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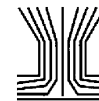
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Particulate Respiratory Dose to Indian Women from Domestic Cooking

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Particulate mass size distributions were measured during cooking and non-cooking periods in three Indian urban household kitchens with Liquefied Petroleum Gas as fuel. Based on the measured mass size distributions, fraction of particulate deposition in the respiratory system were calculated for a healthy Indian female using a deterministic lung deposition model. Respiratory physiological data of Indian women were collected from the published data. These physiological parameters were incorporated in the model to determine the particulate deposition in the respiratory system. The cooking generated very high concentration of particles 4 to 5 times more than the non-cooking background periods. Particulate size distributions in both cooking and non-cooking periods showed bimodal characteristics. Cooking process generated particles predominantly in accumulation mode (0.1–0.3 μm) whereas during non-cooking periods particulates are found in coarse mode (1.0–2.0 μm). Also, during frying process, the particulates were found to have a predominant coarser/droplet mode 0.7–1.0 μm . The highest deposition was observed in pulmonary region during cooking periods. The study shows that the daily particulate dose to the urban Indian women from domestic cooking is comparable with the dose resulting from outdoor particulate exposure.

1. INTRODUCTION

Indoor particulate pollutant concentrations are important in the determination of total human exposure to pollutants mainly because individuals spend a major fraction of their time indoors

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and so exposures to these contaminants inside residences can constitute a potential health hazard. The principal sources of airborne particles in indoor environment are cooking, smoking and re-suspension. In countries like India, the problem of indoor air quality faced by a huge population, as primarily identified in the various studies is due to the usage of biomass as cooking fuel (Dash 2002; Venkataraman and Rao 2001). Apart from fuel combustion, the Indian style of cooking, which involves considerable frying and grilling also generates large amount of vapors and aerosols. Vaporized edible oils and fat can nucleate/condense easily to form particulates in indoor environments and thus are considered as a major particulate source even when fuel combustion sources are absent or minimal. This is especially true for cooking in urban areas where the Liquefied Petroleum Gas (LPG) is used as cooking fuel. The constituents of these vapors and particulates are toxicologically relevant and are chemically irritative, carcinogenic and mutagenic (Rogge et al. 1991; Vainiotalo and Matveinen 1993). While there are some data available on particulate generation from biofuel cooking, there is dearth of reliable data on size distribution of the particulate matter from cooking with LPG in urban areas of India.

Many researchers have assessed Particulate Matter (PM) exposure from cooking in the Indian urban areas (Kulkarni and Patil 1999; Saxena et al. 2002). The estimation of potential health risks in these studies was based on the particulate concentrations in different microenvironments. However, health effects of the inhaled particulate are determined by complex sets of physiological and physical, chemical and biological properties of both respiratory system and the particulate (Dreher 2000). Therefore, size distributions and deposition potential of the particles on the vulnerable regions of the lung can be used as a more effective identifier to link particulate exposure and health effects. Empirical, morphologically based, stochastic or mixed type mathematical models are available to predict deposition plus clearance of particulate matter in humans. However, such models have not been used to estimate respiratory doses from

cooking in Indian subjects. In the present study, we measured mass size distributions of particulates during cooking events in typical Indian urban households. The respiratory deposition of these particulates generated from cooking was estimated using a lung mathematical model. We also examine the doses to women resulting from exposure to particles generated during non-cooking events. The pulmonary function data reported for Indian women were collected from literature and used as the inputs for the lung model. The main objectives of this article are to report particulate size distribution from cooking with LPG as fuel in Indian urban conditions, and to estimate the particulate respiratory dose to Indian women from cooking. The morphological parameters used in the present model are scaled down from a "standard size man" (Yeh and Schum 1980). Therefore, the calculated deposition may differ from actual deposition on Indian women. However, the deposition patterns computed in the present study for Indian women may represent average population values.

2. METHODOLOGY AND EXPERIMENTS

2.1. Study Area Description

In this study all the experiments were carried out in the residential area of IIT Bombay campus. The present study was a part of one-year (June 2003–June 2004) study to assess the exposure from indoor air pollutants to campsites with women as target group (Gangamma 2004). The campus is in the suburbs of Mumbai and is in a relatively clean area so the impact of outdoor pollution is low. The houses were typical middle-income group residences in the region. The measurements were carried out in the kitchen of three different houses, two of the houses (Denoted as H-1 and H-2) were three rooms while the third (denoted as H-3), was two room tenement house. The houses were occupied during part of study period. Cooking was found to be the major indoor activity for generating particulates. These houses were devoid of air condition or any other air regulating equipments except air circulating ceiling fans. The kitchens had no particulate control systems other than the natural ventilation. The salient features of the study houses are shown in Table 1.

2.2. Particulate Measurements

Size segregated particulate mass was measured by cascade impactor with Quartz Crystal Microbalance (PC-2 QCM, California Measurements, INC, USA). The sampling flow rate of 0.240 lpm was found suitable for continuous indoor particulate monitoring without creating any airflow of its own. The impactor has ten stages and segregates aerosol samples into ten size ranges between 25 and 0.05 microns. The cut sizes for the impactor were reported for spherical particle of density 2 g cm^{-3} by the manufacturer. However, these cut diameters are different from the values reported in the calibration studies on the instrument (Fairchild and Wheat 1984; Horton et al. 1992). In this

TABLE 1
Salient features of the test houses and average and standard deviation of activity time of women in kitchen

House no	Kitchen volume m^3	Kitchen area m^2	Openings (area in) m^2	Height (m)	Activity time	
					Cooking (min)	Non cooking (min)
H-1	26.73	8.92	2.35	3.00	185 ± 29	80 ± 20
H-2	19.62	6.85	3.68	2.86	157 ± 53	70 ± 24
H-3	23.10	7.80	2.99	2.96	132 ± 73	40 ± 09
Average					158	63

study, the cut sizes of the impactor were adopted from Horton et al. (1992). The performance of the crystals were stable and satisfactory for the aerosols generated by cooking which consists of oily and greasy matter. The particle concentrations observed on stages having cut sizes greater than $3.1 \mu\text{m}$ were very low and found to be statistically unreliable. Therefore, only the mass measured by stages having cut sizes less than $3.1 \mu\text{m}$ were using in our assessment. A study conducted in the laboratory showed that the stability of the quartz crystal is deteriorated in presence of high moisture. But, such high moisture conditions did not occur in the kitchens during our study. Flow rate and leak check were conducted on the impactor using a rotameter before and after each experiment. The rotameter was calibrated with an electronic flow calibrator (SKC instruments Model 712).

Sampling was carried out at a distance of one meter from the cooking place and at a height near to the occupants breathing level. The particulate concentration and size distribution at this height was assumed to represent the occupants actual exposure during cooking. Each house was monitored for two consecutive days. Normally, each day consists of three sets of cooking events. One hour before and after each cooking events were also monitored. The particulates were sampled inside the kitchen with intervals in the range of 5–10 minutes. The number of samples per session is ranged between 4 and 9. The data collected during each session are averaged and are used for further analysis. Depending on the particulate concentration, sampling time was varied between 20–60 seconds for the measurements. The frequency of the crystals was checked regularly to avoid saturation by overloading. If any crystals found overloaded it was taken out and cleaned with Hexane. The temperature and humidity were measured during the experimental period. The measured mass size distribution data were entered into computer manually for the analysis. To extract size distribution parameters, a lognormal was fitted over the mass size distribution data, based on the stage efficiency kernels (Horton et al. 1992) using a least square minimization principle with a priory constrain of lognormal distribution of particulate mass (Ramachandran et al. 1996). The observed low values of least square error during fitting suggest that the particle mass size distribution follows a lognormal

distribution or its combinations. Mass median aerodynamic diameters and geometric standard deviations of the particle size distribution were determined from the fitted curve.

2.3. Lung Deposition Model

In the present study, deposition fractions of particulates in the human respiratory system were computed using a simple deterministic model (Schum and Yeh 1980; Yeh and Schum 1980). The model initially calculates particle deposition fraction during one breathing cycle that includes inspiration, pause, and expiration. Given breathing and lung parameters, the amount of particles deposited in the lung can be calculated from deposition fraction during one breathing cycle. Respiratory dose for an exposure period can be easily found from the amount deposited in one breathing cycle. The extra-thoracic deposition was improved in the model based on empirical models suggested by Martonen and Zang (1992) and Swift et al. (1992). The model is also modified to use the physiological data of Indian population. Functional Residual Capacity (FRC) values are used to scale the lung initial model to suit physiological dimensions of Indian subjects.

2.4. Respiratory Physiological Parameters

The particulate deposition fraction is a strong function of lung dimensions and other physiological parameters, which in turn are functions of various factors like gender, race, and age (ICRP 1994). The physiological data for Indian population were collected in this study from literature (Kamat et al. 1967; Jain and Ramaiah 1967; Udawadia et al. 1987; Vijayan et al. 1990; IAEA 1998). Overall, scattered data available through these studies have shown that physiological parameters of Indians are different from Caucasians, on which lung deposition studies are mostly done (ICRP 1994), and have distinct geographical variability among the Indian population. For example, the static lung volumes, functional residual capacity and Total Lung Capacity (TLC) are about 30% lower for average Indian population than the Caucasians, and the South Indians have comparatively lower values than the rest of the population (Vijayan et al. 1990; IAEA 1998). Also, most of the collected data lacks age and activity specific information. The TLC is varied in the range of 4.6–5.35 L and 3.50–4.20 L for adult men and adult women, respectively. The tidal volume for Indian subjects is in the range 0.51–0.61 L and 0.35–0.42 L, respectively, for adult men and women. Similarly, FRC varied between 2.4–2.8 and 1.9–2.13 L, respectively. In the present study, average lung physiological data of an Indian subjects are used. The respiratory physiological parameters used for the study are of female subjects, primary targets of the particulate exposure from cooking and are summarized in Table 2. Nose breathing and a symmetrical breathing cycle with zero breath hold time was assumed. Breathing rate corresponding to the household activities are adopted from Adams (1993) and is assumed constant over the exposure time. Since activity based lung function data is not available for Indian women, these val-

TABLE 2

Values of respiratory physiological parameters utilized in the model together with some anatomical parameters for an average adult Indian women

Measurements	Unit	Adult Indian women*	
Total lung capacity	L	3.70	
Functional residual capacity	L	2.04	
Extrathoracic volume	ml	40	
Age	Years	30	
Height	cm	151	
Weight	Kg	44.20	
Physical activity		Sitting awake	Light exercise
Tidal volume	L	0.38	0.82**
Breathing rate	min ⁻¹	15	22***

*Vijayan et al. 1990, Udiwadia et al. 1987 and IAEA 1998.

**Extrapolated using Caucasian women data ICRP 1994.

***Adams W. C. 1993 for household activities.

ues are extrapolated from available data using Caucasian woman lung function (ICRP 1994) as a reference.

3. RESULTS AND DISCUSSION

3.1. Particle Size Distributions for Cooking and Non-Cooking Sessions

The particle size distributions in the kitchen were monitored during cooking and non-cooking sessions. The concentrations of particulates in kitchen were found to vary rapidly and widely during various activities. Therefore, the total period of activities in the kitchen were divided into two groups cooking and non-cooking sessions. The cooking period, which involves combustion of fuel, leads to high concentration of particles compared to background. In contrast, non-cooking periods represent the presence of the occupant in the kitchen without any combustion activities. During non-cooking periods, which were mostly before and after the cooking periods, the particle concentrations were comparable to the background values. The primary sources of particle during non-cooking sessions are particle resuspension due to various activities by occupants, residual particles from cooking and infiltration from outdoor. Average concentrations and mass size distribution parameters of particulates from cooking and non-cooking sessions in kitchen in each house are summarized in Table 3. The data presented in the table are the averages of the cooking and noncooking sessions and standard deviation between sessions (average six sessions per house). The concentrations of the particles during non-cooking were in the range of 25–100 $\mu\text{g}/\text{m}^3$ with an average of 57 $\mu\text{g}/\text{m}^3$. The observed concentrations during non-cooking periods varied between cooking sessions, and between days. However, no particular trend is observed for intra house concentration data during non-cooking. The average mass size distributions of particles in

TABLE 3
Summary of observed average mass size distribution parameters with standard deviations of particulates from cooking and non-cooking sessions in kitchen

House	Fine fraction	MMAD1 (μm)	GSD1	MMAD2 (μm)	GSD2	Concentration ($\mu\text{g}\text{m}^{-3}$)
Cooking						
H1	0.77 ± 0.09	0.30 ± 0.05	1.38 ± 0.11	1.52 ± 0.52	1.65 ± 0.31	262 ± 69
H2	0.82 ± 0.03	0.26 ± 0.01	1.40 ± 0.09	1.14 ± 0.15	1.32 ± 0.21	223 ± 19
H3	0.80 ± 0.03	0.24 ± 0.01	1.31 ± 0.07	1.54 ± 0.07	1.33 ± 0.11	240 ± 20
Average	0.79 ± 0.07	0.27 ± 0.04	1.36 ± 0.10	1.45 ± 0.40	1.49 ± 0.29	248 ± 53
Non-cooking						
H1	0.19 ± 0.06	0.23 ± 0.02	1.27 ± 0.14	1.38 ± 0.22	1.47 ± 0.15	70 ± 30
H2	0.27 ± 0.08	0.28 ± 0.02	1.45 ± 0.10	2.11 ± 0.26	1.34 ± 0.03	60 ± 8
H3	0.32 ± 0.07	0.31 ± 0.04	1.44 ± 0.14	1.69 ± 0.23	1.38 ± 0.04	40 ± 23
Average	0.27 ± 0.08	0.28 ± 0.04	1.40 ± 0.21	1.79 ± 0.36	1.39 ± 0.09	57 ± 23

each house during non-cooking sessions are plotted in Figure 1a. The average particulate size distributions during non-cooking were found to be bimodal with MMADs $0.28 \pm 0.04 \mu\text{m}$ and $1.79 \pm 0.36 \mu\text{m}$ with GSDs 1.40 ± 0.21 and 1.39 ± 0.09 . The particulate size distribution shows a predominant coarse mode present in $1.0\text{--}2.0 \mu\text{m}$ ranges with average mass fraction 0.73. The possible source of larger sized particles was the resuspension of particles from cleaning or other activities of the occupants and the variability in mode size and mass fraction (Figure 1b) indicated a source of highly varying characteristics.

Highest indoor concentrations during non-cooking periods are observed in H1 with considerable variation between concentrations in houses. The lowest concentrations were observed in H3 that may be attributable to the activity level of the occupant in the house. It has observed that the activity level in H3 was less than that of other two houses. This is also supported by the fact that the difference in concentration between H1 and H3 is mainly due to the coarse fraction content 81 versus 68%, respectively. But the concentrations in the indoor are not only governed by indoor activities, but also by the outdoor concentrations and exchange rate. Therefore, it is not possible to fully attribute the variation between houses to the indoor activities.

Generally, cooking produced 4–5 times more particle concentration than the non-cooking periods. The average particle concentration during cooking was $248 \mu\text{g}/\text{m}^3$ with a standard deviation of $53 \mu\text{g}/\text{m}^3$. The particle concentrations during cooking varied widely and ranged up to $1700 \mu\text{g}\text{m}^{-3}$. The average mass size distributions of particles generated during cooking in the study houses are plotted in Figure 2a and 2b. Most of the mass was concentrated below $1 \mu\text{m}$ diameter (fine particles). Distinct accumulation and droplet modes were apparent. The particles generated from cooking activity are of two origins. One is originated from the combustion process of the fuel and second due to emission from the cooking activities like frying, boiling, and so on. In this study, experiments were conducted in

urban households using LPG as the fuel for domestic cooking. LPG combustion produces fine particulates along with limited amount of volatiles whereas, cooking process may produce considerably large amounts of condensable vapors and fine/coarse particulates. Primary particles generated by combustion of LPG have size less than $0.040 \mu\text{m}$. These particles rapidly coagulate to form larger particles of around $0.10 \mu\text{m}$ (Dennekamp et al. 2001). Thus, the accumulation mode is formed from rapid coagulation of ultra fine particles generated during combustion process. On the other hand, vapors with low saturation vapor pressure can nucleate to form new particles or condense onto existing particles. Condensation of volatiles on accumulation mode particles may have formed the droplet mode particles. Distinct accumulation and droplet modes were found to be present only during the cooking process, which involves frying with oil and fatty matters (Figure 2b). This figure shows that the frying process produces a higher proportion of bigger particles than with the normal cooking process and is also expected to generate large amount of condensable vapors. Earlier studies on the particulate size distribution from cooking process also yielded similar results, showing bimodal distribution with accumulation ($0.05\text{--}0.2 \mu\text{m}$) and droplet mode ($0.4\text{--}0.8 \mu\text{m}$). The droplet mode diameter was found increased up to $1.6 \mu\text{m}$ for high fat meat cooking (Annis and Annis 1989). Li et al. (1993) also reported similar particle size distributions during frying process. Particulate concentrations during normal cooking and cooking involving oil frying are plotted in Figure 3. Frying processes generated higher particulate concentrations than in the normal cooking process.

In general, the particle generated during cooking was found mostly in accumulation mode (median diameter $0.27 \mu\text{m}$) except during oil frying, with a mass fraction of 0.79. The concentration measurement during cooking showed large intra house variation. This is partly because the particle generation is a discrete event in the cooking process rather than a homogeneous continuous process. The pattern of the cooking, for example oil

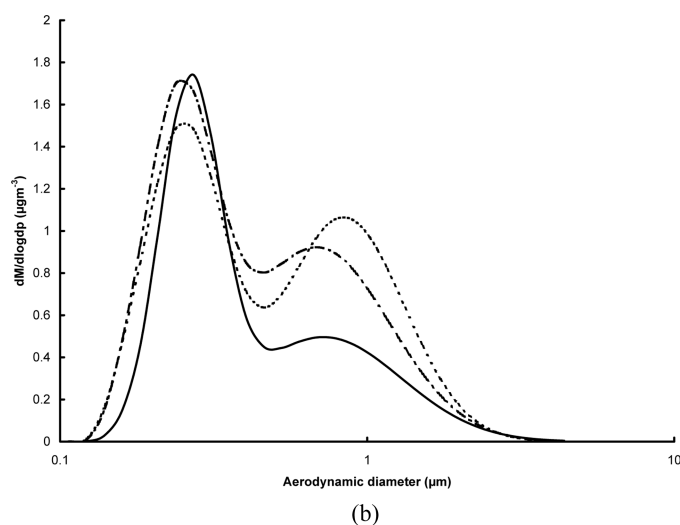
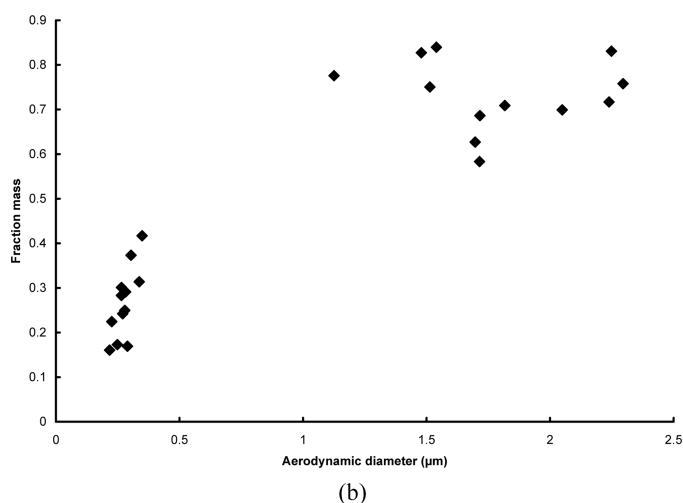
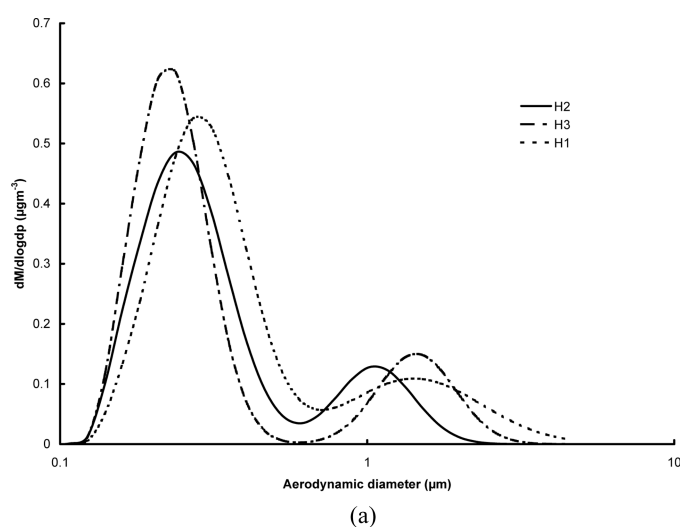
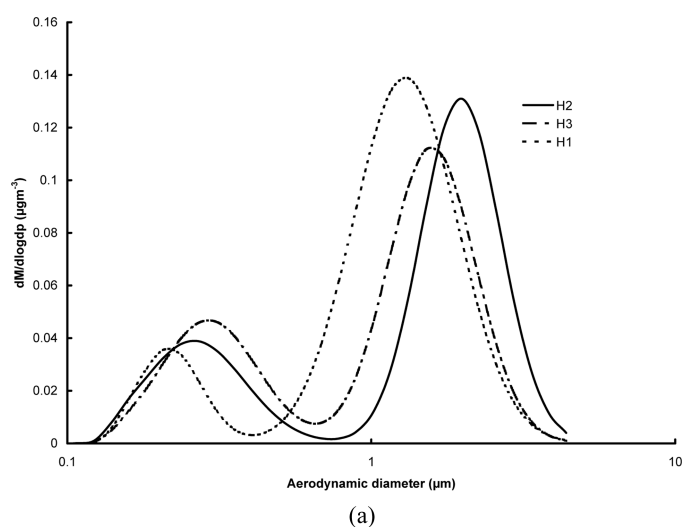


FIG. 1. (a) Particulate mass size distribution observed during non-cooking sessions in three houses, H1, H2, H3. Particulate mass is primarily concentrated in coarse mode. (b) Particulate mass distribution during non-cooking periods measured using QCM. Mass mean diameters are retrieved from stage mass concentrations with the assumption of lognormal distribution and are plotted against respective mode concentration.

FIG. 2. (a) Particulate mass size distribution observed during cooking sessions in three houses, H1, H2, H3. Particulate mass is primarily concentrated in fine accumulation mode. (b) Typical particulate mass size distributions produced during cooking process involving frying of oil and fatty substances. The mass size distribution shows distinct modes of accumulation and droplet.

frying, is also govern the particle concentration and thus added to the variation observed. The concentration variation within a day, between different cooking sessions was found very high (SD $69 \mu\text{g}/\text{m}^3$) for H1. The variation between daily averaged concentrations (SD $29.70 \mu\text{g}/\text{m}^3$) was found much lower than the variation within a day. This observation is expected because the cooking pattern is varied within a day whereas there is a pattern of cooking followed in each day. But the detailed information of such variations of particulate concentration with cooking pattern is limited with the small number of measurements carried out in the present study and needs further investigation. The variation in concentrations for both cooking and non-cooking periods was higher in H1 compared to the other two homes. The average time spent engaged in these activities was also higher in H1 relative

to the other homes. The between home variation in size distribution parameters was small compared to the observed variations in particle concentrations (Table 3).

3.2. Daily Dose from Particulates Exposure During Cooking and Non-Cooking Periods

Daily dose of the particulate matter was estimated based on the concentrations and deposition efficiency data along with time activity pattern of the female (Table 1) occupants during the study period. A detailed activity pattern had been noted down in the kitchen of each house. Average values of duration of cooking activities in kitchen were in the range of 2–3 hours per day and varied from house to house. The non-cooking activity in kitchen is a summation on several small time activities including cleaning, washing operations. This time averaged to be around

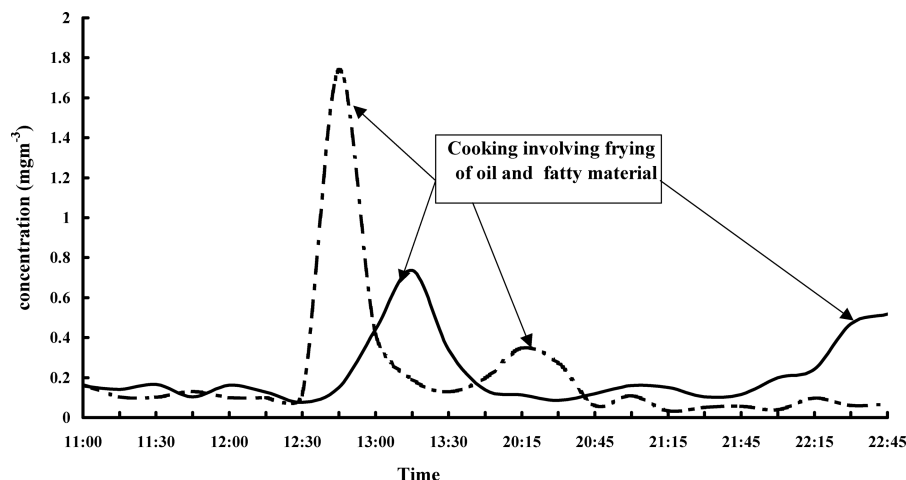


FIG. 3. Variation of concentrations of particulate matter during different types of cooking in house (H1) in two days observation.

one hour for all the houses in day. Average pattern of the three houses studied shows a total period of 3.7 hours spent per day in kitchen with 2.6 hours of cooking and 1.1 hour for non-cooking activities.

The calculated values of particulate deposition patterns and regional lung doses averaged over all the houses for Indian adult women expressed as $\mu\text{g day}^{-1}$ are given in Table 4. The contribution from each mode of the bimodal particle size distribution observed is also summarized in the Table 4.

During cooking, 23.23% of the total inhaled particle is deposited in the respiratory system with 14% deposited in pulmonary region. About 18% of the inhaled accumulation mode particle mass (Mode1) is deposited in the respiratory tract with 11.70% in the PUL, 3.77% in TB and 3.10% in NP regions. About 40.80% of the inhaled coarser particles are deposited in respiratory tract with 23.29% in the PUL, 7.0% in TB, and

10.47% in the NP regions. Major part of mass of the cooking generated particles is found in accumulation mode (Mode 1). But the efficiency of the cooking generated particulate matter deposition is found comparatively less than the coarse mode particle as the size of cooking generated particle coincides with the deposition minimum.

For non-cooking periods, 9.6% of the total inhaled particle is deposited in the respiratory system with 5.3% deposited in pulmonary region. For this period accumulation mode (Mode 1) and coarse mode (Mode 2) contributed 1.4% and 40%, respectively, of corresponding modal mass to the total deposition. The inhaled accumulation mode particle mass (Mode 1) is deposited in the respiratory tract with 0.93% in the PUL, 0.3% in TB, and 0.24 % in NP regions. During cooking as well as non-cooking periods particulates in the both modes were predominantly deposited in pulmonary region. The thoracic deposition of particulates per day was $178.40 \mu\text{g day}^{-1}$ and $67.46 \mu\text{g day}^{-1}$ correspondingly for cooking and non-cooking periods. The average thoracic particulate deposition of an adult with a tidal volume of 1000 cm^3 and breathing rate of 15 per minute ($21.6 \text{ m}^3 \text{ day}^{-1}$) in an urban setting is reported to be around $231 \mu\text{g day}^{-1}$ (Venkataraman and Kao 1999). The above calculation was carried out based on an average outdoor concentration of $150 \mu\text{g m}^{-3}$ (MMD 0.2 and $5.0 \mu\text{m}$, GSD 1.7 and fine fraction 0.57). The particulate dose to women during cooking alone is comparable to dose due to the total outdoor exposure. Therefore, the study points out that the particulate respiratory dose to women is significantly governed by the exposure during domestic cooking.

TABLE 4

Particulate deposition patterns and regional lung deposition averaged over all the houses for Indian adult women and particulate doses per day ($\mu\text{g day}^{-1}$) during cooking and non-cooking sessions

Lung Region	Daily dose ($\mu\text{g day}^{-1}$)			
	Pulmonary	Trachea-bronchial	Nasopharyngeal	Total
Cooking				
Mode1	46.93	14.18	21.10	82.21
Mode2	88.75	28.54	23.51	140.80
Total	135.67	42.72	44.61	223.01
Non-cooking				
Mode1	7.04	2.25	1.86	11.15
Mode2	43.92	14.25	22.85	81.02
Total	50.96	16.50	24.71	92.17

4. CONCLUSIONS

Our study showed that the particulates during cooking sessions are primarily in fine accumulation mode. The particles generated from cooking causes high respiratory dose in the pulmonary region of the human respiratory system with implications on the health of the occupants, especially for women.

Studies investigating the adverse effects of Particulate Matter, in urban settings have used size specific or non-size specific outdoor ambient mass concentration of particulates as the indicator for the particulate exposures. Though this is possibly the most simple and most accessible indicator, it may not accurately represent the respiratory dose of inhaled particulates. The study pointed out that the daily particulate dose to the urban Indian women from domestic cooking is comparable with the dose resulting from outdoor particulate exposure. The particle size distribution during cooking showed that cooking activities, especially oil frying generates the large portion of particles other than fuel combustion. The particulate size distribution has found vary between cooking conditions and affected the respiratory dose. But the variation of size parameters was found low compared to particulate concentration variations. There is a paucity of physiological data of Indian subjects available for lung deposition prediction. Such data are essential for accurate prediction of toxic effects of inhaled particulates.

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