

TOXICITIES OF PM_{2.5} POLYAROMATICS HYDROCARBON ON INDOOR AIR EMITTED DURING FUMING OF INCENSE AND MOSQUITO COIL MATERIALS

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ABSTRACT

In this work, the concentration and fluxes of particulates polycyclic aromatic hydrocarbons emitted during fuming of incense (IS) and mosquito coil (MC) materials are described. The concentration of PM_{2.5} in the indoor environments during fuming of IS and MC (n = 10 each) materials was ranged from 1374-7645 and 260-677 μ g/m³ with mean value of 4054 ± 3233 and 392.70 ± 210 μ g/m³, respectively. Whereas, the concentration of PM₁₀ was ranged from 1465-8597 and 384-784 μ g/m³ with mean value of 4296 ± 3284 and 522.4 ± 209 μ g/m³, respectively. The mean PM_{2.5}/PM₁₀ ratio for IS and MC materials was found to be 0.94 ± 0.02 and 0.75±0.01, respectively. The concentration of PM_{2.5} and PM₁₀ was almost same; therefore the detailed investigation of the PM_{2.5} was carried out. The concentration of PAHs (i.e. Phe, Cor, Fla, Bgh, Baa, Cry, Bbf, Ant, Dba, Ind, Pyr, Bkf and Bap) in PM_{2.5} emitted during fuming of the IS and MC materials are described. The chemical composition of the PM_{2.5} is also discussed.

Key words: PM, PAH, IS, MC, Fuming, Chemical composition.

INTRODUCTION

Indoor air particulate pollution have more exposures arising from burning of materials like biomass, coal, incense (IS), mosquito coil (MC), etc. is several folds more dangerous than the outdoor air pollution^{1,2}. The IS materials i.e. sticks, agarbati, lobhan, kapoor, dhoop etc. are fumed for several purposes by people in India. The mosquito problem is sustained for the most of period of a year, and to repel the mosquito the coils are fumed in the home, office, kitchen, restaurant, library, even in the bathroom, washroom etc. Their incomplete combustions generate smokes, which are made of complex chemical harazadous particulate matter and are of rising environmental and health interests. The ash produced from these materials contains typical regulated elements such as As, Ba, B, Cd, Cr, Pb, Hg,

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Se, etc. When these elements released to the environment, there is very significant impact to the human, wildlife and ecosystem. The major active ingredients of MC are pyrethrins, which is toxic compound and accounting for about 0.3-0.4% of coil mass, on burning it evaporate along with smoke³. The smoke released are complex mixture of particulate matters (PM), silica, metals, carbonate carbon (CC), organic carbon (OC), black carbon (BC) etc. The above mentioned compound are of significant sources for PM, polycyclic aromatic hydrocarbons (PAHs), carbon monoxide (CO), isoprene, persistent organic pollutants (POPs), benzene etc⁴.

These newly born fine and ultrafine size levels particles is serious threat and cause an elevated health risk due to the small size these PM to penetrate the furthest into the human organism and cause severe oxidative stress and the lungs is being inflammated to the exacerbation of asthma symptoms in susceptible individuals⁵. The toxicological effects of MC smoke can induce asthma, lung cancer, low reproductive toxicity in animals and low chronic toxicity to humans, although headache, nausea, dizziness etc. were observed⁶ and persistent wheeze in children, etc.⁷⁻⁹.

The objectives of the proposed work are:

- Investigation of PM_{2.5} concentration in the indoor environments during burning of materials (IS and MC).
- Investigation of concentration of PAHs (i.e. Phe, Cor, Fla, Bgh, Baa, Cry, Bbf, Ant, Dba,Ind, Pyr, Bkf and Bap) associated to the PM_{2.5} during burning of materials in the indoor environments.
- Evaluation of emission fluxes of the PM as well as their chemical constituents during burning of materials.
- Exposure assessment of toxicants associated with the PM and ash residues.

EXPERIMENTAL

Methodologies

Materials

Ten IS and MC materials of different made were selected for the present studies (Table 1). Most of the IS are manufactured from the mixture of fragrant gums, woods, resins, herbs and spices wrapped around a thinbamboo stick¹⁰. The IS materials are made by blending several solid scented ingredients into a paste and then, rolled the paste onto a

bamboo stick. Kapoor is also known as camphor, a waxy in nature, white or transparent solid having a strong, aromatic odour which give a lot of smoke when burnt. It is a terpenoid with the chemical formula of $C_{10}H_{16}O$ and found in wood of the camphor laurel¹¹. Similarly, MC are made of biomass base materials, which contains harmful and toxic insecticides such as pyrethrins, which account for 0.3-0.4% of coil mass. The remaining components of coil are aromatic and organic fillers, dyes, binders and other additives capable of fuming well. The fillers make up 99% of the MC¹².

S. No.	Materials	S. No.	Materials
IS1	Chandan	MC1	Hit
IS2	Gayatri	MC2	Jet
IS3	Bansuri	MC3	Mortein
IS4	Parivar 100	MC4	Tartoise
IS5	Bharat Darshan	MC5	Summit
IS6	Bhakti Puja	MC6	Coghlan's
IS7	Singarpuri	MC7	Murphy's
IS8	Dhoop	MC8	Off Patio and Deck
IS9	Gulab	MC9	CLD Brands
IS10	Camphor	MC10	Pic

Table 1: Name of IS & MC materials selected for experiment

Fuming of materials

A standard room $(3 \times 2 \times 3 \text{ m}^3)$ equipped with one window $(1 \times 1 \text{ m}^2)$ was selected for fuming of the IS and MC materials during October 2015. The window and the door were closed during fuming processes. The stand was used for their fuming. They were kept over in the stainless steel plate to collect the resulting bottom ash.

Collection and analysis of PM

The material IS and MC was burnt, and the PM generated was collected over the quartzfilter (47 mm) by sucking air with air samplers (Thermo Scientific Partisol and UC Davis, USA). The sampled PM filter paper was weighed by using the Mettler Toledo balance type AG245. For the calculation of mass distribution of the PM in the aerosol mass was divided with volume of the air passed through filter paper. For the evaluation of PM

fluxes the materials were burnt in a closed chamber $(0.5 \times 0.5 \times 0.5 \text{ m}^3)$ equipped with the UC Davis (USA) portable air sampler and exhaust fan. The PM emitted was collected over the quartz filter. The PM mass was weighed out and the calculation of flux was done by dividing the PM mass with amount of the material burnt. The weighed amount of the materials (IS and MC) was burnt over the titles and the ash residues produced were collected manually by the help of plastic spoon, further their mass was weighed out and the ash percentage (%) was recorded. The prepared samples were dried over 60 °C in the oven for overnight. The chemical species i.e. carbons and PAHs in the particulate matters and ash residues were analysed by using following techniques.

Flux measurement

The flux of $PM_{2.5}$ was determined by burning the materials in a closed chamber (0.5 x 0.5 x 0.5 m³) equipped with the room exhaust fan and a portable air sampler UC Davis (USA). The $PM_{2.5}$ emitted in a closed chamber was collected over the weighted 47 mm quartz filter. The $PM_{2.5}$ mass was weighted out, and the evaluation of flux was done by dividing the $PM_{2.5}$ mass with amount of the material burnt. The flux for the species associated to the $PM_{2.5}$ was calculated by using the following equation:

$$A_{flux} = PM_{flux} \times F$$

Where, A_{flux} = Fluxes of PAHs to the $PM_{2.5}$, PM_{flux} = PM_m/W , PM_m and W denote the mass of $PM_{2.5}$ in the filter and amount of the materials for burning, F = Chemical fraction in the $PM_{2.5}$.

Analysis of PAHs

A Dionex ASE 200 accelerated solvent extractor (Dionex GmbH, Idstein, Germany) having the extraction cells for leaching of the PAHs from the quartz filter samples was used. The cartridge was loaded with the weighed amount of the aerosol quartz filter and spiked with a solution of pyrene-d10 and 6-methylchrysene as internal standards, mixed with 2.0 g of pelletized diatomaceous earth (Germany, Separtis, Grenzach Wyhlen) and then extracted with hexane and acetone (7:3, v/v) mixture at pressure, temperature and flush volume of 10.7 Mpa, 373 K and 150%, respectively¹³. The extract (ca. 30 mL) of the ASE was filled in a Buchan vessel and reduced to a volume of ca. 0.3 ml on Buchan syncore evaporator controlled by a vacuum controller V-800 (Büchi, Flawil, Switzerland). The residual solvent was filled in the volumetric flask of 1 mL and evaporated under a gentle stream of N₂ in a Barkey vapotherm mobile S (Barkey, Leopoldshöhe, Germany). The volumetric flask was filled to the mark with acetonitrile, a 0.2 µm Spartan filter unit of 13/0.2 RC (Schleicher and Schuell, Dassel, Germany) solution is used for the sample filtered inbuilt with an auto

sampler vial and then used for HPLC analysis. For separating the PAHs an acetonitrile gradient temperature at 35°C and at a flow rate of 0.5 mL/min. The separation started with 58% acetonitrile, gradually increased to 100% in 35 min the content organic solvent and finally held at 100% acetonitrile for the last 12 min. An aliquot of the solution, 10 µL was injected, and detection and quantification PAHs were achieved with time programmed fluorescence detector (Table 2). For the quantification of 13PAHs HPLC method was externally calibrated in the range from 2.0-800 pg/µL with the help of chromatographic peak areas versus concentration as it has been observed the resulting correlation coefficients of the calibration curves (n = 6) were all higher than 0.99. The recoveries except for the high volatile analyte (i.e. naphthalene, acenaphthene and fluorine) of spiked quartz fibre filters and certified reference material were found to be good. This was also supported by the recoveries of the internal standards pyrene-d10 and 6-methylchrysene (80-98%), but nevertheless extraction efficiency and losses during evaporation and sample processing was controlled and corrected if necessary using the recoveries of the internal standards, respectively. During analysis of real samples calibration standards were analysed regularly and the calibration was renewed.

PAHs	Mol. structure	Retention time (min)	Ring number	λ _{em} (nm)	λ _{ex} (nm)	LOD (pg/µL)
Phenanthrene (Phe)	$C_{14}H_{10}$	12.3	3	370	246	0.6
Coronene(Cor)	$C_{24}H_{12}$	47.1	7	430	290	1.1
Fluoranthene (Fla)	$C_{16}H_{10}$	15.5	4	450	280	1.9
Benzo(ghi)perylene(Bgh)	$C_{22}H_{12}$	38.3	6	410	290	1.1
Benz(a)anthracene (Baa)	$C_{18}H_{12}$	21.9	4	420	260	0.7
Chrysene (Cry)	$C_{18}H_{12}$	27.5	5	420	260	0.7
Benzo(b)fluoranthene (Bbf)	$C_{20}H_{12}$	27.5	5	430	284	1.5
Anthracene (Ant)	$C_{14}H_{10}$	14.1	3	370	246	0.3
Dibenz(ah)anthracene (Dba)	$C_{22}H_{12}$	36.3	5	410	290	0.6
Indeno(1,2,3-cd)pyrene (Ind)	$C_{22}H_{12}$	38.3	6	500	250	0.1
Pyrene (Pyr)	$C_{16}H_{10}$	16.8	4	390	270	1.3
Benzo(k)fluoranthene (Bkf)	$C_{20}H_{12}$	30.2	5	430	290	0.7
Benzo(a)pyrene (Bap)	$C_{20}H_{12}$	32.1	5	430	290	0.8

Table 2: Optimal HPLC analytical conditions for PAHs analysis

RESULTS AND DISCUSSION

Distribution, emission fluxes and toxicities of PM in indoor air Distribution

The mean concentration of the PM in the indoor air during fuming of IS and MC is shown in Fig. 1. The concentration of $PM_{2.5}$ and PM_{10} for IS smoke (n =10) was ranged from 1374-7645 and 1465-8597 μ/g^3 with mean value of 4054 ± 3233 and 4296 ± 3284 $\mu g/m^3$, respectively. However, the concentration of $PM_{2.5}$ and PM_{10} for MC smoke (n = 10) was ranged from 260-677 and 384-784 $\mu g/m^3$ with mean value of 392 ± 210 and 522 ± 209 $\mu g/m^3$, respectively. The higher PM concentration was found with the IS than the MC smoke due to the higher fuming rates ($\approx 0.2 \text{ g/min}$), Fig. 1. The $PM_{2.5}/PM_{10}$ ratio for IS and MC smokes was ranged from 0.87-1.0 and 0.65-0.85 with mean value of 0.94 ± 0.02 and 0.75 ± 0.01, respectively. It means that all PM was mostly lie in the fine modes during fuming processes.



Fig. 1: Mean concentration of PM in indoor air during fuming of materials i.e. IS & MC

Emission fluxes

The emission fluxes of $PM_{2.5}$ for the IS and MC materials are shown in Fig. 2. The $PM_{2.5}$ emission fluxes for the IS and MC materials during the combustion were ranged from 1368-6159 and 10638-18799 mg/Kg with mean value of 4095 ± 3250 and 15856 ± 6644

mg/Kg, respectively. The higher emission fluxes of the PM were observed with the MC fuming, may be due to presence of ingredients i.e. sodium benzoate, potassium nitrate, etc.



Fig. 2: Emission fluxes of PM_{2.5} &ΣPAH₁₃ (mg/Kg)

Toxicities

The particulates generated during the fuming processes were generally in the fine modes^{14,6}. These particles produce strong pulmonary inflammatory responses in lungs alveoli^{15,16}. At least 95% particulates were found in the fine modes during the IS and MC fuming. In the present study, the mean $PM_{2.5}$ concentration for the all IS and MC smokes was exceeded the values recommended by ASHREA¹⁷ i.e. 65 and 150 µg/m³ for PM₂.

Distribution, emission fluxes and toxicities of PAHs in indoor air

Distribution

The mean concentration of thirteen PAHs i.e. Phe, Cor, Fla, Bgh, Baa, Cry, Bbf, Ant, Dba, Ind, Pyr, Bkf and Bap in $PM_{2.5}$ in the indoor air is summarized in Fig. 3. The concentration of total ΣPAH_{13} in the indoor air for the IS and MC smokes was ranged from 3417-6241 and 22-53 ng/m³ with a mean value of 4509 ± 4137 and 37 ± 20 ng/m³, respectively. The distribution trend of thirteen PAHs in the air with the IS smoke is: Dba > Baa > Cry > Bbf > Fla > Pyr > Bap > Bkf > Phe > Ind > Bgh > Ant > Cor. However, different trend with the MC smoke is: Phe > Cry > Baa > Ind > Bap > Ant > Bgh > Pyr > Bbf > Bkf > Dba > Fla > Cor.

Significantly, higher ΣPAH_{13} concentration with the IS smoke was observed, may be due to use of the perfume product as ingredients (Fig. 4).



Fig. 3: Mean concentration of PAHs₁₃ in PM_{2.5} in indoor air during fuming of materials i.e. IS & MC



Fig. 4: Value of ΣPAH₁₃ & BapE, Benzo (a) pyrene equivalent carcinogenic

Emission fluxes

The emission fluxes of ΣPAH_{13} for the IS and MC materials (n = 10) are shown in Fig. 5. The ΣPAH_{13} emission fluxes for IS and MC materials were ranged from 3.09-17.71 and 0.81-2.50 mg/Kg with mean value of 6.03 ± 5.43 and 1.46 ± 0.40 mg/Kg, respectively. The several folds higher fluxes of ΣPAH_{13} with the IS fuming were observed, may be due to higher fuming rates (≈ 0.2 g/min), may be due to addition of the organic ingredients.



materials i.e. IS & MC

Toxicities

Amongst thirteen PAHs, the Bap has been reported the most toxic compound. The Bap concentration in the IS and MC smokes was ranged from 124-895 and $0.5-5.7 \text{ ng/m}^3$ with mean value of 353 ± 535 and $2.3 \pm 2.2 \text{ ng/m}^3$, respectively. The recommended value of the Bap in the air reported^{13,18} is 1.0 ng/m³. The Bap concentration in the IS and MC smokes during the fuming was found to be 353 ng/m^3 and 6 folds higher than the recommended limit, respectively. Among 13 PAHs, six compounds i.e. Baa, Bbf, Bkf, Bap, Dba and Ind were reported in the list of carcinogenic compounds¹⁹. They have different toxicity and standardized with respect to most toxic compound (Bap). The benzo(a)pyrene equivalent (BapE) carcinogenic potentiality was calculated by using the following formula²⁰:

$$BapE = 0.06 (Baa) + 0.07 (Bbf) + 0.07 (Bkf) + (Bap) + 0.6 (Dba) + 0.08 (Ind)$$

The mean BapE value for the PAH_s in the IS and MC smokes was observed to be 931 ± 754 and 4.4 ± 3.9 ng/m³, respectively. The highest carcinogenic toxicity potentiality was marked with the IS smoke may be due to addition of the organic ingredients (Fig. 4).

Sources of PM and PAHs

The correlation matrix of PAH_s for the IS and MC smokes is presented in Tables 3 & 4. The good correlation (r = 0.81-0.99) of the PM among themselves in the IS and MC smokes was observed, indicating origin from the burning processes (Fig. 6). The PAHs are generated by the atmospheric reactions at the fuming temperature. The higher PAHs are

generally present in the particulate phase unlikely to the lower ones. The higher PAHs (i.e. Bkf, Bap, Bgh, Dba, Ind and Cor) were well correlated (r = 0.51-0.99) among themselves in the IS and MC smokes indicating origin during the fuming processes.



Fig. 6: Correlation of PM_{2.5} with PM₁₀ in IS & MC smoke

Table 3: Correlation (r) matrix of PAHs in IS smoke

	Phe	Cor	Fla	Bgh	Baa	Cry	Bbf	Ant	Dba	Ind	Pyr	Bkf	Bap
Phe	1.00												
Cor	0.96	1.00											
Fla	0.56	0.46	1.00										
Bgh	0.51	0.31	0.97	1.00									
Baa	0.15	0.11	-0.32	-0.06	1.00								
Cry	0.51	0.54	-0.16	-0.06	0.97	1.00							
Bbf	0.38	0.32	-0.18	-0.07	0.92	0.95	1.00						
Ant	-0.49	-0.65	0.38	0.53	-0.20	-0.54	-0.48	1.00					
Dba	0.49	0.24	0.94	0.87	-0.58	-0.42	-0.52	0.52	1.00				
Ind	-0.46	-0.65	0.45	0.52	-0.12	042	-0.39	0.99	0.48	1.00			
Pyr	-0.77	-0.84	-0.45	-0.28	0.51	0.09	0.35	0.56	-0.57	0.59	1.00		
Bkf	-0.74	-0.82	0.10	0.14	-0.42	-0.06	-0.68	0.90	0.34	0.89	0.56	1.00	
Bap	-0.57	-0.65	0.26	0.43	-0.07	-0.52	-0.38	0.99	0.38	0.98	0.68	0.89	1.00

	Phe	Cor	Fla	Bgh	Baa	Cry	Bbf	Ant	Dba	Ind	Pyr	Bkf	Bap
Phe	1.00												
Cor	0.95	1.00											
Fla	-0.35	-0.65	1.00										
Bgh	0.88	0.98	-0.61	1.00									
Baa	0.56	0.74	-0.45	0.85	1.00								
Cry	0.64	0.81	-0.34	0.91	0.98	1.00							
Bbf	-0.48	-0.64	0.88	-0.47	-0.07	-0.08	1.00						
Ant	-0.65	-0.79	0.89	-0.65	-0.28	-0.32	0.98	1.00					
Dba	-0.47	-0.68	0.97	-0.58	-0.28	-0.27	0.97	0.94	1.00				
Ind	-0.65	-0.84	0.88	-0.74	-0.31	-0.39	0.99	0.98	0.98	1.00			
Pyr	-0.44	-0.75	0.75	-0.59	-0.12	-0.18	0.98	0.87	0.85	0.97	1.00		
Bkf	-0.52	-0.73	0.99	-0.65	-0.54	-0.45	0.84	0.95	0.89	0.91	0.76	1.00	
Bap	-0.68	-0.89	0.95	-0.78	049	-0.54	0.92	0.99	0.96	0.97	0.88	0.98	1.00

Table 4: Correlation (r) matrix of PAHs in MC smoke

Chemical composition of PM

IS4

7.42

The fraction of the chemical constituents in the particulates (PM) is shown in Table 5. The total Σ PAH₁₃ content in the IS and MC PM was ranged from 3417-6241 and 22.3-53.7 mg/Kg with a mean value of 4509 ± 418 and 39 ± 30.4 mg/Kg, respectively. The high content of the PAHs in the IS PM is expected due to the fast fuming rates (≈ 0.2 g/min). The overall total fraction of carbons and PAHs in the IS and MC PM was ranged from 57.1-84.8 and 15.1-42.8% with mean value of 72.4 ± 10.5 and 27.5 ± 13.4%, 67.7-94.3 and 5.6-32.2% with mean value of 81.2 ± 10.4 and 18.78 ± 21.65%, respectively.

S. No. Phe Cor Fla Bgh Baa Cry Bbf Ant Dba Ind Pyr Bkf IS1 6.57 0.00 10.78 3.30 3.96 23.35 9.18 1.61 2.63 4.20 11.17 1.57 IS2 4.90 0.00 6.87 12.73 31.02 10.21 8.79 10.97 1.93 0.00 12.69 4.51 IS3 2.91 12.68 10.52 15.09 0.00 4.19 8.45 30.95 11.28 11.28 0.00 2.49

0.00 18.27 17.83 9.10 47.45 26.56 6.48 90.61 21.69 23.36 8.81

Table 5: Fraction of PAHs in PM_{2.5}, mg/kg %

Cont...

Bap

3.32

7.38

5.18

9.02

S. No.	Phe	Cor	Fla	Bgh	Baa	Cry	Bbf	Ant	Dba	Ind	Pyr	Bkf	Bap
IS5	9.56	5.94	14.17	0.00	12.08	11.17	6.09	24.77	17.36	16.93	4.66	33.19	25.73
IS6	2.77	5.46	0.00	3.56	5.45	1.87	7.78	4.80	7.51	6.82	11.28	3.52	0.00
IS7	25.00	11.65	27.57	15.58	43.45	0.00	19.32	0.00	24.22	10.39	15.78	26.46	10.73
IS8	2.33	0.00	5.61	1.67	1.67	6.03	6.11	1.28	6.43	1.32	0.00	4.87	11.93
IS9	25.33	0.00	7.57	17.10	17.16	18.99	19.17	7.40	21.60	21.60	0.00	27.10	19.17
IS10	35.54	9.01	23.62	15.70	0.00	21.00	21.00	15.26	38.08	18.60	15.55	17.08	39.39
MC1	1.58	0.45	1.13	2.33	2.33	1.95	1.95	0.71	0.56	2.22	1.95	1.20	1.80
MC2	0.89	0.00	0.39	0.69	1.77	1.28	0.95	1.15	0.92	0.00	1.34	0.30	1.87
MC3	1.35	0.00	0.75	1.03	0.42	1.77	0.00	0.89	0.18	0.50	0.68	0.24	0.77
MC4	0.30	0.42	0.00	0.64	0.98	1.52	0.38	0.70	0.36	0.92	0.18	0.68	0.48
MC5	0.35	0.21	0.31	0.83	1.26	0.86	0.71	0.41	0.38	0.34	0.18	0.77	0.16
MC6	0.20	0.00	0.18	0.84	0.46	0.48	1.12	0.00	0.00	0.38	0.36	0.30	0.14
MC7	2.92	0.69	0.39	0.00	0.00	0.59	0.95	0.79	0.26	1.70	0.79	0.62	0.69
MC8	2.11	1.07	0.69	0.81	2.15	1.84	1.88	1.61	1.11	2.07	0.77	0.31	0.00
MC9	2.96	0.51	0.00	0.39	1.54	0.90	1.86	1.22	0.55	2.06	1.13	0.55	0.32
MC10	1.59	0.27	0.30	0.63	1.30	0.90	0.00	1.69	1.03	0.93	1.03	0.93	0.70

CONCLUSION

The indoor environment is contaminated with very high content of carbonaceous PM during the burning processes, being several folds higher than in the outdoor environment. However, at least 5-folds higher concentration of the Σ PAH13 in the IS smoke was marked. The MC smoke is seems to be more dangerous than the IS smoke, due to the acidic particulate environment during fuming processes. In all cases, the concentration of the most toxic PAHs i.e. Bap was found several folds higher than the recommended value. The indoor air pollution is largely responsible to be expected for appearing of the respiratory diseases in humans exposed in.

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