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Carcinogenic risk from exposure to $PM_{2.5}$ bound polycyclic aromatic hydrocarbons in rural settings

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ABSTRACT

In the study, first-time personal exposure level of polycyclic aromatic hydrocarbons (PAHs) was measured during cooking hours in participants of three different types of kitchen both in the particulate and gaseous phase using traditional and improved cookstoves. Along with that, indoor particulate matter (PM) concentration was also estimated during the cooking hours to examine the impact of intervention in different kitchens. The results of the study clearly revealed that the kitchen characteristics and type of cookstove technology have a significant impact on $PM_{2.5}$, PM_1 and PAHs concentration. Cookstoves intervention has resulted in maximum reduction of PM_1 i.e. 75% in an enclosed kitchen followed by semi-enclosed and open kitchen having 71% and 52%, respectively. In addition, correlation analysis of $PM_{2.5}$ and PM_1 with PAHs showed a strong association ($r^2 = 0.9$), showing the affinity of PAHs to bind to fine range of particles. Health risk assessment was also carried out to assess the PM daily dose and carcinogenic and non-carcinogenic risk due to inhalation of PAHs. The study confirmed the personal concentration of PAHs compounds was significantly high ($p < 0.05$) during use of traditional cookstove compared to improved cookstove among all the three kitchens. Furthermore, to measure the toxicity levels, PAHs concentrations have been converted to benzo[a]pyrene equivalence for calculating cancer and non-cancer effects using toxicity equivalency factors. The overall lifetime carcinogenic risk was the highest 2.5E-03, 6.4E-04 among women who prepared meals in the enclosed kitchen compared to 8.4E-04, 1.3E-04 in semi-enclosed and 2.2E-04, 4.6E-05 in the open kitchen during use of traditional and improved cookstoves, respectively, which exceeded the US EPA standard i.e. 1×10^{-6}. The study underlined the importance of personal monitoring for exposure, and risks-based studies along with the time-activity of user to measure the actual inhalation risk for the participants. These findings indicated that women are exposed to hazardous smoke in the indoor kitchen and are at greater risk of developing cancer, especially in rural areas.

1. Introduction

All over the world, around 3 billion people rely on inefficient source of cooking out of which 2.7 million prepare foods on these cookstoves using solid biomass fuels (SBFs) such as wood, animal dung and crop residue (WHO, 2018; Arora and Jain, 2016). Traditional cookstoves (TCS) are the major source of household air pollution (HAP) that releases a number of toxic air pollutants such as particulate matter (PM), carbon monoxide (CO) and hydrocarbons which include polycyclic aromatic hydrocarbons (PAHs) and volatile organic compounds (VOCs) in the indoor environment (Sharma and Jain, 2019; Arora and Jain, 2015; Arora et al., 2014, 2013; Haritash and Kaushik, 2009). PAHs are ubiquitous organic compounds originating from a variety of energy sources i.e. domestic cooking, open biomass burning, industrial and vehicular emissions etc. (Yury et al., 2018; Zheng et al., 2018; Liu et al., 2017). PAHs are the products of incomplete combustion, and domestic activities contribute around 60% of global emissions of PAHs (Rengarajan et al., 2015). The World Health Organization (WHO, 2018) reported high levels of exposure to $PM_{2.5}$ resulting in heart and respiratory diseases and responsible for lung cancer in human beings. Furthermore, a few studies reported that PAHs are likely to absorb in the fine range of particles ($PM_{2.5}$) that provides a large surface area to organic compounds causing toxicological effects, i.e. oxidative stress, cytotoxicity, genetic mutations, systemic inflammation and cardiovascular diseases (Rabha et al., 2018; Yang et al., 2012). Moreover, long term exposure to PAHs may result in damage to human cell lines, pulmonary tissue damage, and cardiopulmonary mortality (Rengarajan et al., 2015; Lal et al., 2011). Risk assessment associated with inhalation

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Table 1
Details of sampling along with time-activity of the participants among selected households.

Features analyzed	Type of kitchen	Enclosed kitchen		Semi-enclosed kitchen		Open kitchen	
	Household No.	HH1	HH2	HH3	HH4	HH5	HH6
No. of sample collected for TCS and ICS with 3 replicates for two cooking sessions	TCS (n = 3)	6	6	6	6	6	6
	AFD (n = 3)	6	6	6	6	6	6
Total number of samples	PM and gaseous samples: 12 × 2 = 24 samples from each household; Total = 24 × 6 = 144						
Average cooking hours/day (Morning + Evening)	TCS	3.42	3.23	2.40	2.47	3.00	2.45
	AFD	3.05	2.51	2.10	2.05	2.30	2.19
Actual time in front of stove (hours/day)	TCS	3.10	2.52	2.20	2.30	2.35	2.15
	AFD	2.40	2.20	1.45	1.35	2.05	1.45
Time spend in doing other household work along with cooking (minutes)	TCS	32	31	20	17	25	30
	AFD	25	31	25	30	29	34
No. of family members in each household		7	6	6	7	6	5

HH: Household; TCS: Traditional cookstove; AFD: Annapurna forced draft; n = replicates.

of PAHs is often assessed based on the Benzo[a]Pyrene (B[a]P) concentration in air (IARC, 2010). Past research reported that SBF burning in TCS results in higher PAHs emissions (Masih et al., 2010). Nevertheless, in most of the developing nations, use of SBFs in domestic cooking is still dominant. One of the alternatives to this issue is deployment of improved cookstoves (ICS) to reduce the personal exposure of residents by such interventions (Sharma and Jain, 2019). In addition, the study also highlighted the importance of personal sampling, which gives actual concentration of pollutants inhaled (Lin et al., 2016). Thus, aim of the study was to monitor the indoor PM and personal PAHs concentrations, to characterize and quantify the personal exposure to PAHs during cooking hours in the participants, i.e. women cook in the respective households under different kitchen characteristics in pre and post intervention conditions. Further, PM daily dose, PAHs carcinogenic and non-carcinogenic risk has also been calculated among the women (chief cook) based on the $B[a]P_{equiv}$ and HQ respectively, during cooking hours.

2. Material and methods

This section briefly gives an outline of the methodology. Table 1 presents the number of households selected along with their kitchen characteristics and methods used in consort with time-activity of the participants during cooking and the number of samples collected. Details of household selection, indoor and personal sampling of PM and PAHs and their analysis are discussed in following sections.

2.1. Study area and selection of households

The study was conducted in Jagdishpur, Amethi district in the state of Uttar Pradesh, India (details has been presented in Sharma and Jain, 2019). Six households were selected through a pilot survey, based on the kitchen characteristics, type of cookstoves and use of SBFs like wood, dung cake and crop residue for their daily cooking activities and willingness to participate. The survey data showed that 80–90% of the households used only biomass fuel for cooking and other domestic activities. The primary cooks were women and average number of family members was 6–8. Fuel consumption pattern showed that majority of households use fuel wood ~84% for their daily cooking activities. It was observed that along with TCS, around 40% of the households were using ICS (40%) for their daily cooking and nearly 6% were using only ICS, while approx. 54% were dependent on TCS as their primary cookstove. Households were selected to have enclosed kitchen (covered by four walls and have minimal ventilation), semi-enclosed kitchen (covered with a thatched roof) and open kitchen located in the veranda and open to the sky. The selected households were using TCS along with ICS i.e. Annapurna forced draft-TERI SPT-0610. It was a top loading cookstove, made of steel and has an inbuilt fan to enhance the mixing of combustion gases. In each household, cooking was done two times a day i.e. morning between 6:00 a.m. to 9:00 a.m. and evening from 5:30 p.m. to 8:30 p.m. Apart from the information on the household characteristics, participants were also inquired about the fuel consumption and their time-activity pattern presented in Table 1. Thus, this information helps in conducting the personal monitoring in a more efficient way.

2.2. Sampling: Indoor PM and personal PAHs measurements

The PM ($PM_{2.5}$ and PM_1) was monitored using an aerosol spectrometer (Optical Particle Sizer – OPS model – 3330, TSI) that works on a single particle light scattering principle. The instrument provides real-time PM concentrations in 10 size channels starting from 0.3 μm to 10 μm with a user choice sampling time interval (1 min in the present study). The instruments installed in the kitchen area were kept at a distance of ~100 cm away from the cookstove chamber and a height of ~145 cm from the floor, which closely represented the breathing zone of the person involved in cooking task (more detail has been presented in Sharma and Jain, 2019).

The personal monitoring system (PMS) was consisted of battery-operated pumps (model 224-44XR) with a flow rate of one lpm, with one-stage $PM_{2.5}$ inertial impactors (SKC Personal Environmental Monitor, model 200) that capture particles on 37 mm quartz filters (Gelman R2PJ037). Personal PAHs sampling for gaseous phase was carried out using XAD-2 sorbent tubes (8 × 110 mm size, 2-section, 200/400 mg sorbent, with GS ends, WWW separators i.e. glass wool separators between the sorbent layer for the uniform pressure drop and tube cover) connected between the impactor and pump (Masih et al., 2012). Flow rates were measured before and after each sampling period using a calibrated rotameter. The impactor was attached to the collar of the cook near her breathing zone and sampling pump was fitted in a small bag that was attached around the waist during complete cooking cycle as presented in Fig. S1, section A of supplementary information (SI). The cook was informed and trained about the importance of keeping the impactor in proper position. All the samples were properly labelled and wrapped in aluminum foils to store at freezing temperature in deep freezer (-18 °C) to prevent degradation (Chen et al., 2016). Blank samples were also treated in the same way as the test samples. Mean ambient temperature and relative humidity recorded during the monitoring was 35 °C ± 5.8 and 45–80%, respectively. The personal sampling at each household was done for three days which includes two cooking cycles per day i.e. morning and evening with three replicates on each cookstove to get a representative sample. Therefore, around 8 weeks intensive and extensive monitoring was conducted to collect 12 samples from each household (TCS: 6 and ICS: 6) for particulate and gaseous phases simultaneously. Total 144 samples were collected for particulate and gaseous phases in three types of kitchens from six households as shown in Table 1.

2.3. PAHs extraction and analysis

The PAHs concentrations were extracted from the $PM_{2.5}$ collected on the quartz filter and XAD-2 extraction using US EPA method TO-13 A for PAHs analysis (US EPA, 1999). PAHs extraction was carried out using half of the quartz filter further cut into strips and extracted with 30 ml of HPLC grade dichloromethane (DCM) using ultrasonic agitation for 30 min (Orakij et al., 2017). The procedure was repeated 2–3 times to get maximum recovery of PAHs. The extract was evaporated using a rotary evaporator at a temperature between 30 and 40 °C and then finally adjusted to exactly 1 mL by DCM after filtering through membrane filter (PVDF 0.5 mm micro syringe). Similarly, the XAD-2 tube was broken, and content was dissolved in DCM for extraction using ultrasonic agitation and the procedure was repeated two times (60 min) followed by rotary evaporation and filtered through membrane filter (Downward et al., 2014). All field blanks were also extracted in the same way as the test samples following the US EPA method TO-13 A and no PAHs compounds were detected. All glassware was rinsed with DCM before using them for the sample extraction, clean up and storage. The final extract obtained was stored in glass vials that were pre-conditioned in the oven. The extracts were then injected into Gas Chromatography Mass Spectrometry (Shimadzu GC–MS-QP, 2010 Plus model) for analysis (Zheng et al., 2018). The GC–MS was equipped with a capillary column Rtx-5 (dimensions: 0.25 μm film thickness, 0.25 mm internal diameter, and 30 m in length). Injection volume was 1.0 μL, and the pulsed split less time was set at 1 min. For qualitative screening of contaminants, scan mode was carried, and vaporizing temperature was 300 °C and carrier gas was Helium. All the analysis was done in the Advance Instrumentation Research Facility (AIRF), Jawaharlal Nehru University, New Delhi.

2.4. Estimation of PM daily dose and exposure indices

PM inhalation dose was estimated using the general equation for chronic daily intake (CDI) given by United States Environmental Protection Agency (US EPA, 1993). The equation used to determine the dose is as follows:

$$Daily\ dose\ (mg/kg-day) = C\ x\ IR\ x\ ET\ x\ EF\ x\ ED/(BW\ x\ AT) \quad (1)$$

where, *C: $PM_{2.5}$ and PM_1 concentration during cooking hours (mg/m^3).*

IR: inhalation rate: 20 m^3/day
ET: exposure time (hours): Participants actual cooking time (TCS and ICS)
EF: exposure frequency: 350 days/year
ED: exposure duration: Number of years exposed (actual data collected from the participants under study)
BW: body weight (kg): Weight of the person involved in cooking
AT: average time (days)

The PM concentration monitored during cooking was compared to see the impact of intervention under different kitchen characteristics on the daily dose of participants. The IR, EF and AT were taken as constant values from US EPA (2011); whereas, ET, ED and BW were collected from the field while doing survey and indoor monitoring. Thus, the study gives the actual dose of participants involved in cooking in the respective kitchens. In addition, indoor $PM_{2.5}$ and PM_1 concentrations were also compared with 24-h $PM_{2.5}$ concentrations in the ambient air based on health and air quality categories given by US EPA and the Department of Health and Human Services Victoria (EPA Victoria, 2018). In view of that, the present study develops exposure indices of $PM_{2.5}$ and PM_1 under health and air quality categories to understand the complexity of HAP during cooking.

2.4.1. Calculation of B[a]P equivalency

Earlier different methods were followed to measure the toxicity of PAHs and one of the methods is to estimate the $B[a]P_{equiv}$ toxicity using toxicity equivalency factors (TEFs) used in various studies (Liu et al., 2017; Kaushik et al., 2012). The risk relative to B[a]P was calculated by the TEFs for individual PAHs given by Nisbet and LaGoy (1992). $B[a]P_{equiv}$ was calculated by multiplying individual PAH concentration with its equivalent TEFs for all kitchen types. A few studies have used the same approach for $B[a]P_{equiv}$ calculations based on the TEFs (Ramírez et al., 2011; Ohura et al., 2004). Equation (2) was used for calculating the $B[a]P_{equiv}$.

$$\boldsymbol{B[a]Pequiv} = \sum_{i=1}^{n} \boldsymbol{C_i \times TEF_i} \quad (2)$$

whereas, C_i = Concentration of compound *i*.

TEF_i = Toxicity equivalence factor for individual PAHs

In order to quantify the carcinogenic and non-carcinogenic risk from inhalation exposure, an inhalation unit risk (UR) of 6×10^{-4} per μg/m^3 was applied given by Integrated Risk Information System, (US EPA, 2017). Similarly, to quantify HQ, a reference concentration (RfC) for developmental (HQ_D) and reproductive (HQ_R) effect applied was 2×10^{-6} and 3×10^{-6} mg/m^3, respectively, for B[a]P (US EPA, 2017). Moreover, it was also reported that overall confidence while estimating inhalation RfC for non-carcinogenic risk varies from low-to-medium. Thus, carcinogenic risk was calculated by multiplying the $B[a]P_{equiv}$ (from equation (2)) and UR, as given below in equation (3). HQ was calculated by dividing the actual inhalation concentration by RfC (Liu et al., 2015).

$$Carcinogenic\ risk = B[a]Pequiv\,(ng/m^3) \times UR \quad (3)$$

$$HQ = C/RfC \quad (4)$$

whereas, C = Inhalation concentration (mg/m^3) — $B[a]P_{equiv}$

2.5. Statistical analysis

The SPSS version 20 was used to calculate the descriptive statistics, analysis of variance (ANOVA), and Pearson's correlation to test the relationship between PM ($PM_{2.5}$ and PM_1) and PAHs concentrations in all three types of kitchen. Two-way ANOVA was applied to test the differences between means of diverse kitchen characteristics for PM and PAHs concentration for TCS and ICS. Further, Student's t-test was applied to determine the statistical significance ($P < 0.05$) of the differences between the means determined for individual PAHs concentration.

3. Results and discussion

3.1. Indoor PM during cooking hours

The average concentration of $PM_{2.5}$ and PM_1 estimated among three types of kitchen in the selected households has been presented in Table 2. The cooking hours' average $PM_{2.5}$ and PM_1 concentration was found highest for TCS in enclosed kitchen (818 and 756 μg/m^3) followed by the semi-enclosed (455 and 354 μg/m^3) and open (161 and 118 μg/m^3) kitchen, respectively. Intervention has resulted in average reduction in indoor PM concentration in all the kitchens, having highest reduction in enclosed and semi-enclosed kitchen (76% and 71%) for PM_1 followed by $PM_{2.5}$ (54% and 64%), respectively. It was observed that kitchen characteristics, type of cookstoves and its handling were important factors causing variability in indoor concentrations, especially during cooking time. The variability among enclosed and semi-enclosed kitchens were attributed to low ventilation and small size of kitchens that results in accumulation of smoke in the indoor environment. While open kitchen has low concentration and less reduction in $PM_{2.5}$ (34%) and PM_1 (52%) as compared to other two kitchens. This is

Table 2
The mean $PM_{2.5}$ and PM_1 concentration and daily doses estimated under different kitchen categories during pre and post intervention phases.

Type of kitchen	HH No.	$PM_{2.5}$						PM_1					
		TCS (n = 3)		ICS (n = 3)		Reduction		*p*- value	TCS (n = 3)		ICS (n = 3)		Reduction		*p*- value
		C (μg/m^3) Mean ± SD	DD (mg/kg-day)	C (μg/m^3) Mean ± SD	DD (mg/kg-day)	C %	DD %		C (μg/m^3) Mean ± SD	DD (mg/kg-day)	C (μg/m^3) Mean ± SD	DD (mg/kg-day)	C %	DD %	
Enclosed kitchen	HH1	656 ± 713	3.24E-02	219 ± 180	9.66E-03	67	70	0.01	620 ± 696	3.06E-02	158 ± 116	6.97E-03	75	77	0.01
	HH2	980 ± 1166	3.56E-02	567 ± 494	1.60E-02	42	55	0.03	892 ± 730	3.24E-02	202 ± 162	5.48E-03	77	83	0.00
Semi-enclosed kitchen	HH3	400 ± 402	8.92E-03	169 ± 105	3.29E-03	58	63	0.00	347 ± 363	7.74E-03	99 ± 95	1.85E-03	71	77	0.01
	HH4	509 ± 539	1.42E-02	153 ± 59	3.54E-03	70	75	0.01	361 ± 392	1.00E-02	108 ± 105	2.49E-03	70	75	0.01
Open kitchen	HH5	135 ± 95	5.87E-03	89 ± 41	3.01E-03	34	49	0.00	75 ± 56	3.25E-03	34 ± 23	1.22E-03	55	64	0.01
	HH6	187 ± 95	6.11E-03	122 ± 75	3.57E-03	35	42	0.01	160 ± 142	5.24E-03	81 ± 72	2.38E-03	49	54	0.01

HH: Household; HH No.: Household number; TCS: Traditional cookstoves; ICS: Improved cookstoves; C: Concentration; DD: Daily dose; SD: Standard deviation; n = replicates; *p*-value (using Student's t-test).

attributed to the impact of ambient environment in the kitchen area concentration of pollutants, as open kitchen has direct influence of surrounding environment (Sharma and Jain, 2019). Similarly, Deepthi et al. (2019) also reported higher indoor $PM_{2.5}$ and PM_1 concentrations in enclosed kitchen (278 and 176 μg/m^3) as compared to outdoor kitchen (65 and 49 μg/m^3). Moreover, dusting was other major activity done before and after cooking sessions contributing significantly to PM concentrations and results into overlapping with peaks generated during cooking. The indoor cooking concentration was tested using two-way ANOVA to compare the performance of TCS with ICS among three kitchen types. Since the p-values for $PM_{2.5}$ and PM_1 are < 0.05 (shown in Table 2), null hypothesis was rejected, which showed that there was a significant difference between the indoor PM concentrations for TCS and ICS among all the kitchens ($p = 0.001$ and 0.0002).

3.2. Personal inhalation of PAHs

The personal inhalation concentration levels of PAHs were estimated for TCS and ICS among three selected kitchen types and summarized in Fig. 1. The average PAHs concentration and percentage distributions in three kitchens are shown in Fig. 1(a) and (b), respectively. In case of TCS, the average personal total PAHs concentration was 55371, 32092 and 17239 ng/m^3 in enclosed, semi-enclosed and open kitchens, respectively, as shown in Fig. 1(a). However, in case of ICS, total PAHs concentration was 13599, 4518 and 1764 ng/m^3, respectively. It is important to note that intervention of improved cookstove resulted in a significant reduction of total PAHs concentrations by 75% (p value = 0.004), 86% (p = 0.01) and 90% (p value = 0.02) in case of enclosed, semi-enclosed and open kitchens, respectively. Further, it is worth noting that kitchen characteristics also have a significant impact on personal exposure of the cook. In the present study, a reduction in PAHs concentration ranges from 42% (enclosed vs. semi-enclosed); 69% (enclosed vs. open); 46% (semi-enclosed vs. open) in case of TCS; while, 67% (enclosed vs. semi-enclosed); 87% (enclosed vs. open); 61% (semi-enclosed vs. open) in case of ICS. Downward et al. (2014) also reported that particulate-bound PAH concentration was 4–10 times lower in more ventilated homes compared to enclosed or unventilated homes, which are in line with the results of present study. In our study, both lower and higher molecular weight PAHs compounds were identified in the personal samples as reported in other studies during biomass combustion (Ingale et al., 2011).

Furthermore, the average personal PAHs concentrations in particulate and gaseous phases are shown in Fig. 1(c), (d) and (e) for enclosed, semi-enclosed and open kitchen, respectively. In an enclosed kitchen for TCS, PAHs concentrations in particulate and gaseous phase was 25701 ng/m^3 and 29670 ng/m^3 respectively. However, for ICS, it was 6139 and 7469 ng/m^3, respectively, which resulted in a reduction of ~75% in personal PAHs concentrations both in particulate and gaseous phase as compared to TCS. Similarly, the total PAHs concentrations in semi-enclosed kitchen was maximum during TCS use i.e., 19546 ng/m^3 and 12546 ng/m^3 compared to ICS 2317 ng/m^3 and 2201 ng/m^3 in gaseous and particulate phases, respectively. The results of the study were comparable to the previous studies conducted for personal as well as indoor concentration of PAHs (Tiwari et al., 2016; Ingale et al., 2011). Bhargava et al. (2004) also reported that PAHs concentration varied with type of fuel used and high concentration was reported in case of dung cake (16850 ng/m^3) compared to wood (9170 ng/m^3) in breathing zone. Lisouza et al. (2011) also reported that kitchens having poorly ventilated thatched roof was a major source of particulate born PAHs, similar to the present study. However, in an open kitchen only seven individual PAHs were identified out of 16 PAH-mix, the total PAHs for TCS was highest in gaseous phase, and pyrene was dominant in both phases. It is attributed by impact of ambient environment, which resulted in significantly lower PAHs concentrations in the breathing zone of open kitchen compared to enclosed and semi-enclosed kitchens.

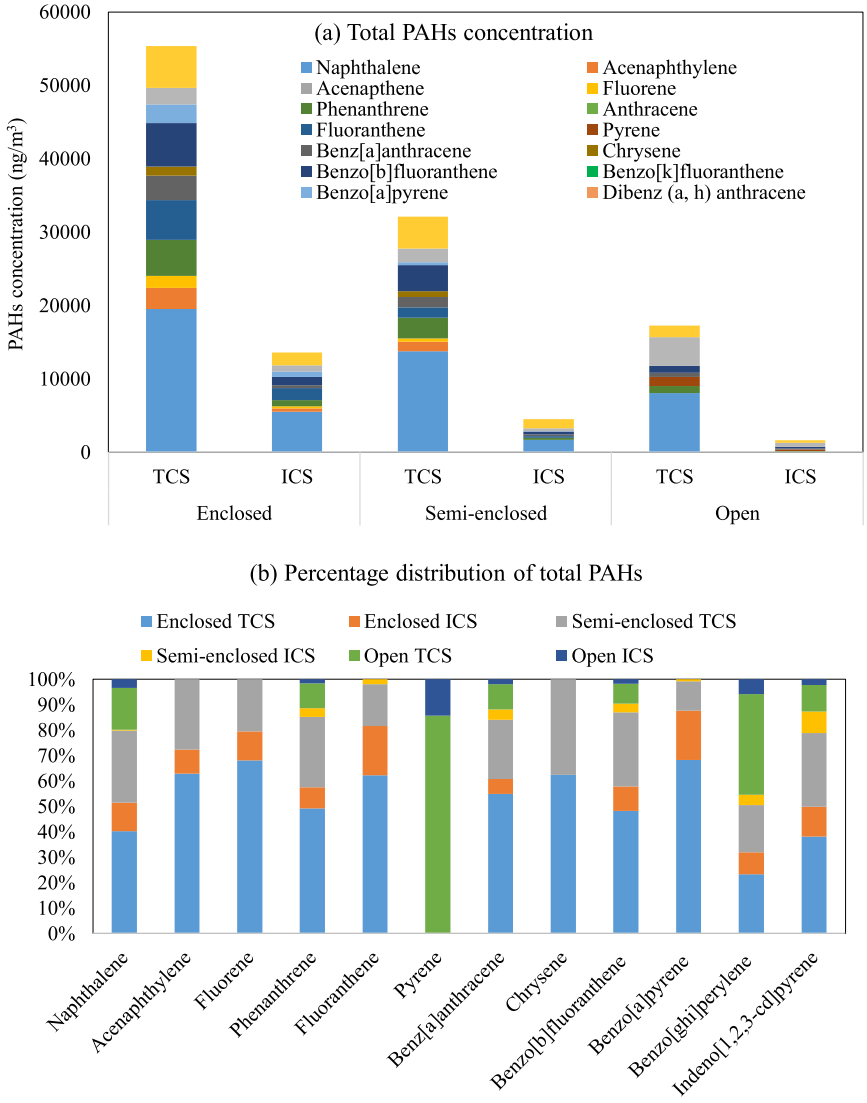

Fig. 1. (a) Average personal concentration of $PM_{2.5}$-bound total PAHs in particulate and gaseous phase measured during cooking hours. The stacked bars are showing total PAHs concentration having statistically significant difference ($p < 0.05$) between TCS and ICS. (b) Percentage distribution of PAHs classified as enclosed, semi-enclosed and open kitchen for TCS and ICS. It presents the distribution of 12 individual PAHs identified among three kitchens. (c-e): Profile of personal $PM_{2.5}$-bound average PAHs concentrations identified in particulate and gaseous phases while using TCS and ICS among three kitchens i.e. (c) Enclosed, (d) Semi-enclosed and (e) Open, respectively. The stacked bars represents the individual concentration of PAHs having significant differences ($p < 0.05$) among three kitchen types. The legend of figure (c-e) shows the list of 16 PAHs representing a compound through different color.

Moreover, among individual concentration of PAHs the most common $PM_{2.5}$ bound PAHs found in particulate phase were 5–6 ring compounds like, B[a]P, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene and benzo[b]fluoranthene as also reported by Haritash and Kaushik (2011) in the residential area. Moreover, B[a]P has been recognized as strong carcinogen (EU, 2005), its concentration in the enclosed kitchen was 1732 and 463 ng/m^3 in particulate and 796 and 259 ng/m^3 in gaseous phase during TCS and ICS, respectively. Further, B[a]P concentrations were also reported by Chen et al. (2017) in rural China (308 ng/m^3) and Bhargava et al. (2004) in rural India (730 ng/m^3) from wood combustion in the kitchens, which is very close to gaseous phase PAHs concentration in the present study. Additionally, the personal concentration of B[a]P in semi-enclosed kitchen during TCS and ICS was 425 ng/m^3 and 31 ng/m^3, respectively which was lower to previously reported concentration i.e., 700 ng/m^3 in similar conditions by Bhargava et al. (2004). As discussed, high variability in PAHs concentrations in breathing zone was not only dependent on cookstove performance but also influenced by time-activity of the user (Liu and Zhu, 2001). Moreover, the impact of user's cookstoves operation and handling and cooking time have significant impact on variability of PAHs as observed in the present study while recording their diverse activities. For e.g., the cook took almost 30% less time to prepare the meals while using ICS compared to TCS. Moreover, it is important to highlight that the cook spent 10–20% less time directly in front of the cookstove compared with actual cooking hours as presented in time-activity Table 1. Orakij et al. (2017) reported that time spent in cooking increases the inhalation exposure to PAHs. Though, the pilot study aimed to measure the personal inhalation exposure during pre and post intervention phases, but observations based on time-activity of users gives a new insight in terms of actual inhalation exposure of participants.

3.3. Correlation between PAHs and PM

The personal PAHs and indoor $PM_{2.5}$ and PM_1 concentrations showed a positive and strong (R^2 = 0.84 to 0.95) correlation amongst

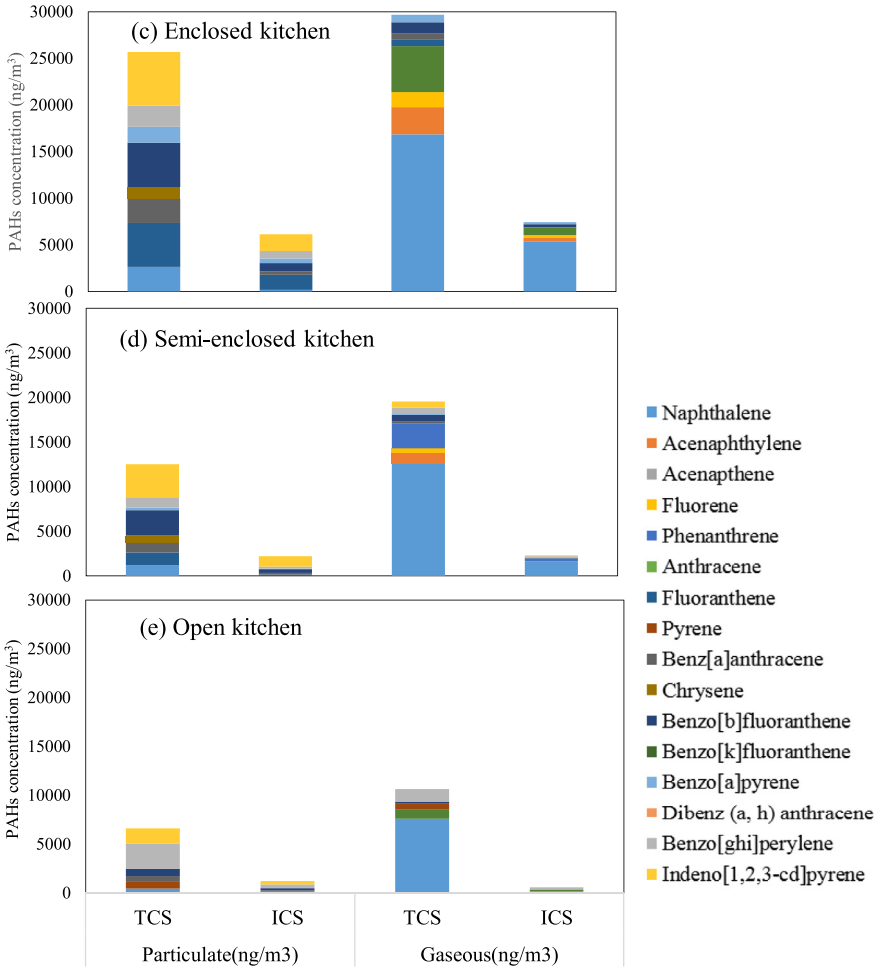

Fig. 1. (*continued*)

all associations that showed PAHs are bound to fine PM i.e. $PM_{2.5}$ and PM_1 as presented in Fig. 2(a–d) and details are also shown in Table S1 (SI, Section B). Similarly, Yang et al. (2012) also reported strong correlation between $PM_{2.5}$ and PAHs as compared to PM_{10} and highlighted that PAHs are bound to $PM_{2.5}$ attributed to small size of particles that provides large surface area to absorb organic compounds. However, in present study because of non-availability of PM_1 impactor we have estimated the PAHs associated with $PM_{2.5}$ only. However, fine and ultra-fine range of particles needs to be analyzed so that the toxicity level can be assessed more comprehensively in health-based studies.

3.4. PM daily dose and exposure indices

Table 2 presents the average daily doses of $PM_{2.5}$ and PM_1 concentrations exposed during cooking sessions under three different types of kitchens. The average dose estimated in the enclosed kitchen during TCS was 3.40E-02 and 3.15E-02 mg/kg-day, followed by semi-enclosed (1.00E-02 and 7.72E-03 mg/kg-day) and open kitchen (5.52E-03 and 3.99E-03 mg/kg-day) for $PM_{2.5}$ and PM_1, respectively. It is important to highlight that the average inhalation dose during TCS was ~3–4 times higher as compared to ICS and become intensified due to the poor source of ventilation in the kitchen area. However, the intervention has resulted in an average reduction of 40–80% among different kitchens in $PM_{2.5}$ and PM_1, respectively. Likewise, Deepthi et al. (2019) reported that indoor doses of $PM_{2.5}$ and PM_1 were 9–10 times higher as compared to outdoor kitchens, attributed to poor ventilation and size of kitchen. Consequently, the study clearly indicates a high dose in a poorly ventilated kitchen which aggravates the health risk of women, thus putting them to very hazardous health category as presented in Fig. 3.

The health categories illustrate the range of $PM_{2.5}$ concentrations from low to 'extremely hazardous' in terms of exposure indices. Fig. 3(a) and (b) showed the comparison of ambient 24-h $PM_{2.5}$ concentrations with indoor $PM_{2.5}$ and PM_1 concentrations during cooking hours. $PM_{2.5}$ concentrations in the enclosed kitchen lie in the 'hazardous' category for both TCS and ICS. While, in case of semi-enclosed and open kitchen, $PM_{2.5}$ concentrations were in 'hazardous' and 'unhealthy' category for TCS, but post intervention concentration falls in 'unhealthy' category. Therefore, intervention of ICS would be beneficial, to a certain extent, if implemented and handled properly. In addition, the air quality category also shows the condition was very poor for indoor $PM_{2.5}$ concentrations compared with both 24-h and 1-h $PM_{2.5}$ concentrations given by EPA Victoria. Similarly, Matawle et al. (2017) also reported that 24-h indoor $PM_{2.5}$ concentrations were several times higher than the National Ambient Air Quality Standard (NAAQS) of India i.e. 60 μg/m^3. Therefore, the study confirms that use of TCS exposes the cook to very hazardous smoke, and this concentration increases to be several folds in an enclosed kitchen. The results clearly show that there is a high level of health risk associated with HAP, which should be of prime concern. Therefore, there is a dire need to have IAQ regulatory standards to understand the complexity of type, toxicity and variability in indoor pollutants.

3.5. Carcinogenic and non-carcinogenic risk assessment

Fig. 4 showed the carcinogenic risk estimated for individual exposed

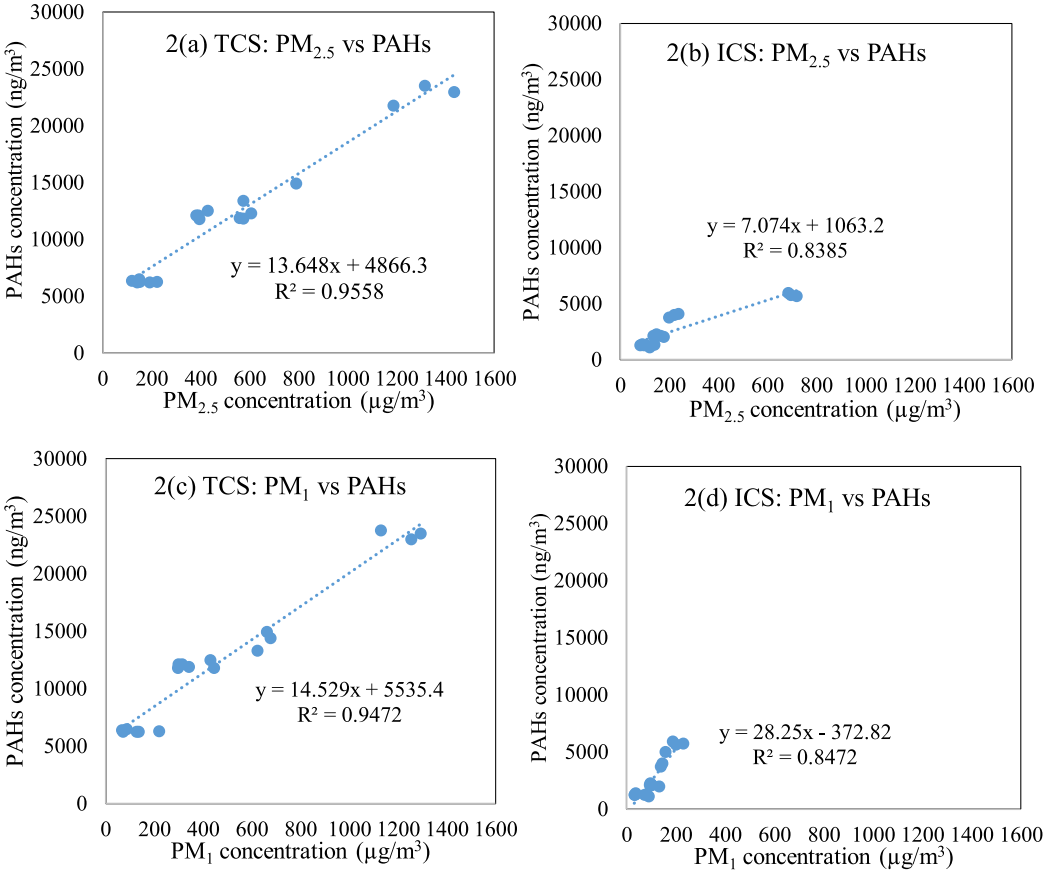

Fig. 2. Correlation analysis between $PM_{2.5}$ and PAHs concentration: (a) & (b) shows association between indoor $PM_{2.5}$ with personal PAHs; (c) & (d) association of indoor PM_1 with personal PAHs concentration in the kitchen area during cooking hours for TCS and ICS, respectively.

to personal PAHs concentrations among three types of kitchen while using TCS and ICS cookstoves (details are shown in Table S2, Section B of SI). The overall life time carcinogenic risk estimated was highest, i.e., 2.5E-03, 6.4E-04 in enclosed compared to 8.4E-04, 1.3E-04 in semi-enclosed and 2.2E-04, 4.6E-05 in open kitchens during use of TCS and ICS, respectively. It is important to note that exposure to B[a]P or its equivalence concentration could be reduced from 66% to 93% by replacing the TCS with ICS in the rural areas. Similar recommendations have been made by Chen et al. (2017) for rural China. In present study, total PAH concentrations in breathing zone clearly showed that carcinogenic risk estimated in the studied area through inhalation of PAHs in particulate and gaseous phases exceeded the US EPA standard i.e. 1×10^{-6}. It is clearly reflected while considering the individual toxicity of the PAHs, the compound that has the highest risk was B[a]P in both enclosed and semi-enclosed kitchen and benzo[ghi]perylene in nearly all kitchens. According to US EPA, it is a possible human carcinogen and contributed nearly 50% of the total predictable risk in case of participants using SBFs during cooking (IARC, 2010). Similar findings have been reported in the past highlights the high lifetime lung cancer risk in smoking homes (Rabha et al., 2018; Castro et al., 2011).

Further, Table 3 summaries the $B[a]P_{equiv}$ and non-carcinogenic risk (HQ_D and HQ_R) estimated for individual exposed to personal PAHs concentrations among three types of kitchen. The non-carcinogenic risk estimated in enclosed kitchen showed the highest risk in terms of HQ_D and HQ_R during use of TCS (2045, 1365) as compared to ICS (533, 356), followed by semi-enclosed (HQ_D = 702, HQ_R = 468, for TCS and HQ_D = 114, HQ_R = 76, for ICS) and open kitchen (HQ_D = 181, HQ_R = 120, for TCS and HQ_D = 38, HQ_R = 25, for ICS). The individual HQ for B[a]P and other high molecular weight compounds exceeds unity and showed high HQ_D and HQ_R risk in participants of all kitchens. However, as stated by US EPA, non-carcinogenic risk due to B[a]P varied low-to-medium confidence, but values estimated in case of enclosed and semi-enclosed kitchens using both the cookstove technology are very much alarming showing high probability of toxicity in terms of developmental and reproductive effects. Similarly, high HQ was also reported for respiratory and cardiovascular effect by Piersanti et al. (2018) due to exposure to PAHs concentration. Generally, the results would help in understanding the situation, where merely by increasing the ventilation in kitchen area can decrease the resultant health risk. In such conditions, schemes like Pradhan Mantri Ujjwala Yojana (PMUY) could play a very important role by providing the liquid petroleum gas (LPG) and would help in reducing the burden of cancer risk and other associated diseases.

3.6. Conclusion

The present work underlined the impact of kitchen characteristics and cookstove technology on indoor and personal exposure of the participants to PM and PAHs concentration in rural areas. The study suggested that women in rural areas who use SBFs were exposed to high indoor $PM_{2.5}$ and PM_1 and associated PAHs concentration thereby increasing possibilities of developing cancer and other chronic health impacts. Health risk assessment suggested the overall life time carcinogenic risk was the highest in case of women cooking meals in enclosed kitchens compared to semi-enclosed and open kitchens. The current research also advocated that it is necessary to have appropriate ventilation in the kitchen areas in order to reduce health risk from exposure to toxic compounds. Thus, it is worth noting that the IAQ regulatory guidelines should be established in India, which give details of type of pollutants and their indoor concentration levels and

A. Health category	24-h $PM_{2.5}$ (μg/m^3)	Cooking hours average $PM_{2.5}$ (μg/m^3) concentrations in traditional and improved cookstoves											
	Ambient air	Enclosed kitchen				Semi-enclosed kitchen				Open kitchen			
		$PM_{2.5}$		PM_1		$PM_{2.5}$		PM_1		$PM_{2.5}$		PM_1	
		TCS	ICS	TCS	ICS	TCS	ICS	TCS	ICS	TCS	ICS	TCS	ICS
Low	0–8.9												
Moderate	9.0–25.9												
Unhealthy-sensitive	26.0–39.9												
Unhealthy all	40.0–106.9								101		106		59
Very unhealthy	107.0–177.9						161			161		118	
Hazardous (high)	180-249.9				180								
Hazardous (extreme)	≥ 250	818	393	756		455		354					

B. Air quality category	24-h $PM_{2.5}$ (μg/m^3)	1-h $PM_{2.5}$ (μg/m^3)	Cooking hours $PM_{2.5}$		
			Enclosed kitchen	Semi-enclosed kitchen	Open kitchen
Very good	0–8.2	0–13.1			
Good	8.3–16.4	13.2–26.3			
Fair	16.5–25.0	26.4–39.9			
Poor	25.1–37.4	40–59.9			**ICS**
Very poor	≥ 37.5	≥ 60	**TCS and ICS**	**TCS and ICS**	**TCS**

Fig. 3. (A) Comparison of 24-h $PM_{2.5}$ concentrations in the ambient air with indoor $PM_{2.5}$ and PM_1 concentrations on the basis of health categories as given by EPA; (B) Comparison of $PM_{2.5}$ air quality categories for 24-h and 1-h $PM_{2.5}$ with $PM_{2.5}$ measured during cooking hours as given by EPA (Source: EPA Victoria, 2018).

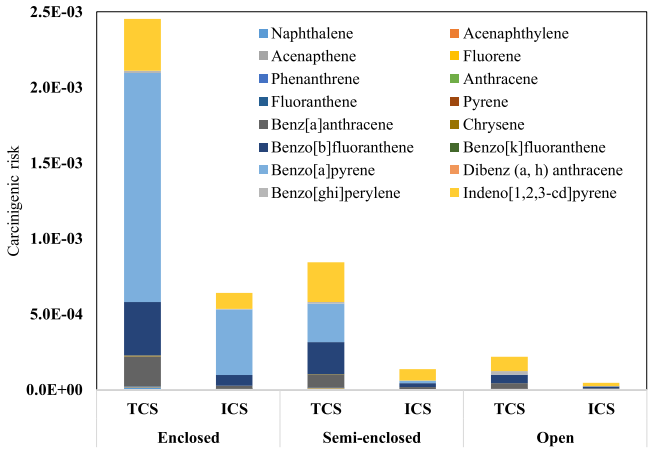

Fig. 4. Estimated carcinogenic risk for individuals exposed among three kitchens while using TCS and ICS. The stacked bars present the risk due to exposure to individual PAHs compounds. The Benzo[a]pyrene is causing the highest risk in enclosed and semi-enclosed kitchens.

subsequent health impacts. Therefore, this study provides significant information that can be used by various stakeholder's and policymakers while making regional priorities for interventions and mitigations to be taken to use more efficient and clean devices for cooking along with access to clean fuel. Along with that, awareness should be made in rural communities about the importance of ventilation in kitchens and households by making some structural changes in the households, so that ventilation could be increased in kitchens. This would further help in significantly reducing the exposure of women and children and hence health impacts. Overall, the findings of the study are useful for better understanding about personal exposure to toxic air pollutants for improvement in cookstove designs, ventilation conditions in the kitchens and relevant policy and indoor standards formulations. Limitation of the study is that gravimetric measurement of $PM_{2.5}$ quartz filters were not made due to the unavailability of sensitive mass balance on field. Thus, future studies should do real-time time measurements of $PM_{2.5}$ to assess the toxicity under various set of conditions.

Author contribution statement

Deepti Sharma: Methodology, field measurements and data analysis, review and editing. **Suresh Jain**: Research design, Indoor air quality monitoring guidelines, field measurements, data analysis and review and editing.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://

Table 3
Average B[a]Pequiv (ng/m^3) and non-carcinogenic risk (HQ) among participants of three kitchens.

16 PAHs compounds	Enclosed Kitchen				Semi-enclosed Kitchen				Open Kitchen			
	TCS (n = 3)				ICS (n = 3)				TCS (n = 3)			
	BaP equiv	HQ_D	HQ_R	BaP equiv	HQ_D	HQ_R	BaP equiv	HQ_D	HQ_R	BaP equiv	HQ_D	HQ_R	BaP equiv	HQ_D	HQ_R
Naphthalene	0.020	9.8	6.5	0.006	2.8	1.8	0.014	6.9	4.6	0.0001	0.83	0.55	0.008	4.0	2.7	0.0017	0.1	0.0
Acenaphthylene	0.003	1.4	1.0	0.000	0.2	0.1	0.0013	0.6	0.4	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Acenapthene	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Fluorene	0.002	0.8	0.5	0.000	0.1	0.1	0.0005	0.2	0.2	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Phenanthrene	0.005	2.5	1.6	0.001	0.4	0.3	0.003	1.4	0.9	0.0003	0.2	0.1	0.001	0.5	0.3	0.0002	0.1	0.1
Anthracene	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Fluoranthene	0.005	2.7	1.8	0.002	0.8	0.6	0.0014	0.7	0.5	0.0002	0.1	0.1	0.000	0.0	0.0	0.000	0.0	0.0
Pyrene	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.001	0.6	0.4	0.0002	0.1	0.1
Benz[a]anthracene	0.329	164.7	109.8	0.035	17.5	11.6	0.140	70.1	46.7	0.025	12.3	8.2	0.059	29.8	19.8	0.012	5.9	3.9
Chrysene	0.013	6.4	4.2	0.000	0.0	0.0	0.008	3.8	2.6	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Benzo[b]fluoranthene	0.589	294.3	196.2	0.117	58.4	38.9	0.356	178.0	118.6	0.043	21.4	14.2	0.095	47.6	31.7	0.022	10.9	7.3
Benzo[k]fluoranthene	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Benzo[a]pyrene	2.528	1264.0	842.7	0.721	360.5	240.3	0.425	212.7	141.8	0.031	15.6	10.4	0.000	0.0	0.0	0.000	0.0	0.0
Dibenz(a, h) anthracene	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0	0.000	0.0	0.0
Benzo[ghi]perylene	0.023	11.4	7.6	0.008	4.2	2.8	0.0182	9.1	6.1	0.0040	2.0	1.3	0.039	19.3	12.9	0.006	2.9	1.9
Indeno[1,2,3-cd]pyrene	0.573	286.7	191.1	0.177	88.5	59.0	0.4369	218.5	145.6	0.1255	62.8	41.8	0.158	78.8	52.6	0.035	17.4	11.6

TCS: Traditional cookstoves; ICS: Improved cookstoves; BaP_{equiv} (ng/m^3): Benzo[a]Pyrene equivalent; HQ: Hazard quotient; HQ_D: Hazard quotient developmental; HQ_R: Hazard quotient reproductive; n = replicates.

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