Analysis of gaseous polycyclic aromatic hydrocarbon emissions from cooking devices in selected rural and urban kitchens in Bomet and Narok Counties of Kenya

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Abstract

Traditional combustion devices and fuels such as charcoal, wood and biomass, are widely utilised in rural and urban households in Africa. Incomplete combustion can generate air pollutants which are of human toxicological importance, including polycyclic aromatic hydrocarbons (PAHs). In this study, portable multi-channel polydimethylsiloxane rubber traps were used to sample gas phase emissions from cooking devices used in urban and rural households in Bomet and Narok Counties of Kenya. The results showed a wide range of total PAH concentrations in samples collected (0.82 – 173.69 µg/m^3), which could be attributed to the differences in fuel type, combustion device, climate, and nature of households. Wood combustion using the 3-stone device had the highest average total PAH concentration of ~71 µg/m^3. Narok had higher indoor total gas phase PAH concentrations averaging 35.88 µg/m^3 in urban and 70.84 µg/m^3 in rural households, compared to Bomet County (2.91 µg/m^3 in urban and 9.09 µg/m^3 in rural households). Ambient total gas phase PAH concentrations were more similar and ranged between 1.26 – 6.28 µg/m^3 (Narok) and 2.44 – 6.30 µg/m^3 (Bomet). Although the 3-stone device and burning of wood (especially wet wood) accounts for higher PAH emissions, toxic equivalence quotient (TEQ) values suggest that the jiko stove with locally made charcoal as fuel, has the highest TEQ value (9.87 µg/m^3) and may present more health hazards due to release of higher concentrations of high molecular weight PAHs. Determination of the various levels of PAH produced by these cooking devices and fuels is critical to public health and sustainable pollution mitigation.

Keywords: Polycyclic aromatic hydrocarbon; combustion device, household combustion, domestic air quality.

1. Introduction

A vast portion of the population, particularly in developing countries, rely on solid fuels like wood, charcoal, dung, crop wastes, and traditional stoves for heating and cooking (Bonjour et al., 2013, Johansson et al., 2012). This is a common challenge in African countries where over 600 million people still rely on traditional sources of energy to meet their basic energy needs (Makonese et al., 2018, WHO, 2016). The challenge with traditional energy sources is the emission of potentially harmful toxic compounds which can pose serious human health effects through inhalation (Yury et al., 2018). These emissions can have a negative impact on indoor air quality, which is a vital determinant of global health as humans spend up to 90% of their time indoors (Klepeis et al., 2001). Studies conducted by the Global Burden of Disease established that approximately 3.5 million premature deaths worldwide and various health issues (e.g. cancer and cardiovascular diseases) can be associated with exposure to smoke from households (Patelarou and Kelly, 2014, Suter et al., 2018).

Emissions from household combustion devices can consist of various organic aerosols, the detailed analysis of which may require the use of pollution markers as surrogates for the pollutant species. Polycyclic aromatic hydrocarbons (PAHs) are a common class of combustion products and have received global interest as markers for assessing indoor air pollution (Shen et al., 2013a, Shen et al., 2017, Riva et al., 2011, Chen et al., 2016).

PAHs are a class of semi-volatile organic compounds with two or more fused benzene rings in different configurations. These compounds are of toxicological interest due to their potential mutagenicity and carcinogenicity (Boström et al., 2002, Umbuzeiro et al., 2008). Their occurrence in the air is mainly as a result of pyrolysis or incomplete combustion of organic matter including wood, charcoal, coke, gas, and diesel. Besides these anthropogenic sources, other natural sources of PAHs include forest fires and volcanic eruptions. About 60% of the 16

US EPA priority PAHs are associated with solid fuel combustion (Shen et al., 2013b). In countries like Finland, Chile, and the United States, the 16 US EPA priority PAHs arising from residential wood combustion constitute 78, 72, and 46% of the national PAH emission totals, respectively (Shen et al., 2013b, Shen et al., 2017).

One of the possible challenges for the lack of widespread air monitoring of PAHs is the complex and expensive sampling and extraction techniques typically required. Multi-channel polydimethylsiloxane (PDMS) rubber traps have been successfully used by our group as sorbents for sampling gaseous PAHs in different studies (Forbes et al., 2013, Forbes and Rohwer, 2015, Geldenhuys et al., 2015). The versatility of these simple sampling devices for airborne PAHs has been demonstrated in various applications including sugarcane burning emissions, tunnel air pollution studies, household fire emissions, and diesel emissions from underground mining (Forbes et al., 2013, Geldenhuys et al., 2015, Forbes and Rohwer, 2009, Munyeza et al., 2020).

Review of studies from African countries suggests that there is still limited data on the occurrence of atmospheric PAHs and their associated health effects (Munyeza et al., 2019, Kalisa et al., 2019). This is of great concern, as most developing African countries still rely on solid organic matter (wood, charcoal, etc) as fuel sources. For example, about 85% of households in Kenya mainly use wood as a source of fuel in cooking devices under poorly ventilated conditions (Lisouza et al., 2011, Rahnema et al., 2017, Osano et al., 2020). While there have been studies conducted on the general use and performance of cooking devices in Kenya (Adkins et al., 2010, Lozier et al., 2016, Tigabu, 2017, Pilishvili et al., 2016, Osano et al., 2020), there has been limited reporting on the quantification of PAHs that are emitted by these devices (Gachanja and Worsfold, 1993, Lisouza et al., 2011).

The study by Gachanja and Worsfold (1993) looked at particulate-bound and gaseous PAHs from charcoal stoves that are commonly used in Kenya, specifically ceramic-lined and traditional metal stoves. They found that the ceramic stoves produced significantly lower PAH emissions compared to the traditional counterpart which were 33% higher. On the other hand, the study by Lisouza et al. (2011) focused on PAHs in soot emissions from traditional thatched rural households in Western Kenya, and did not take into consideration gas phase PAH concentrations. While these studies provide some useful insights on the PAH levels, they are limited in that (i) they did not consider PAH levels in the breathing zones of those tending the combustion device in urban and household kitchens, (ii) they did not study the ambient concentrations of PAHs which are importance for human health assessments, and (iii) they did not study PAHs in the gas phase, but rather focused solely on particulate PAH concentrations. These gaps were addressed in a study conducted by our research group, which focused on households in coastal Counties of Kenya (Munyeza et al., 2020).

The objective of the present study was to expand on the previous campaign in characterizing and quantifying indoor PAHs levels from cooking devices in both rural and urban households in selected inland Counties of Kenya, namely Bomet and Narok. This extension from our previous study was important in order to take into consideration variations in fuel availability, cultural practices, climate, altitude, and different home dwellings. PDMS rubber traps were again utilized as simple and cost-effective samplers for gas phase PAHs and these were subsequently extracted using an in-house developed plunger-assisted solvent extraction (PASE) technique, followed by analysis with gas chromatography-mass spectrometry (GC-MS). This study will provide useful insights into the possible factors that can influence indoor PAH levels emitted from cooking devices. Factors such as the population density, type of dwelling, ventilation, geographical location and related climate, source of fuel, and type of combustion device for each of the sampling areas were explored. Such information can serve as a basis for improving household energy usage in order to mitigate the potentially harmful PAH emissions that combustion devices generate.

2. Materials and methods

2.1 Sample collection

The air sampling campaign was conducted in October 2019, at the various sites detailed in **Table 1**. Sampling was conducted in two Counties in south-western Kenya, namely Bomet and Narok, as shown in **Figure 1.** The population, land area and population density of the two Counties are presented in **Table S1** of the Supplementary Information. For each study area, samples were taken from cooking devices in urban and rural dwellings, and ambient samples were also taken in each area.

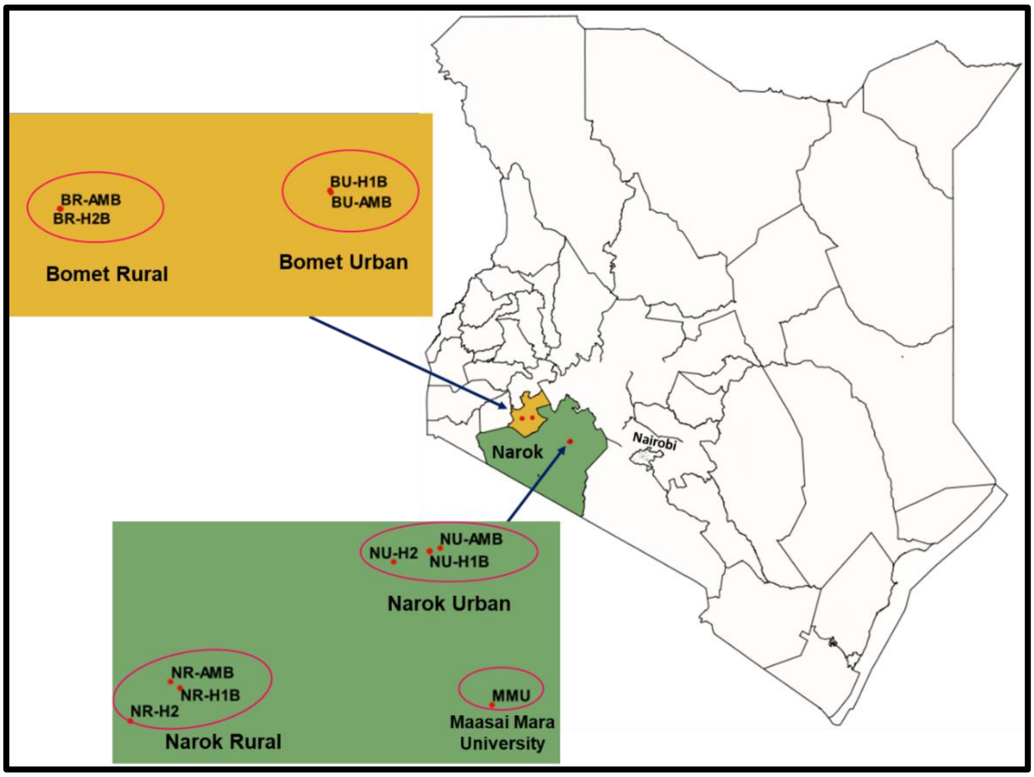


Figure 1 *Map of Kenya showing the sampling locations in Bomet and Narok.*

2.2 Air sampling

The traps used for air sampling were pre-conditioned in an off-line Gerstel™ TC 2 Tube conditioner (Chemetrix, Midrand, South Africa) using a hydrogen gas flow of 100 $mL.min^{-1}$. Field samples of combustion emissions were taken using a PDMS trap as shown in **Figure 2** (consisting of 22 parallel PDMS tubes of 0.3 mm i.d. compactly arranged in a 178 mm long glass tube) coupled to a portable GilAir® Plus sampling pump *(*Sensidyne® Industrial Health and Safety Instrument, Florida, USA) which was operated at a flow rate of 500 mL min^{-1} for 10 min (total volume of air sampled per trap was ~ 5 L). Throughout the sampling campaign, the PDMS traps were positioned at a consistent distance of 65 cm from the stove and 36 cm above the ground (**Figure 3**). Duplicate samples were taken in some cases (**Table 1**).

Furthermore, in all cases, the same aluminium cooking pot containing one litre of water was used and the water was heated to boiling point before sampling commenced. For each sampling point, the sampling position, type of fuel, combustion device, and type of dwelling were noted and are presented in **Table 1**. The majority of the households used either wood or charcoal as a source of fuel, and the common stoves were the jiko, the 3-stone or improved 3-stone stove, and the kerosene stove. This is consistent with an initial survey study that was conducted by our group (Osano et al., 2020).

Indoor ambient conditions (temperature and altitude) were measured using a Kestrel 4500 portable weather station (Kestrel Weather and Environmental Meters, Boothwyn, USA). Ambient gas phase samples were taken outdoors away from buildings at 1 m above the ground. For quality control purposes field blank samples were collected for each of the four sampling regions and these were subjected to the same treatment as the emission samples.

After sampling, the PDMS traps were sealed with glass caps, wrapped in aluminum foil, and stored in a cooler box with ice packs to ensure their integrity during transportation. The samples were stored in the laboratory in a freezer at –18 °C prior to analysis.

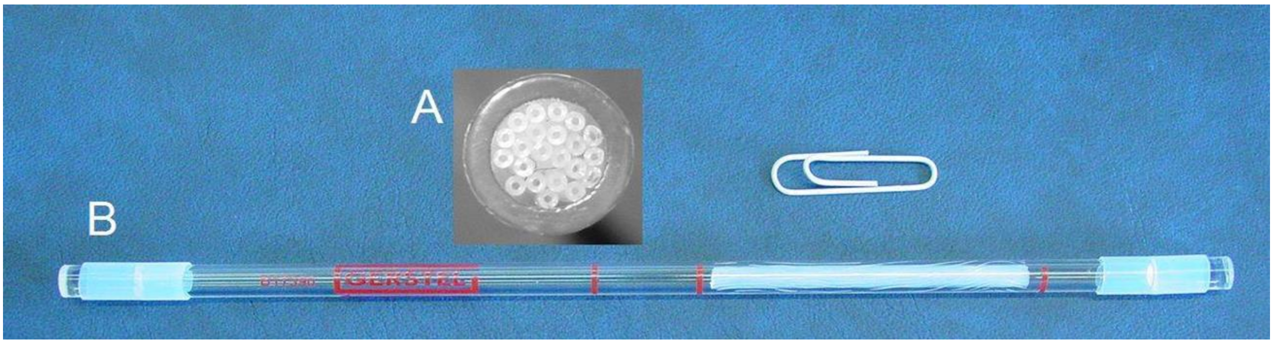


Figure 2. The PDMS sampling trap used in the study in (A) cross section and (B) side view showing glass storage end caps held in place with Teflon (Reprinted from Naudé et al., 2009 with permission from Elsevier).

Figure 3. Typical sampling setup used at (A) Narok rural HH#1 (Improved 3-stone) (B) Bomet rural HH#1 (Improved 3-stone) (C) Bomet urban HH#1 (Jiko), and (D) Narok urban HH#2 (Jiko). For all samples, the PDMS trap was positioned at 65 cm from the stove and at a height of 36 cm above the ground.

Table 1. Details of dwelling type, sampling location, fuel type and combustion device used in various households in Bomet and Narok Counties.

Household (HH #)	Sample abbreviation	Type of dwelling	Combustion device	Type of fuel	Sampling position	Location description
Bomet urban ambient	BU-AMB	n/a	n/a	n/a	1 m above ground level	About 100 m from Bomet urban Household #1(HH#1) at end of dirt road (at T-junction of another dirt road)
Bomet urban HH#1 (Duplicate samples)	BU-H1A & BU-H1B	Brick house with galvanized zinc roof	Wood stove / Jiko "improved"	Wood - cyprus (small pieces)	In kitchen on stool 36 cm above ground and 65 cm from the fire	The fire was under a chimney
Bomet urban HH#1	BU-H1C	Brick house with galvanized zinc roof	Jiko	Charcoal	In kitchen on stool 36 cm above ground and 65 cm from the jiko	The fire was under a chimney
Bomet rural ambient	BR-AMB	n/a	n/a	n/a	1 m above ground level	About 30 m from Bomet rural Household #1 (HH#1) (at end of dirt driveway where it joined the dirt road) amongst farmland

Bomet rural HH#1 (Sample 1)	BR-H1A	Wooden house with galvanized zinc roof	Improved stove - (cement structure)	Wood cyprus	In kitchen on stool 36 cm above ground and 65 cm from the fire	Well ventilated kitchen with open windows
Bomet rural HH#1 (Sample 2)	BR-H1B	Wooden house with galvanized zinc roof	Jiko	Briquettes (made from sawdust & bagasse): water had not begun to boil	In kitchen on stool 36 cm above ground and 65 cm from the jiko	Well ventilated kitchen with open windows
Bomet rural HH#1 (Sample 3)	BR-H1C	Wooden house with galvanized zinc roof	Jiko	Briquettes (made from sawdust & bagasse): second sample from same fire once water had started to boil	In kitchen on stool 36 cm above ground and 65 cm from the jiko	Well ventilated kitchen with open windows
Bomet rural HH#1 (Sample 4)	BR-H1D	Wooden house with galvanized zinc roof	Improved jiko	Charcoal (from wood fire in sample 1)	In lounge area/veranda adjacent to the kitchen, 36 cm above ground and 65 cm from the jiko	Sampled in this area to prevent cross contamination from previous combustion in the kitchen. Well ventilated with open door and windows
Narok urban ambient	NU-AMB	n/a	n/a	n/a	1 m above ground level	Adjacent to dirt road near Maasai Mara University
Narok urban HH#1 (Duplicate samples)	NU-H1A & NU-H1B	Zinc with galvanized zinc roof	Kerosene stove	Kerosene	On stool in kitchen 36 cm above ground	Inside zinc kitchen

					and 40 cm from the stove	
Narok urban HH#2	NU-H2A	Open outdoor shack made of plastic with wooden supports outside the house	3-stone	Wood & sticks	Outdoors next to zinc kitchen on a stool 36 cm above ground and 87 cm from the fire	Plastic shelter around fire used for outdoor cooking
Narok urban HH#2	NU-H2B	Brick	Jiko	Charcoal	On stool in kitchen 36 cm above ground and 65 cm from the jiko	Kitchen located inside house, with open door and window
Maasai Mara University (Duplicate samples)	MMUA & MMUB	Brick with tiled roof. Ground floor of 2-story building	Jiko	Briquettes doped with sodium citrate	In student office 36 cm above ground and 65 cm from the jiko	Open door and window
Narok rural ambient	NR-AMB	n/a	n/a	n/a	1 m above ground level	In farmyard in open farming area
Narok rural HH#1 (semi-rural)	NR-H1	Mud walls & galvanized zinc roof	3-stone type	Wood	On stool in kitchen 36 cm above ground and 65 cm from the fire	Inside kitchen with an open door and window
Narok rural HH#2	NR-H2	Galvanized zinc shack	Jiko	Charcoal (bought from trader)	On stool in kitchen 36 cm above ground and 65 cm from the jiko	In separate kitchen shack (with door but no windows) adjacent to the house
Narok rural HH#3	NR-H3	Manyata with clay/dung walls and roof	3-stone type	Wood	On stool in kitchen 36 cm above ground and 70 cm from the fire	Bedroom led directly off kitchen with very limited ventilation (no windows)

2.2 Chemicals and reagents

The overall analytical procedure, including calibration, was performed using a certified standard PAH mix solution (Supelco, USA) containing 15 US EPA priority PAHs. The nominal concentration of each compound in the mixture dissolved in methylene chloride was 2000 ng/µL. Stock solutions were prepared in n-hexane and working solutions in the range of 0.5 to 4 ng/µL were prepared by appropriate dilutions of the stock solutions before use. All solvents including toluene and n-hexane were of analytical grade (99% purity) and were purchased from Sigma Aldrich (Bellefonte, USA).

2.3 Extraction and GC-MS analysis of PAHs

All samples were extracted using the PASE method developed by Munyeza et al. (2018). Briefly, the traps were plunged 10 times with two portions of 1 mL hexane which were then combined to give a total volume of 2 mL. These extracts were concentrated by blowing down with nitrogen to near dryness, after which they were reconstituted in 100 µL hexane in amber vials. Pre-washed plungers, clean vials and pure solvents were used for the PASE extraction of each sample to prevent carryover of samples or cross-contamination. Sequential extractions were carried out with fresh portions of solvent to reduce losses due to trace analytes that may remain in the residual solvent in the PDMS tubes or heavy PAHs that may adhere to the glass walls. Two sequential extractions have been shown to result in optimum overall extraction efficiencies of the target PAHs, which ranged from 76% for naphthalene to 99% for phenanthrene, with percentage relative standard deviations (%RSDs) below 6% (Munyeza et al., 2018). The enhanced recovery due to sequential extraction was more evident for heavier target PAHs (4-6 rings). Relatively lower recoveries of lower molecular weight PAHs may be due to losses due to volatilization, especially for naphthalene.

Sample analysis was performed using a gas chromatograph (GC, Agilent 6890) connected to a mass spectrometer (MSD, Agilent 5975C) in electron impact ionization mode. The GC-MSD conditions are provided in **Table 2**. A mass range of m/z 40-350 was recorded in full scan mode. Compounds were identified based on a comparison of retention times and mass spectra to those of pure individual standards. For better sensitivity, the selected ion monitoring (SIM) mode was employed to detect compounds and quantify the analytes (**Table 3**).

Quantification of the selected PAHs was carried out using seven-point calibration curves. The calibration was set-up by spiking of traps with concentrations ranging from 0.5 ng/µL to 4 ng/µL (including blanks) for the 15 US EPA priority PAHs included in this study, the abbreviations of which are provided in **Table 3**. The limit of detection (LOD) and limit of quantification (LOQ) was calculated as 3 times and 10 times the S/N ratio (**Table 4)**. Samples were corrected for PAHs found on the respective field blank sample for that area.

2.4. Toxic equivalent quotient (TEQ) determination

The carcinogenicity of a PAH mixture or inhalation risk is often described in terms of its TEQ value, similar to the benzo(a)pyrene equivalent concentration ($B[a]P_{eq}$) (Xia et al., 2013; Munyeza et al., 2020). The TEQ of gas-phase PAH emissions from different cooking devices in this study was calculated according to equation (1):

$$TEQ\ or\ B(a)Peq = \Sigma_{i=1}^{n} C_i\ x\ TEF_i \text{ ---------------------------------------(1)}$$

where C_i = concentration of the PAH congener *i*; TEF_i = the toxicity equivalency factor (TEF) of PAH congener *i* (**Table S2**).

Table 2. GC-MSD conditions employed in the analysis of PAHs in PASE extracted samples.

Parameter	Details
Column	Restek Rxi®-PAH
Column dimensions	60 m, 0.25 mm ID, 0.10 µm d_f
Oven program	80 ^oC (1 min), 30 ^oC/min to 180 ^oC, 2 ^oC/min to 320 ^oC
Injection volume	1 µL
Inlet mode	Splitless (1 min), purge flow 30 mL/min (1 min)
Inlet liner	Restek SKY™ precision splitless liner without wool
Solvent delay	6.5 min
Inlet temperature	275 ^oC
Carrier gas	Helium, constant flow mode, 1 mL/min
Transfer line temperature	300 ^oC
Ionization energy	70 eV, electron impact mode (EI+)
Mode of detection	
Compound identification/ confirmation	*Full scan mode m/z 40 – 350*
Quantification	*Selected ion monitoring mode (SIM) m/z 128, 136, 152, 154, 166, 178, 188, 202, 212, 228, 240, 252, 276, 278*
MS temperature	230 ^oC (ion source), 150 ^oC (quadrupole)
Total run time	74.33 min

3. Results and discussion

3.1 PAH quantitation

The calibration method was employed for the quantification of target PAHs and correlation coefficients (R^2) for all analytes were higher than 0.920 (**Table 3**). The limits of detection (LODs) and limits of quantification (LOQs) based on average sample volumes (0.005 m^3) were also evaluated and are reported in **Table 4**. The concentration of PAHs sampled on each trap was calculated using equation (2):

$$C_{PAH} = \frac{M_v}{V_{air}} \text{------------------------------} (2)$$

where C_{PAH} is the concentration of each PAH per unit volume of air sampled (µg/m^3); M_v (ng) is the amount of target analyte determined from the linear regression calibration equations (ng/µL), divided by 1000 (to convert to µg), and multiplied by the volume of final extract (100 µL); and V_{air} is the volume of air sampled on the PDMS trap (~ 0.005 m^3).

The plunger-assisted solvent extraction (PASE) method described by Munyeza et al. (2018) was employed for the analysis of samples collected from indoor cooking-related combustion activities and ambient air samples, with a final extract volume after blowdown under N_2 of 100 µL. For improved sensitivity and selectivity, the selective ion monitoring (SIM) mode was employed to quantify the target PAHs (Adeola and Forbes, 2020, Munyeza et al., 2018). A representative GC SIM chromatogram is shown in **Figure S1**. Carryover between samples and contamination from solvent blanks did not occur, as PAHs were not detected in analytical grade solvents (99% purity) injected between sample runs. A trace amount of target compounds, especially volatile naphthalene, was detected in field blank samples and was deducted from sample concentrations accordingly. The Narok urban field blank sample was lost during analysis, thus the average of the other three field blanks was used for correction of Narok urban samples.

Seven target PAHs out of the 15 US EPA priority PAHs were above the limit of quantification (LOQ) in the samples (**Figure 4, Table S3**). Where analytes were detected in some samples but were <LOQ in others in the sample set (for example within Bomet rural samples), the LOQ was used in the calculation of average values as a worst case scenario. There was a vast variation in the total gas phase indoor concentrations of PAHs in households which were detected, ranging from 0.82 to 173.69 µg/m^3. Low molecular weight (LMW) PAHs were ubiquitous in the gaseous phase due to their relatively high vapor pressure; however, they are less toxic to humans. The high molecular weight (HMW) PAHs are more predominant in the particulate phase due to their low vapor pressures, with proven carcinogenicity (Dat and Chang, 2017). The particle phase was not sampled in this study due to low sampling volumes, which would have resulted in particle phase PAH concentrations being below the LODs. The importance of gas phase PAH emissions in determining exposure levels has been previously demonstrated (Geldenhuys et al., 2015, Munyeza et al., 2020).

Table 3. List of PAHs included in this study. Chemical formulae and number of fused benzene rings are shown along with the linear regression (R^2) calibration correlations (n=3).

Analyte (PAH)	**Abbreviation**	**Quantification ion (m/z)**	**Formula**	**Number of rings**	**R^2**
Naphthalene	Nap	128	$C_{10}H_8$	2	0.992
Acenaphthylene	Acy	152	$C_{12}H_8$	3	0.987
Acenaphthene	Ace	154	$C_{12}H_{10}$	3	0.988
Fluorene	Flu	166	$C_{13}H_{10}$	3	0.988
Phenanthrene	Phen	178	$C_{14}H_{10}$	3	0.981
Anthracene	Ant	178	$C_{14}H_{10}$	3	0.946

Fluoranthene	FluAn	202	$C_{16}H_{10}$	4	0.989
Pyrene	Pyr	202	$C_{16}H_{10}$	4	0.984
Benzo[a]anthracene	BaA	228	$C_{18}H_{12}$	4	0.984
Chrysene	Chry	228	$C_{18}H_{12}$	4	0.944
Benzo[k]fluoranthene	BkF	252	$C_{20}H_{12}$	5	0.924
Benzo[a]pyrene	BaP	252	$C_{20}H_{12}$	5	0.937
Dibenz[a,h]anthracene	DahA	278	$C_{22}H_{14}$	5	0.998
Indeno[1,2,3-cd]pyrene	IcdP	276	$C_{22}H_{12}$	6	0.929
Benzo[g,h,i]perylene	BghiP	276	$C_{22}H_{12}$	6	0.949

Table 4. Limits of detection (LODs) and limits of quantitation (LOQs) of PAHs based on SIM ions for the PASE method. The LOD was calculated based on a signal to noise (S/N) ratio of 3 and the LOQ on a S/N ratio of 10.

Target analyte (PAH)	**LOD (Injected) (ng/µL)**	**LOD (Trap) (ng/100µL)**	**Calculated LOD (µg/m^3)**	**LOQ (Injected) (ng/µL)**	**LOD (Trap) (ng/100µL)**	**Calculated LOQ (µg/m^3)**
Nap	0.005	0.5	0.10	0.016	1.6	0.320
Acy	0.003	0.3	0.06	0.009	0.9	0.180
Ace	0.002	0.2	0.04	0.006	0.6	0.120
Flu	0.007	0.7	0.14	0.024	2.4	0.480
Phen	0.003	0.3	0.06	0.009	0.9	0.180
Ant	0.004	0.4	0.08	0.015	1.5	0.300
FluAn	0.006	0.6	0.12	0.020	2.0	0.400
Pyr	0.003	0.3	0.06	0.011	1.1	0.220
BaA	0.002	0.2	0.04	0.005	0.5	0.100

Chry	0.007	0.7	0.14	0.022	2.2	0.440
BkF	0.001	0.1	0.02	0.003	0.3	0.060
BaP	0.002	0.2	0.04	0.007	0.7	0.140
DahA	0.025	2.5	0.50	0.082	8.2	1.640
IcdP	0.006	0.6	0.12	0.018	1.8	0.360
BghiP	0.003	0.3	0.06	0.010	1.0	0.200

Generally, Narok County samples had a higher average total PAH concentration, ranging from 70.84 µg/m^3 in rural homes to 35.88 µg/m^3 in urban households, compared to Bomet County (9.09 µg/m^3 in rural homes to 2.91 µg/m^3 in urban households) (**Table 5**). This could be attributed to the difference in atmospheric conditions, such as relative humidity, temperature, etc., as well as combustion devices, combustion fuel, ventilation, nature of households, etc. (Munyeza et al., 2020, Zou et al., 2003, Shen et al., 2011, Hellén et al., 2017). Other factors that could have contributed to the variation in PAH concentrations and related toxicity are further discussed in Sections 3.2 to 3.4. Naphthalene was present at the highest concentration in most of the households investigated in this study (**Figure 4, Table S3**), similar to earlier reports on combustion of different biomass fuels (Zou et al., 2003, Shen et al., 2011). This could be attributed to the fact that naphthalene has the highest vapor pressure and volatility and lowest molecular weight, thus will readily be found in the gas phase (Abdel-Shafy and Mansour, 2016). Elevated concentrations of naphthalene, as the most abundant PAH in most household kitchens in the study areas, was equally reported by studies carried out in coastal regions of Kenya (Munyeza et al., 2020), in Burundi (Viau et al., 2000), and Japanese kitchens (Ohura et al., 2004).

Literature suggests that the total PAH concentration in the gaseous/vapor phase often increases with an increase in temperature, and that lower relative humidity (RH) enhances the burning or combustion of biomass and gaseous release (Hellén et al., 2017). In this study, indoor temperatures averaged 24.3 ºC (ranging from 20.6 ºC to 27.9 ºC). This may have contributed to the lower gas phase PAH concentrations found in samples collected from Bomet and Narok Counties, compared to previous results obtained in Mombasa and Taita Taveta where the average indoor temperature was 31 ºC (Munyeza et al., 2020). These findings further emphasize the influence of seasonal variations in atmospheric temperature and relative humidity on the occurrence of vapor phase pollutants, and the need for adequate consideration of atmospheric factors in toxicological profiling and risk assessment of PAHs, and other gas phase pollutants.

Table 5. PAH concentrations in µg m^{-3} in indoor and ambient air from rural and urban inland Counties of Kenya

Sampling location	Sample abbreviation	Total PAHs (µg/m^3)	Average Total Household PAHs ± Std Dev (µg/m^3)
Bomet rural			9.09 ± 4.13
Bomet rural-household 1	BR-H1A	9.63	
Bomet rural-household 1	BR-H1B	3.13	
Bomet rural- household 1	BR-H1C	12.39	
Bomet rural- household 1	BR-H1D	11.20	
Bomet rural- ambient	BR-AMB	2.44	
Bomet urban			2.91 ± 1.82
Bomet urban-household 1	BU-H1A	4.16	
Bomet urban- household 1	BU-H1B	3.74	
Bomet urban- household 1	BU-H1C	0.82	
Bomet urban- ambient	BU-AMB	6.30	
Narok rural			70.84 ± 90.58
Narok rural-household 1	NR-H1	35.88	
Narok rural-household 2	NR-H2	2.94	
Narok rural-household 3	NR-H3	173.69	
Narok rural-ambient	NR-AMB	1.26	

Narok urban			35.88 ± 49.59
Narok urban-household 1	NU-H1A	4.60	
Narok urban-household 1	NU-H1B	133.10	
Narok urban-household 2	NU-H2A	3.96	
Narok urban-household 2	NU-H2B	41.68	
Maasai Mara University	MMU A	19.01	
Maasai Mara University	MMU B	12.90	
Narok urban-ambient	NU-AMB	6.28	

3.2 Role of combustion devices and fuel employed on gas phase PAH emissions

Studies have shown that the composition of gaseous emissions varies with different cooking devices and fuel sources (Shen et al., 2013a). This is because the combustion conditions often influence the concentration of pollutants that are released during the combustion of fuels, whether clean or not (Orasche et al., 2012, Orasche et al., 2013). Therefore, the four most prominent cooking devices found in the study area, which are jiko, 3-stone, improved 3-stone (molded with clay or bricks - see **Figure S2**), and the kerosene stove; were investigated in this study (**Figure 5**).

As illustrated in **Figure 5**, the combustion of wood in the 3-stone cooking device; which is a traditional fire-making method for cooking and is still in practice in developing countries; resulted in the highest average total PAH emissions relative to other cooking methods (70.69 μg/m^3). This is followed by the kerosene stove (68.85 μg/m^3); the jiko stove with charcoal as a fuel source (12.43 μg/m^3) and then the improved 3-stone stove (5.69 μg/m^3), which involves wood combustion but under more controlled conditions than traditional 3-stone stoves. Note that error bars are not included in this figure (and subsequent figures) due to the wide variation in results between samples. A similar result was reported for wood and charcoal cooking devices in rural areas of Tanzania (Titcombe and Simcik, 2011) and coastal areas of Kenya (Munyeza et al., 2020).

The profiles revealed that the concentrations of naphthalene were far above the concentrations of other PAHs for all cooking devices. Inefficient charcoal production, substandard cooking devices, and burning of wet wood will result in relatively higher PAH emissions, which may lead to variations in emissions reported for the 3-stone, improved 3-stone and jiko combustion devices in this study. These findings agree with studies that affirm that availability of proper ventilation, nature of wood (moisture content or wood type) and burning duration influences the concentration of smoke and PAHs released in households (Munyeza et al., 2020, Chomanee et al., 2009). It should be noted that substantially better reproducibility in terms of total PAH concentrations between duplicate samples for both wood burning (BU-H1A and BU-H1B) and briquette burning (MMU-A and MMU-B) jiko stoves was obtained than for the kerosene stove duplicate samples (NU-H1A and NU-H1B). This may point towards poor efficiency of the device tested.

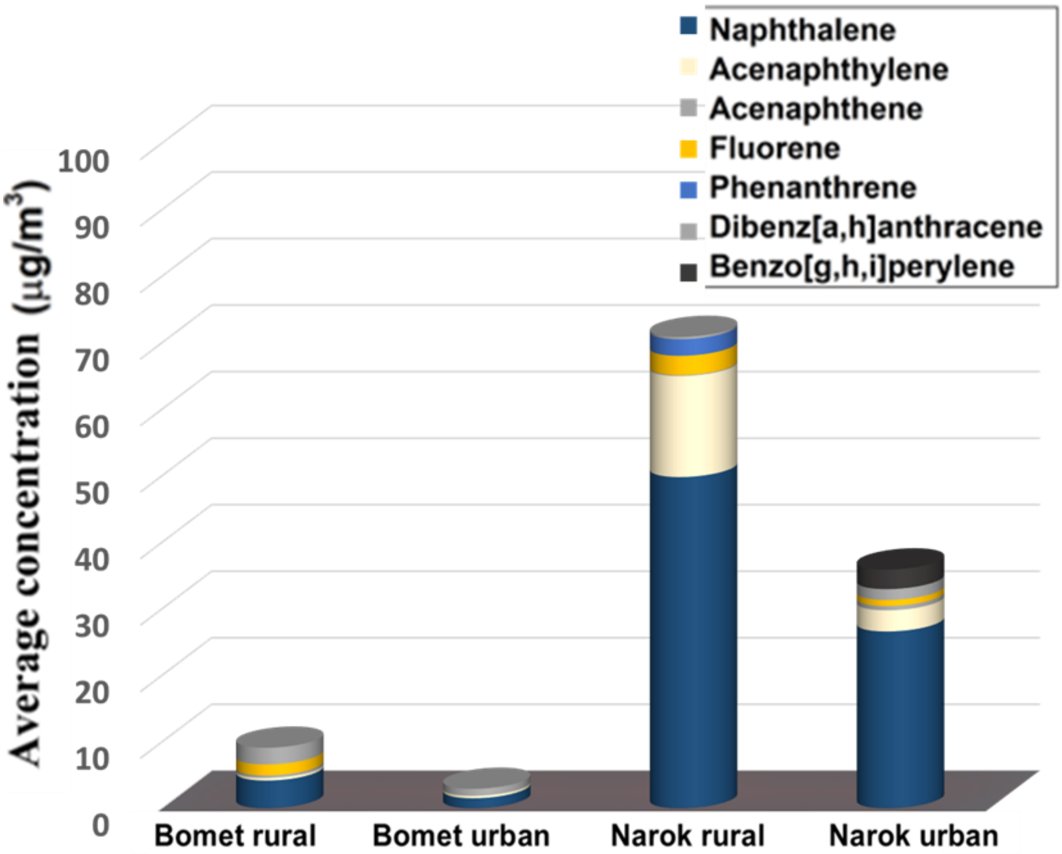


Figure 4. *Average gaseous PAH concentrations in urban and rural households of Narok and Bomet Counties of Kenya. LOQs were used in the average calculation where [analyte]<LOQ.*

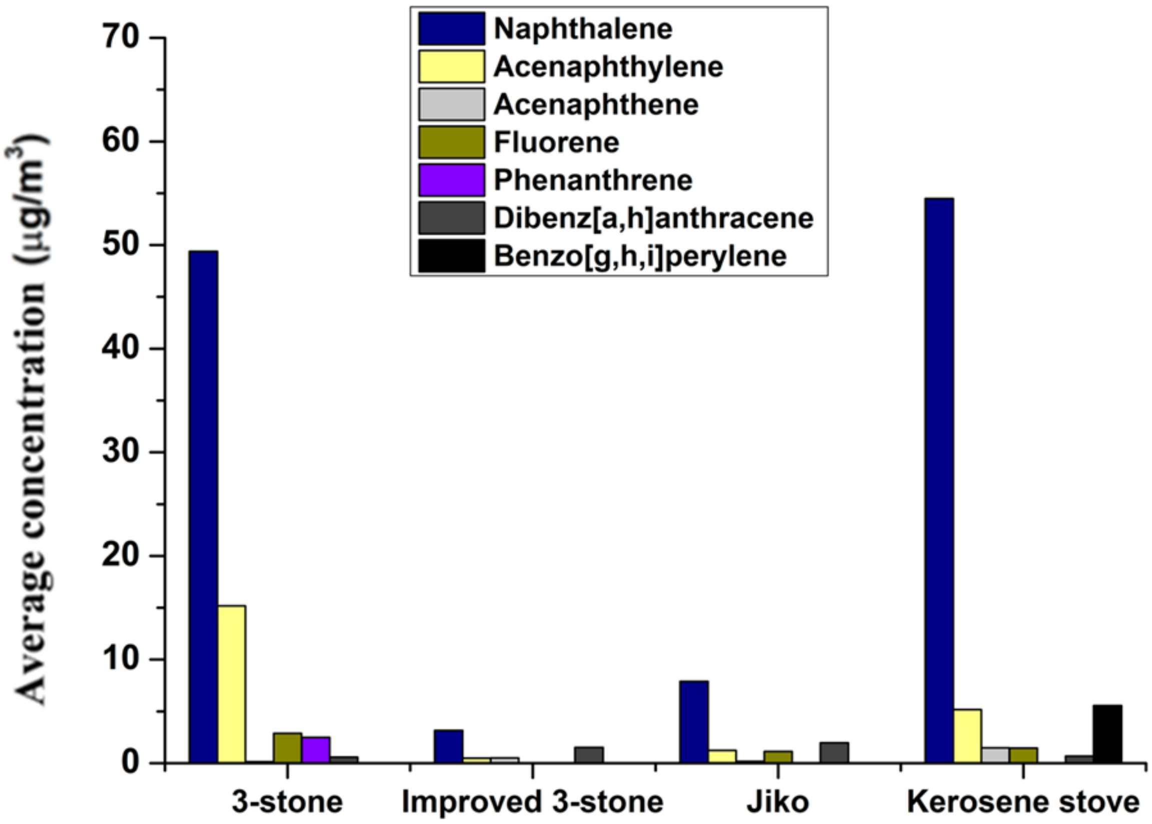


Figure 5. *Average gaseous PAH concentrations from various combustion devices.* $N_{(3\text{-stone})} = 3$, $N_{(\text{Improved 3-stone})} = 3$, $N_{(\text{Jiko})} = 8$, $N_{(\text{Kerosene stove})} = 2$. *LOQs were used in the average calculation where [analyte]<LOQ.*

3.3 PAH variation in rural and urban households

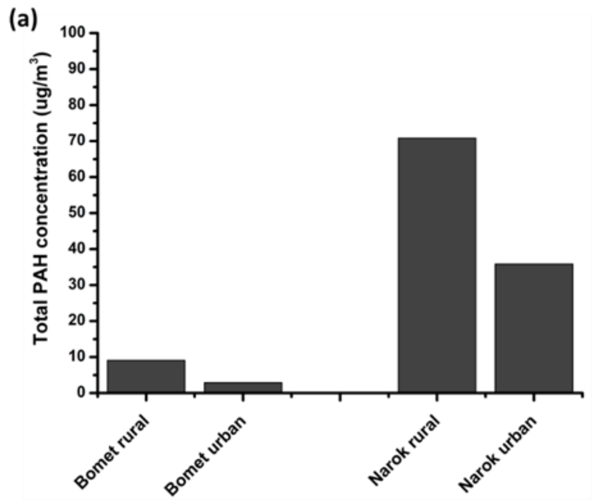
As illustrated in **Figures 4 & 6**, there is a marked difference in the concentration of PAHs in rural and urban households. Most rural households are poorly ventilated in general, with poor roofing structures and walls made of clay (**Figure S3 & S4**). In some cases, no chimneys were present, and the walls and roofs were consequently darkened with the smoke from combustion cooking devices. The housing structure, substandard cooking devices, and dependence on wood of all kinds as fuel, could be responsible for the higher PAH concentrations in rural compared to urban kitchens (**Table 5**). Although the total PAH concentration in rural and urban kitchens in Narok households was far higher than those found in Bomet homes, the ambient

PAH concentration in outdoor air samples in rural Bomet was somewhat higher than that found in rural Narok (**Figure 6a & b**). This affirms that household PAHs, generated *in-situ,* only contribute a portion to outdoor PAH concentrations and that several anthropogenic and outdoor activities such as vehicular and industrial emissions, as well as population density, contribute more to ambient PAH concentrations. During the sampling campaign, it was observed that a specific type of Maasai Mara traditional housing called a “manyata” predominates in rural Narok County (**Figure S3b**, sample NR-H3). This structure holds both the kitchen and bedroom of residents, with very limited ventilation. This contributes to the elevated level of PAHs found in the gas phase in households in rural Narok (**Figure 4**) and consequently increases the risk of exposure of residents to toxic gaseous pollutants generated from cooking within their living spaces.

Furthermore, naphthalene, acenaphthylene, acenaphthene and dibenz(a,h)anthracene were PAHs consistently and prominently detected in samples collected from wood combustion in urban and rural kitchens (**Figure 5**). The average ΣPAH level recorded as a result of the combustion of wood was as high as 173.69 $\mu g/m^3$ for six detected PAHs. Elevated average ΣPAH levels as high as 43 $\mu g/m^3$ for 12 detected PAHs were reported in rural households of Burundi by Viau et al. (2000). Vietnam recorded levels as high as 957 $\mu g/m^3$ for 18 ΣPAHs (Oanh et al. 1999). An earlier survey carried out revealed that the preference for a particular type of combustion device employed in rural and urban areas is influenced by the cost of the device, energy required/cost of fuel type, its availability, and cultural beliefs (Osano et al., 2020).

The pattern revealed in **Figure 6b** suggests ambient/outdoor gas phase air quality with respect to PAHs was better in rural areas of Narok and Bomet, this is expected due to relatively more anthropogenic activities and higher population density in urban areas that could negatively impact air quality (**Table S1**). Narok rural is near the Maasai Mara reserve, with much lower population density and limited local sources of PAHs, resulting in less ambient pollution in comparison to Bomet. Furthermore, Bomet is in the South Rift Valley region of southwestern Kenya, thus the topography and temperate climate of Bomet may reduce the dispersion of air pollutants to some extent (Osano et al., 2020), particularly as it was the wet season.

Primitive or traditional devices such as 3-stone and improved versions thereof were mainly employed in rural areas because they are often self-made and wood is abundant in villages which can be used as firewood, therefore 3-stone stoves are affordable by rural dwellers. Kerosene stoves, gas stoves, and coal devices (jiko) are mainly prevalent in urban residences as residents could afford them and have access to the fuel required. Discussions with rural dwellers during the sampling campaign also indicated that cooking using traditional methods is part of their cultural heritage and certain local meals such as 'ugali' are considered to be more delicious when made using firewood and a 3-stone device.



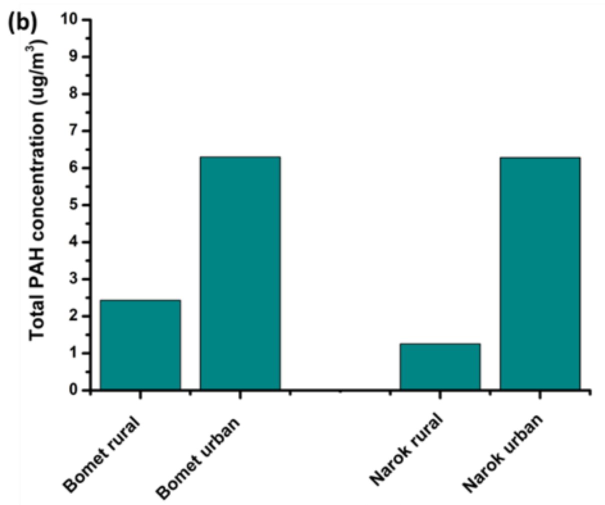


Figure 6. *(a) Total average PAH concentrations in rural and urban kitchens (b) total PAH ambient concentration from the sampled rural and urban areas.* $N_{(Bomet\ rural)} = 4$*,* $N_{(Boment\ urban)} = 3$*,* $N_{(Narok\ rural)} = 3$*,* $N_{(Narok\ urban)} = 6$*. LOQs were used in the average calculation for kitchen samples where [analyte]<LOQ.*

3.4 Toxicity assessment of detected gas phase PAHs

The carcinogenic potency and toxicity of PAHs were evaluated in this study, considering the PAH concentrations and relative distribution of different ringed PAHs. The human health risk (carcinogenicity) of PAHs released by the different cooking devices was calculated using Toxic Equivalence Factors (TEFs) proposed by Nisbet and LaGoy (1992). Based on the TEF values and average gas phase PAH concentrations, toxic equivalence quotient (TEQ) values for each device were estimated, as shown in **Table S2**. The sum of TEQ values of individual PAHs quantified and averaged for each cooking device were 3.0, 7.6, 9.9, and 3.6 $\mu g/m^3$ for 3-stone, improved 3-stone, jiko, and kerosene combustion devices, respectively (**Figure 7**). Furthermore, although **Figure 5** revealed the highest total PAH emissions from 3-stone stoves, TEQ values suggest that jiko stove emissions are more carcinogenic due to the relatively higher concentration of dibenz[a,h]anthracene released and its toxic equivalence factor (**Table S2)**. Thus, the quality of charcoal should be examined, and process technology involved in charcoal production should be standardized in the study area due to potential carcinogenic risks posed to residents utilising this fuel.

According to Nisbet and Lagoy (1992), the TEF value of two- to four-ringed PAHs (LMW) is 0.001, except for anthracene with 0.01. While five- to six-ringed PAHs (HMW) have TEF values ranging from 0.01 to 5. **Table 6** also reveals the gradual increase in the carcinogenicity of the PAHs as their molecular weight increases, except for the unique potency of benzo[a]pyrene (Patra, 2003). The total TEQ value is mainly influenced by the concentration of heavier PAHs present at sampling sites, due to higher TEFs of these PAHs. Similar to this study, the presence of benzo[g,h,i]perylene (a 6-ringed PAH) in gas phase ambient samples collected by the roadside has been reported (Nadali et al. 2021). The presence of heavier PAHs in the ambient gas phase was attributed to light-duty vehicular emissions and pyrogenic activities. **Figure 8** reveals that outdoor air samples also contained the 5-ring dibenz[a,h]anthracene, which is of concern. There is an overall higher proportion of HMW PAHs (dibenz[a,h]anthracene and benzo[ghi]perylene) in indoor samples from Narok County than in Bomet County. The presence of HMW PAHs in the gas phase can be a result of sampling near the source of emissions, thus condensation and equilibration thereof onto particles had not yet occurred.

Table 6. Relative toxicity and cancer potency of selected PAHs according to the US EPA and IARC (Nisbet and LaGoy, 1992; Patra 2003)

PAH	Toxic equivalency factor	Relative cancer potency
Benzo[a]pyrene	1	1.0000
Chrysene	0.01	0.0044
Benzo[k]fluoranthene	0.1	0.020
Benzo[a]anthracene	0.1	0.145
Dibenzo[a,h]anthracene	5	1.11

According to the International Agency for Research on Cancer (IARC), eight of the 16 priority PAHs listed by US EPA are potentially carcinogenic, namely benzo[a]pyrene, dibenzo[a,h]anthracene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, indeno[1,2,3-cd]pyrene and benzo[g,h,i]perylene (IARC, 2022; Wang et al., 2019). Two of these PAHs were above the limit of quantification in some samples collected during this study (dibenzo[a,h]anthracene and benzo[g,h,i]perylene) (**Table S2**).

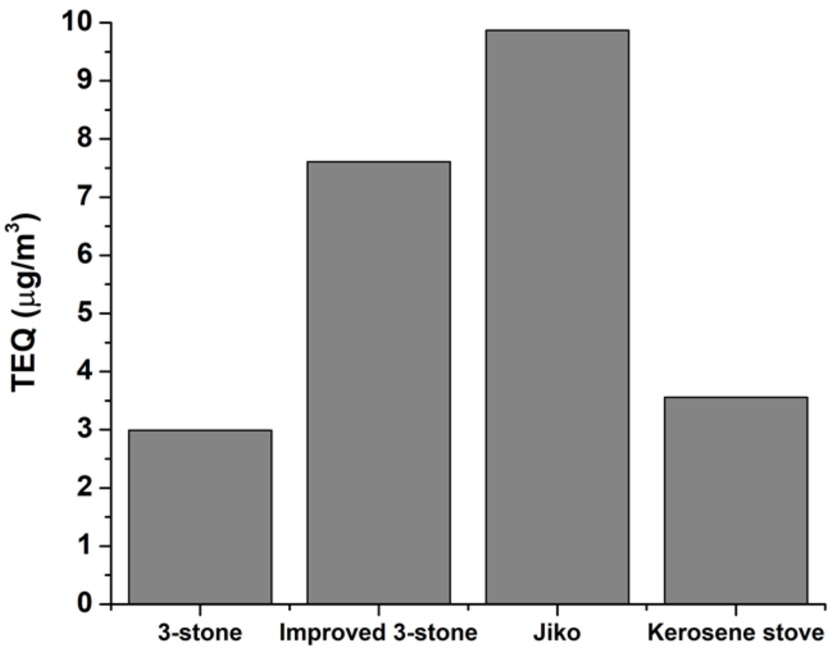


Figure 7. *Average toxic equivalent (TEQ) concentrations relating to different combustion devices for each PAH. N(3-stone) = 3, N(Improved 3-stone) = 3, N(Jiko) = 8, N(Kerosene stove) = 2. LOQs were used in the average calculation where [analyte]<LOQ.*

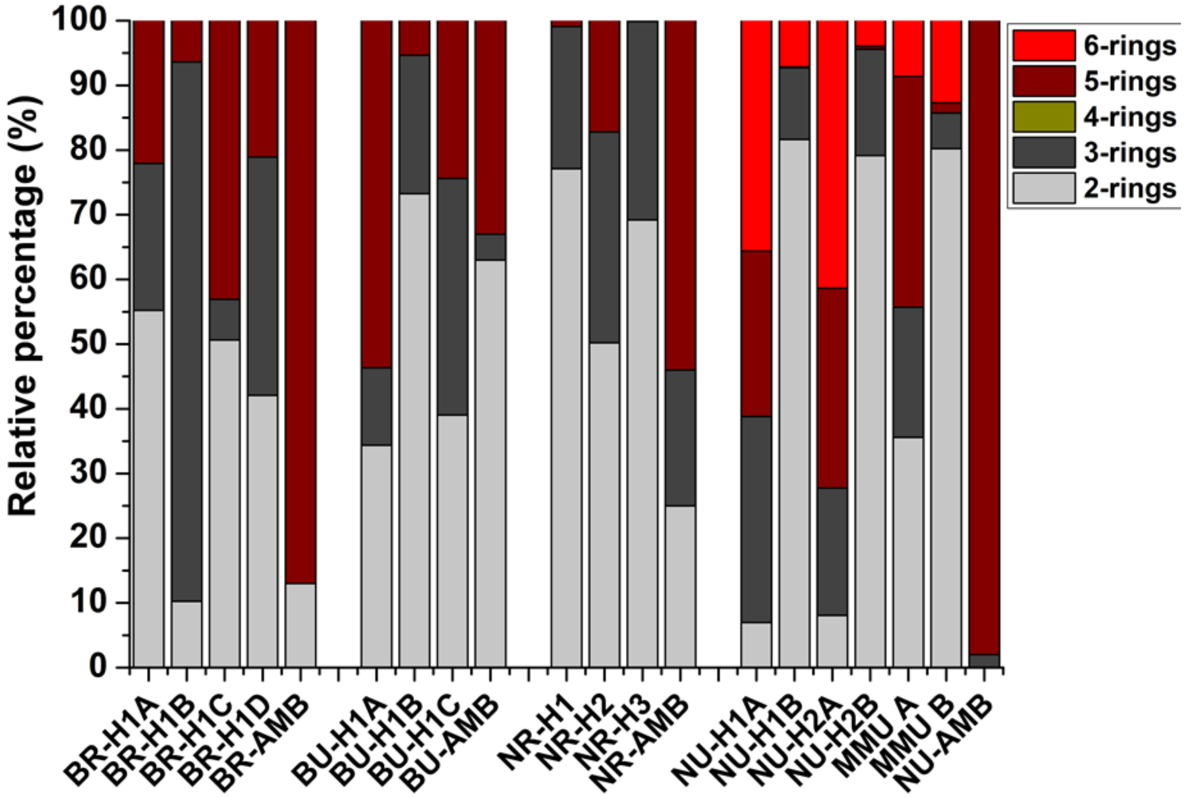


Figure 8. *Relative percentage of 2- to 6- ring PAHs present in the gas phase of individual household and ambient air samples.*

4. Conclusion

This study was performed to gain insight into the levels of PAHs released in the gas phase using local cooking devices, combusting wood, charcoal, and kerosene in the Narok and Bomet inland Counties of Kenya. The evaluation of emission sources and corresponding health risk assessment was possible using low volume portable samplers with subsequent plunger-assisted solvent extraction, and GC-MS analysis of extracts. Although large variations in PAH emissions were observed between houses due to differences in fuels, combustion devices, climate, and household ventilation, it was clear that naphthalene is the main contributor to indoor PAHs. Charcoal combustion using jiko stoves contributed the highest PAH toxic equivalent quotient (TEQ) whilst firewood combustion (in 3-stone stoves) resulted in the highest total PAH emissions in the studied households. Generally, people living in manyattas (traditional houses) in rural Narok are exposed to higher doses of cooking-related gaseous PAHs. The ambient/outdoor gas phase air quality with respect to PAHs was better in rural areas of Narok and Bomet which may be attributed to relatively more anthropogenic activities and higher population density in urban areas that could negatively impact air quality. The Narok rural sampling location is near the Maasai Mara nature reserve, with much lower population density and limited local sources of PAHs, resulting in lower ambient rural pollution in comparison to Bomet. The difference in PAH levels reported in coastal and inland Counties in Kenya was attributed to the difference in climatic conditions, fuel types, prevailing combustion devices, and type of households observed during the sampling campaign. The low molecular weight PAHs found at elevated levels in this study may react with atmospheric molecules such as O_3 and NOx to form highly toxic derivatives (nitro- and oxy-PAHs), thus monitoring thereof should be considered in future studies to allow for their inclusion in risk assessments. There is a need for local/on-site interactions with residents in the study area, and rural communities in developing countries in general, on the need for adequate ventilation in household kitchens and to promote the transition to cleaner fuels as integral aspects of pollution control and healthy living.

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Data availability

Data is available from the corresponding author upon request.

Conflict of interest

The authors declare no conflict of interest.

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