162	35	6-753	42	17	14-95	27	15	6-51
2. Acenaphthene	57	-	5-258	23	16	8-42	15	80	5-35
3. Fluorene	5.1	5	11-202	66	13	7-275	31	30	19-51
4. Phenanthrene	2 10	29	14-667	156	77	17-404	23	16	5-89
5. Anthracene	32	-	10-80	22	10	5-63	12	9	4- 76
6. Fluoranthene	267	122	16-750	317	136	20-1095	63	45	19-200
7. Pyrene	283	216	45-726	195	146	31-850	176	8	30-500
8. Benzo(a)anthracene	213	96	15-1021	284	197	34-820	178	158	22-473
9. Chrysene	353	148	17-1439	286	265	5 2-845	169	103	16-424
10. Benzo(e)pyrene	327	136	11-1106	267	105	5 1-805	171	150	17-376
11. Benzo(b)fluoranthene	219	93	16-903	209	153	29-477	146	115	18-389
12. Benzo(k)fluoranthene	211	142	14-824	169	117	16-313	84	72	22-202
13. Benzo(a)ovrene	462	214	39-1645	375	265	27-865	399	370	71-793
14. Dibenz(a.h)anthracene	221	154	17-958	477	212	47-840	5 10	305	35-862
15. Benzo(q,h,i)perylene	263	199	37-802	25.9	122	45-506	194	157	40-511
16. Indeno(123-cd)pyrene	155	110	18-670	195	202	64-3 18	121	96	12-317
Total PAHs (ug/cu m)	3.56	1.73	0.54-18.1	93.46	2.15	0.72-9.83	2.01	1.93	0.83-4.54
TSP (mg/cu m)	6.95	69.9	1.20-14.19	9 6.22	6.0 1	2.25-12.67	5.73	5.02	2.04-8.87

Table 1. (continued)

Compound				Тур	Type of cooking fule	ng fule			
		Coal			Kerosene	Эс		LPG	
	Mean	Median	Range	Mean	Median	Range	Mean	Median	Range
• •	20 4	17	5-24 2-6	15	15	14-16 1-3	2 2	~ -	1-3
 Fluorene Phenanthrene 	68 23	69 24	43-89 11-32	53 22	5.2 20	38-70 9-39	5 10	10	3-18 3-10
5. Anthracene 6. Fluoranthene	1 29	0.8 28	0.5-2 23-35	0.4 8	0.4	0.3-0.5	0.2	0.2	0.1-0.3
	120	130	65-153	57	55	18-99	21	19	7-37
	48 48	75 75	18-57	2 2	^y 5	5-15 8-31	, p	9 6	5-8 7-14
 Benzo(e)pyrene Benzo(b)fluoranthene 	17 26	12 22	8-45 15-50	14 8	10	5-39 4-11	ω α	7 6	5-13
	30	29	10-42	16	12	6-25	7	, _	4-11
	56	58	13-92	17	13	6-30	13		8-23
	80	92	15-111	18	-	9-44	13	13	8-19
15. Benza(g,h,i)perylene	32	52	10-80	9	7	5-30	6	8	4-15
	22	23	12-52	6	ω	7-15	ω	7	4-12
Total PAHs (µg/cu m) TSP (mg/cu m)	0.55	0.5 1	0.28-0.62	0.25	0.23	0.14-0.38 0.59-1.02	0.13	0.13	0.11-0.14

TSP levels in the houses using kerosene and LPG could be presumed to be mainly a contribution of the outdoor environment.

The highest mean levels of PAH compounds in different categories of houses were for benzo(a)pyrene in the houses using cattle dung (462 ng/cu m); dibenz(a,h)anthracene in the houses using admix of cattle dung and wood (477 ng/cu m) and wood alone (5.10 ng/cu m); and pyrene in the houses using coal (120 ng/cu m), kerosene (57 ng/cu m) and LPG (21 ng/cu m). Mean levels of benzo(a)pyrene in these houses were 462. 375, 399, 56, 17 and 13 ng/cu m respectively. Looking to intra-concentration profile of PAHs for each category of houses it could be, in general, observed that levels of high molecular weight hydrocarbons (MW ≥252, compound No. 10-16) were major components in the smoke of biomass fuels, whereas levels of low molecular weight hydrocarbons (MW ≤228, compound No. 1-9) were major components in the smoke of fossil fuels. On comparing the levels of individual PAHs with those observed in houses using LPG as an yard-stick, levels of PAHs in the houses using biomass fuels were higher by a factor of 5 to 45; by a factor of 2 to 10 in houses using coal, and by a factor of 1 to 7 in the houses using kerosene.

In assessment of human exposure and dose, better information can be gathered from 24 hours personal sampling studies carried out for a number of subjects and sampling stations (Meyer and Hartley, 1982). Neverthless, it is useful to attempt some rough estimation of total exposure and resultant dose in order to compare the relative hazards of different activities and locations.

Table 2. Airborne benzo(a)pyrene concentrations and dose estimates.

Type of fuel/ source	Concentration (ng/cu m)		Daily exposure te to ambient air	
		(hrs.)	(hrs.)	(ng)
Cattle dung	462	3	21	1033
Admix of cattle	375	3	21	870
dung and wood				
Wood	399	3	21	914
Coal	56	3	21	272
Kerosene	17	3	21	198
LPG	13	3	21	191
Outdoor air of study area	12.7	continuous		190
Cigarette smoker (Bridbord et al. 1		One pa perday	ck of cigarette	400
Proposed USSR ambient standard	1.0	conti		15

This has been calculated (Table 2) by estimating exposure durations and breathing rates for benzo(a)pyrene as an index pollutant with the assumption that the reported mean concentrations and exposures were representative and uniform throughout the exposure period for each group of subjects. The average cooking time per day was estimated to be 3 hours, meaning that the inhabitants were exposed to smoke generated from cooking fuels for that duration only, and inhaled air

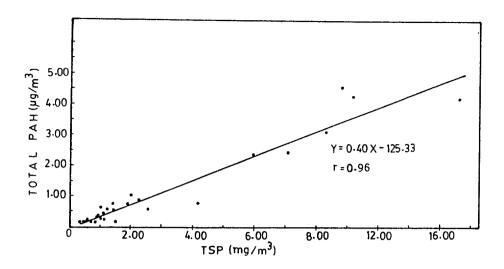


Figure 1. Correlationship between total suspended particulates (TSP) and total PAHs measured from the houses using various cooking fuels.

of general ambient quality for rest of the day. The inhalation volume was assumed to be 15 cu m of air per day. The results of these estimates showed that households using traditional biomass fuels were daily inhaling benzo(a)pyrene equivelent to smoking 2 to 2.5 packs of cigarettes. The exposed population is not generally composed only of adults of middle age and in good health. In general, the cooking women (whether young, old, pregnant, ill or infirm) and other family members like infants, children and the ill are exposed to such high levels of pollutants. Indeed relatively few workers in rather obscure occupations would receive the doses approaching the levels seen in this study.

The correlation of total PAHs with the TSP levels (irrespective of type of fuels) was highly significant (r=0.96, Fig.1). This shows that the indoor built up of PAHs depend on the amount of smoke generated in the use of different types of fuels. Hence, in efforts to solve the high indoor exposure problem associated with biomass fuel use, the greatest consideration needs to be given to adopt new types of improved biomass stoves (chulhas) with chimnies and in improving home ventilation. However, improved stoves with chimnies only help to divert the smoke emission from within the house to the general environment. Thus, although the indoor levels are reduced, the problem of the community environment and general ambient exposure levels remains unsolved. Biomass stoves designed to reduce smoke generation alongwith increased heating efficiency would ideally solve the problem. Alternatively, as charcoal produces less amount of smoke (Patel et al. 1984), supply of charcoal or charcoal pellets should be made available at competitive rates, and use of cattle dung and wood as a cooking fuel must be phased This would result in improvement of combustion characteristics with increased efficiency and flexibility by way of upgrading the traditional biomass fuel into high quality fuel, and reducing the air pollution exposures. With the present patterns of fuel use, it is certain that

the resultant exposures are not compatible with achieving a higher quality of life which is the overall goal of all development efforts.

Acknowledgements. Authors thank to Mr.KM Vyas, Mr.JA Shah and Mr.KR Dabhi for their technical help. Most importantly, we thank the women and their families who so graciously participated in the study. We are also thankful to Mr.DR Lonkar for his secretarial assistance.

REFERENCES

- Aggarwal AL, Raiyani CV, Patel PD, Shah PG, Chatterjee SK (1982) Assessment of exposure to benzo(a)pyrene in air for various population groups in Ahmedabad. Atm Environ 16:867-870.
- Bridbord K, Finklea JF, Wagoner JK, Moran JB, Caplan P (1976) Human exposure to polynuclear aromatic hydrocarbons. In: Frudenthal RI, Jones PW (eds). Carcinogenesis, vol.1; Polynuclear aromatic hydrocarbons: Chemistry, Metaboslism and carcinogenesis. Raven Press, New York, pp 319-324.
- Dave JM (1984) Studies on emissions from coal burning stoves (Sigries) as used in Eastern India. In: Berglund B, Lindvall T, Sundell J (eds). Proc 3rd International Conference on Indoor Air Quality and Climate, Stockholm, vol.4, pp 383-388.
- Deshpandey JM, Nayak UV, Sunita UN (1984) Indoor air quality in Bombay, India with respect to respirable particulates, benzo(a)pyrene and sulphates. In: Berglund B, Lindvall T, Sundell J (eds). Proc 3rd International Conference on Indoor Air Quality and Climate, Stockholm, vol.4, pp 403-408.
- Jani JP, Raiyani CV, Mistry JS, Patel JS, Desai NM, Kashyap SK (1991) Residues of organochlorine pesticides and polycyclic aromatic hydrocarbons in drinking water of Ahmedabad city, India. Bull Environ Contam Toxicol 47:381-385.
- Meyer S, Hartley H (1982) Inventory of current indoor air quality related research. USEPA, Research Triangle Park, North Carolina, EPA-600/57-81-119.
- Patel TS, Raiyani CV, Mohan Rao N, Aggarwal AL, Kartha GP, Chatterjee BB (1984) Indoor air pollution problem: Traditional vs Modern fuels. In: Berglund B, Lindvall T. Sundell J (eds). Proc 3rd International Conference on Indoor Air Quality and Climate, Stockholm, vol.4, pp 295-302.
- Smith KR, Aggarwal AL, Dave RM (1983) Air Pollution and rural biomass fuels in developing countries: A pilot village study in India and implications for research and policy. Atm Environ 17:2343-2362. WHO (1987) Global Environment Monitoring System: Human Exposure
- WHO (1987) Global Environment Monitoring System: Human Exposure Assessment Location Project, publication No.WHO/PEP/87.1. Indoor air pollution study, Maragua area, Kenya, World Health Organization, Geneva.
- WHO (1988) Global Environment Monitoring System: Human Exposure Assessment Location Project, publication No.WHO/PEP/88.3. Indoor Air Quality in the Basse area, The Gambia. World Health Organization, Geneva.

Received June 5, 1992; accepted October 16, 1992.