Original Paper

Measurement of Surface Area Concentration of Fine Particulate Matter in Indoor Environments

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Abstract

Indoor exposure to fine particles (0.01-2.5 μm) is measured in the present study using Surface Area Concentration (SAC) metric for two indoor environment categories based on the cooking fuel used—kerosene and Liquid Petroleum Gas LPG. Study of SAC is very important from the point of view of health aspects. From the collected data, Arithmetic Means, Geometric Means, peak concentration values were determined to compare between the two categories of Indoor environment. The results show the two environments to be significantly different. Also, the results indicate remarkable high indoor surface area concentrations during the cooking and other combustion processes.

Keywords

Fine particulate matter, surface area concentration, air quality, indoor combustion, real-time measurement

1. Introduction

Solid or liquid particles suspended in air form aerosols. Particulate portion is usually referred to as Particulate Matter (PM). PM is chemically and physically diverse substance and range in a wide size range. Aerosols are generated from various sources such as dust, soil, volcanic eruptions or due to anthropogenic processes like combustion activities, agricultural, commercial or industrial practices. Then they also change because of processes such as condensation, nucleation etc. For indoor concentration of aerosols, we can consider the following as the sources: cooking, smoking and other combustion activities, pollen, fungal spores, molds, and bacteria, besides the influx of outdoor aerosols such as road and soil dust.

Fine particulate matter caused air pollution has been considered in recent years as a serious environmental concern. Recent studies (Bruce et al., 2000; Smith, 2000) have found that the respiratory diseases such as TB, cardio-vascular diseases, COPD etc. are associated with the long-term exposure to

fine particulates. India has second highest per capita burden of respiratory illnesses in the world (Smith, 2000). About one million deaths are reported annually in India due to household air pollution caused by fumes from cooking, heating and lighting activities. Till few years back, Mass Concentration (MC) of PM was used to assess the health impacts. Now we know other metrics such as Number Concentration (NC) and Surface Area Concentration (SAC) metrics are important as well and this is confirmed by the studies on cell cultures and animal respiratory system (Brown et al., 2001; Oberdorster, 2000; Tran et al., 2000; Heitbrank et al., 2009). We have already studied and reported the results on number and mass concentration in Indoors (Monkkonen et al., 2004).

Of all the PMs, the fine particulates (0.01-2.5 μm) have a greater probability of delving deep into the lungs (Zhou & Cheng, 2000; Churg & Brauer, 2000). Ratio of surface area to mass becomes greater as we move towards small diameter particles. For a given mass, surface area increases as 1/diameter. Hence the necessity for expressing the regional dose in terms of surface area concentration in the epidemiology studies. Jicheng Gong et al. (2019) study shows that particle size ranges having large SAC and/or volume concentration have significant positive association with pulmonary inflammation. The recent USEPA particle size chosen as standard for epidemiological studies is $PM_{2.5}$, which are particles smaller than 2.5 micrometer in mean aerodynamic diameter (USEPA; 1996; Schwartz et al., 1996). In the (Sahu et al., 2011) study conducted in rural India, the correlation between $PM_{2.5}$ and surface area concentration was found to be low to moderate. The study showed correlation coefficient R=0.38 in the tracheobronchial region and R=0.47 in the alveolar region. This indicates that $PM_{2.5}$ is not a sufficient proxy for particle surface area concentration.

However, to conform to the latest findings, new standard of $PM_{2.5}$ is to be adopted for further study. The USEPA Clean Air Scientific Advisory Committee (CASAC) review (Wolff, 1996) suggested a 24-hr $PM_{2.5}$ standard in the range of 20 to 65 $\mu g/m^3$. Many studies (Kamens et al., 1991; Lighty et al., 2000) have indicated that cooking is the primary activity responsible for generating fine particles in Indoors. According to Kamens et al. (1991) study, particles below 1 μm are major constituent of the emission. The average diameter of these particles in the emission was around 0.8 μm. A recent study (Patel et al., 2020) has confirmed this where they found that NC was highest for sub-10 nm particles during cooking activity with kerosene stove.

In such a study, some of the important tasks included are: measurement of exposure with a particular exposure metric, comparing them with the existing emission or exposure standard, understanding their impact on the exposed population, and also carrying out source-apportionment of the aerosols monitored.

In India, many studies have been conducted on the health effects of biomass fuel usage (Smith, 1986). However, very few studies have compared the kerosene and Liquid Petroleum Gas LPG fuels which are also commonly used for household purposes. According to a recent study (Elf, Jessica L et al., 2018), 57% of urban homes use LPG as the lone and primary fuel, whereas kerosene and wood is used as a secondary main fuel in about 22-25% of homes.

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In India, especially in economically poorer section, women spend primary part of their time indoors cooking with crude fuels with poor ventilation and, also the stoves are inefficient. Kerosene is being used by economically lower-class people who live in poorly ventilated small houses. Studies in India have concentrated on the mass concentration of PM_{10} and PM_5. In the present study SAC of fine particles are measured. We are going to compare the obtained data with respect to the type of fuel used indoors and as well as study their variation patterns.

The present study is conducted in Mysore city situated in the state of Karnataka having average minimum temperature of $15^{o}C$ and maximum temperature is $35^{o}C$. It is a small city with a population of 893.062 (Census of India 2011) and an area of more than 150 sq. km. Cooking activities using kerosene and LPG as main fuel with poor ventilation facility found in many of the houses.

2. Method

In the process of aerosol measurement, the first part is collecting a small sample from the bulk aerosol for analysis. The process is referred to as aspiration. These particles are transported to the measurement instrument. The particles are analyzed in real-time (dynamic measurement). Prior to the collection or analysis, an additional step called pre-classification is added to get an expected aerosol for sampling. For example, when we want to measure particles smaller than 10 μm, we have to remove all particles of size larger than 10 μm before collecting the sample.

2.1 Instrument Used

Diffusion Charger (e.g., LQ1-DC, Matter Engineering, AG)

The technology necessary to measure aerosol exposure in terms of SAC is not widely available. In the present study we are going to use LQ1-DC (Matter Engineering, Switzerland), to directly measure active surface area by the method of diffusion charging.
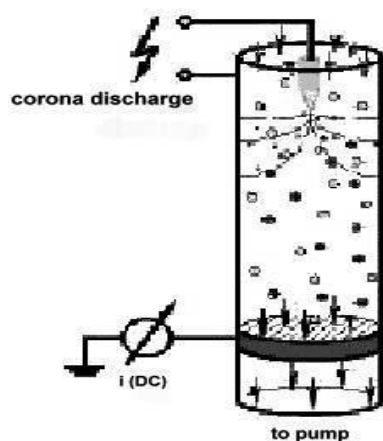

Figure 1. Mechanism of Unipolar Diffusion Charging

Source: www.matter-engineering.com

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The unipolar diffusion charging mechanism used is shown in Figure 1. A small positive voltage of approx. 20 V DC is applied to the grid. Aerosols in the gas stream are guided between the grid and the grounded tube. With the corona discharge from the corona tip to the grid, ions get produced in the carrier gas. These ions attach to the surface of the aerosol particles. The particles carrying positive charge flow to the sensor unit and are absorbed in the measuring filter. The filter is connected to a ultra low current amplifier (resolution 1 fA=10^{-15} A). The voltage signal from the charge amplifier is converted to the calibrated analogue signal 0…5 V DC. This signal is displayed as 0…1000 after adjustment.

The LQ1-DC instrument measures the integral attachment cross section of ions on particles. It is the part of the geometrical surface which is directly accessible from outside; hence the use of term "active surface" or "Fuchs surface area". The measurement gives the total active surface area per unit volume. To overcome multiple charging for particles larger than 10 nm, the in concentration is kept low. Due to coulomb repulsion, the ion attachment rate in case of multiple charging decreases.

We now discuss the technical specifications, pre-sampling set-up, of the instrument LQ1-DC used for measuring the SAC.

Figure 2. LQ1-DC Instrument to Measure Surface Area Concentration

Data logging time-interval for Indoor measurements was set to 10 sec. Before each sampling, Electrical Zero is adjusted to zero using potentiometer.

Response time: few seconds

Particle size range: few nm to 2 μm.

Resolution: 1 $\mu m^2/cm^3$

Gas flow rate: approx. 1.5 l/min

2.2 Sampling Procedures

The study is carried out in the city of Mysore in different households using either kerosene or LPG fuel for cooking. Five houses of each type were randomly selected from different parts of the city. Kerosene and LPG houses were chosen in pairs such that a kerosene house lied in an area close to a LPG house.

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It was also noted during the selection of the houses that the households also used some other fuels wood, dung cakes etc. All measurements were carried out in the period from August to January.

On the day of sampling, the LQ1-DC instrument was taken to the sampling location and were run by battery for 24-hr to collect the real-time SAC data. The instrument was kept in the living room at a height of about 50 cm from the floor.

Main activities within households including cooking were noted down by the family member along with the corresponding timings. After a gap of 4 months, measurements were repeated for the second time in each house. The sampled data stored in the memory unit were downloaded to the PC from the instruments after each sampling.

2.3 Analysis of the Data

Statistical analysis of 1-minute-average time-aligned data file was made using Microsoft Excel software. Statistical entities like the arithmetic or geometric mean averages, standard deviation and variance were calculated from data files of each sampling site using the functions provided in the software. These results were used to make comparisons between the two types of environments. Also, variation aspect of the metric data was studied with these results. The two types of sampling categories were checked for statistical differences in terms of metric concentrations.

To find statistical difference, following procedure was used. First, the mean and standard error of the surface area concentrations of two groups, e.g., kerosene and LPG houses were obtained. Let the number of houses for kerosene and LPG groups are N_1 and N_2 respectively. The standard error for each group, e.g., kerosene group is obtained as

$$SE = \frac{Std.\,Deviation}{square\ root\ of\ (N)}$$

The two groups are statistically different if their error bars do not overlap. For example, let the mean and standard error for kerosene be m_1 and se_1, and those for LPG be m_2 and se_2. Then if $(m_1-se_1) > (m_2+se_2)$, (i.e., the lower bound for kerosene is greater than the upper bound for LPG) then, the two groups are statistically different. Otherwise, the two groups are not different.

The statistical results of the analysis in the present study are presented in the next section in terms of averages and standard deviation representing the particular environmental category. Standard deviation actually gives the amount of dispersion involved in the metric levels. From the variation levels, we can know if any significant change in concentration levels occurs over a day, or over different sampling sites.

3. Results and Discussion

In this section, we will discuss analysis of the data collected to know the particular association of some of the sources with concentration levels, the daily variation pattern, and the average SAC concentration levels for the two indoor categories and comparison between them.

3.1 Association of Peaks with Activities.

The data collected from the LQ1-DC were mapped with the activities recorded by the housewife in the activity log sheet. Following associations were found in general.

Cooking Activities identified with the peaks

   i. Heating water for bath: Time: Morning 6 to 9 am, A high unique peak

  ii. Breakfast: Time: morning 7-9 am

 iii. Meals: Time: morning 11-12 noon, A moderate peak

  iv. Evening cooking: Time 5-7 pm, Moderate peaks

   v. Late night: 11 pm-4 am, steep decrease in concentrations reaching minimum levels.

Along with the cooking fuel, the household were found to use alternate fuels such as coconut husk or dung cakes. The unique high peaks observed in surface area concentration corresponded to this activity of heating water for bath noted down in the activity log sheet which usually occurred in the morning hours from 6 am-9 a.m. (see Figures 3 and 4). This is a major aerosol generating activity and usually would last for 2-3 hours.

Other peaks during the day also corresponded to the activities noted down in the activity log sheet. All other peaks afterwards are found to be associated with the household activities mainly the cooking processes, such as preparing tea, breakfast, meals, etc. So, to further understand the association of aerosol properties with the activities, it is necessary to study each of the peaks relating to the particular activities in detail.
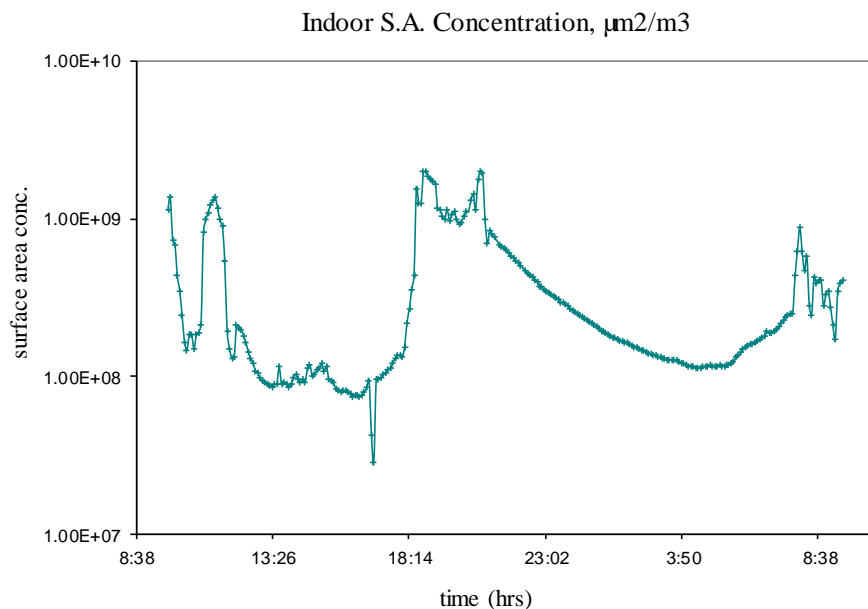
Indoor S.A. Concentration, μm2/m3

Figure 3. Indoor SAC Variation over 24-Hours

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3.2 Variation Pattern over a Day

The SAC variation pattern was similar across the different sampling locations, except during combustion activities. Corresponding to the each particular combustion activity, fuel-specific particle emission takes place influencing the concentrations as well as the size ranges. There was difference in the nature of the peaks from one activity to another which was evident through the graphical plots of the concentration as well as the activity log sheet. This corresponds to the source-specific nature of the aerosols generated. After constant low levels of concentrations in the early hours of the day, sharp peaks are seen from around 5 a.m. to 9 a.m. due to the inflow of particles onset by activities and wind flow. Moreover, after around 6 am to 9 am, combustion activities start to take place. These usually include heating water for bath, preparing morning breakfast, and even sweeping.
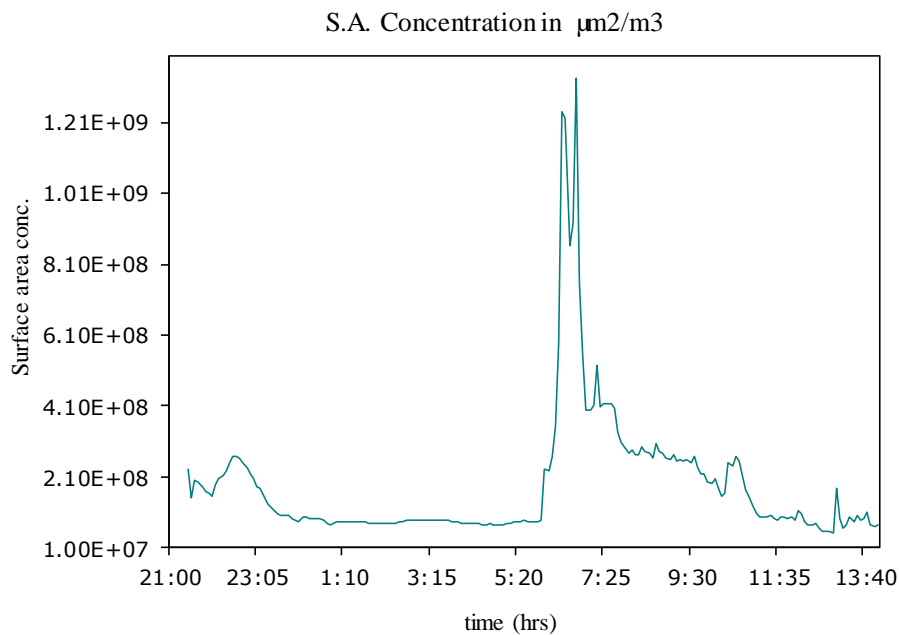

Figure 4. Indoor SAC Variation over 24-Hours

Usually, the afternoon period of 12-4 pm was found to be little calmer durations with lower concentration levels. Concentrations rise to moderate levels from around 5 pm-7 pm and variation pattern is similar to early morning hours, but with little lower concentration levels. This is found to be due to the increase in outdoor activities like movement of vehicles, or due to smaller-duration cooking activities which included preparing tea, snacks, and sometimes dinner also. In the late-night hours, after about 11 pm, there was a steep decrease in concentrations reaching a minimum and remaining constant at that level till around 4-5 am. After 11 pm, family members are taking rest; hence there is no disturbance causing the coarser particles to gradually settle down. No unique difference in variation was found during this period between Kerosene and LPG houses.

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Nighttime average values of concentration during 12 p.m.-4 a.m. are considered as background concentration levels. Since it is no-activity period, there will be fewer disturbances for aerosol movements.

We can refer to results from other studies in this. Study by (Kamens et al., 1991) showed that at midnight, when everyone in the house had gone to sleep, aerosol concentration declined indicating the settling of larger particles. Our study also showed similar results.

The activity log-sheet was helpful to know the cause for unusual variations in concentration levels. The plot shown above (Figure 3 and Figure 4) represents an indoor daily variation with a sudden peak around midnight. The sudden peak is because of the doors being opened when one of the house members came late in the night, which might have caused the wind in-flow, resulting in random movement of the settled particles. It can also be observed from the plot that later on, the concentration levels remained higher than the usual levels throughout the night, with a small gradual decrease in concentration. Study by (Kamens et al., 1991) has showed that the fine particle levels after a peak would remain high for a longer period.

For urban locations, within-day variation was found to be about 2-3 times higher than between-day variation. But for remote locations, not so high difference was found between the two, indicating the influence of lesser sources and activities.

3.3 Concentration Levels of the Metric

Data downloaded from the LQ1-DC after each sampling was 1-min average data of SAC. From 1-min average data, statistical results such as Arithmetic mean, Geometric mean, and Standard Deviation etc. were obtained for all Indoor sites.

Table 1 gives the summary of average concentration, their standard deviation for Indoor measurements. LPG house SAC concentrations are little lower compared to the kerosene households.

One of the kerosene houses sampled was in a slum area, with poor ventilation facility within the house; the influence from neighboring houses also was large, because of too much congestion. In two other kerosene houses, the neighboring house influence on the concentration was large. One kerosene and one LPG house had dusty surrounding environment. Statistical difference was found between the two groups for SAC as the exposure metric. Variance among the households is nearly the same among the two groups.

Table 1. 24-hr Average Data of the Metrics from Indoor Measurements with Kerosene and LPG as the Main Fuel

House ID	Kerosene house	LPG house
	Metric values in each house	Metric values in each house
	Surface area conc. (0.01-2.0) $\mu m^2/m^3$	Surface area conc. (0.01-2.0) $\mu m^2/m^3$
1	1.63E+08	1.49E+08
2	4.30E+08	
6	2.99E+08	3.45E+08
7	7.79E+08	
8		
9		4.31E+08
10	7.76E+08	
11	3.77E+08	1.37E+08
S.D. ($\mu g/m^3$)	**55.34**	**46.31**
Variance (%)	**54.54**	**62**

Standard Error (S.E.) is found out for the two groups using S.D. from Table 1, using the formula discussed earlier. Later, we find that Kerosene (Mean-SE) > LPG (Mean+SE) indicating that the two indoor categories, viz., kerosene and LPG households are statistically different.

Table 2 shows the ratio of concentrations found in two categories of houses is 1.79 indicating that usage of kerosene fuel is leading to higher concentrations than LPG. Thus LPG is a cleaner fuel than kerosene.

Table. 2. Indoor Measurements in Houses: 24-hr A.M. Values of Surface Area Concentration (0.01-2 μm) (n=5) (Background indoor pollution: 36$\pm$16 $\mu g/m^3$)

Kerosene house	LPG house	Kerosene/LPG ratio: range and G.M.
(4.71$\pm$2.54)E+08 $\mu m^2/m^3$	(2.65$\pm$1.46)E+08 $\mu m^2/m^3$	1.76 to 1.82 GM=1.790

Considering both groups of houses in general, for surface area concentration, the ratio of 24-hr mean to background value for each house is found to vary from 1.6 to 3 with an average of 2.5. This shows the influence of the combustion activities which increases the PM concentration to more than two times the normal levels. For instance, in one kerosene house, SAC was found to exceed 2000 $\mu m^2/cm^3$ two times

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in a day. These levels are potentially harmful to the inhabitants who get exposed to them on a daily basis.

The ratio of peak value to 24-hr mean value for each house is found to vary from 4 to 66, with an average of 21.2. This shows the impact of emissions during combustion activities to increase the PM concentrations from normal levels to more than 21 times. This indicates how some combustion activities can create really high dose of short-term particulate exposure to the inhabitants. Ventilation facility in the house influences the concentration levels very much. Results from earlier studies have also shown the dependence of $PM_{2.5}$ exposures on the ventilation levels within the houses (Sarnat et al., 2000). In some cases, the outdoor sources had a major effect on the exposure levels indoors.

4. Conclusion

National Ambient Air Quality standards (NAAQS, 2009) in India stipulate standards for $PM_{2.5}$ at an annual average of 40 μg/m^3 and 24-hr average of 60 μg/m^3. However, there are no standards specified for SAC.

Houses using kerosene fuel for cooking were found to be more (about 1.79 times) polluted with particulate matter than the houses using LPG. Every cooking activity resulted in concentration peaks. Indoor peak values were seen to be about 20 times higher than the 24-hr mean values. Indoor average concentrations exceeded background values by more than two times. In kerosene-using houses, generally poor ventilation facility and congestion was found. Heating water for bath during early morning hours was one major aerosol generating activity which lasted for 2-3 hours. Peaks during heating water for bath in each house were found to far exceed the peaks during the other activities. This indicates the influence of the crude fuels like wood, dung cakes etc. Similar peak was found daily during evening times between 5 p.m. and 7 p.m., mostly due to the increase in outdoor activities, and penetration of outdoor particles into the houses. For kerosene and LPG fuel types, not much difference in daily variation pattern was found.

In the light of health effects associated with aerosol exposure, results of the present study indicate alarmingly high exposure to household women who spend majority of their time indoors doing cooking. There is a risk of high-dose of short-term exposure to the inhabitants. There is the need to ensure that their exposure to particulate matter is well within the permissible limits for a healthy living. May be good ventilation facility within living room and kitchen, cleaner fuel and efficient stoves etc., can lessen the exposure levels to these particulates generated. Also, the use of kerosene fuel, wood and dung cakes etc., will have to be replaced by better fuels like Liquid Petroleum Gas (LPG).

The latest survey released by the National Sample Survey Organization (NSSO, 2012) has revealed that in rural areas, subsidized kerosene was used in less than 1% of kitchens, which relied largely on firewood and chips as the primary source of energy for cooking during 2011-12. More than 67% of rural households in India still depend on firewood or wood chips for cooking. In contrast, around 68% of urban households use LPG for cooking. When it comes to lighting homes in villages, the share of

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kerosene is estimated at 26.5%, with electricity's share estimated at 72.7%. In urban India, 3.2% of households use kerosene for lighting while the share of electricity is 96%.

The encouraging aspect in urban households is that the use of kerosene has declined from more than 23% to 6%. Consumption of kerosene, used primarily for lighting and cooking purposes in rural areas, has dropped by a sharp 21% in 2016-17 from a year ago aided by greater use of cleaner Liquefied Petroleum Gas (LPG) for cooking and coverage of more villages under the rural electrification program, as per data from Petroleum Planning and Analysis Cell, an arm of the oil ministry. In the same period, consumption of LPG jumped by 9.8%. In 2016-2017, there were issued a total 3.25 crore new LPG connections, the highest number of connections given in any year ever. This number included the 2 crore connections given under the "LPG-for poor women" scheme, the Pradhan Mantri Ujjwala Yojana (PMUY). India's fossil fuel consumption trend is suggesting a shift away from inefficient and highly polluting use of hydrocarbons, as a result of efforts to move towards a less-carbon-intensive economy. Recent WHO pollution report mentions this scheme saying "India's PMUY Scheme has provided some 37 million women living below the poverty line with free LPG connections to support them to switch to clean household energy use" (WHO, 2018). Worth mentioning also are some of the non-government organizations such as Confederation of Indian Industry (CII) which has established the Indian Green Building Council (IGBC) and working closely with state governments and central government in promoting green building concepts in India (https://igbc.in/).

The present study has setup a database for SAC metric at two Indoor environments in a medium Indian city and initiated a study of the association of peaks with cooking activities and the variation pattern of the metric. This kind of study may become more satisfactory with the rigorous study of peaks associated with combustion activities, as to the particle size range, density or morphology of particles generated by different sources.

Acknowledgement

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