PAH Emissions from an African Cookstove

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**Abstract**

Combustion of wood and other biomass is a significant contributor to poor air quality in many developing countries. Emissions of particulates and Polycyclic Aromatic Hydrocarbons (PAH) are a major health hazard, particularly in Africa where the use of domestic cookstoves has increased alongside population expansion. Because of economic factors firewood is commonly used in place of the more expensive charcoal; particularly in rural areas. This work conducts a study of PAH emissions from an African cookstove comparing the combustion of both charcoal and firewood. It is demonstrated that PAH and particulate emissions are much higher from the firewood compared to the charcoal. The difference in levels can be interpreted due to the importance of the pyrolysis reactions of the volatile components of wood in PAH formation, whereas these volatiles emissions are much smaller from charcoal. Analysis of the combustion phases (flaming, smouldering) is undertaken and a computer model has been developed to link the composition of the fuels to the emissions of the PAH and particulates. The modelled PAH levels are shown to follow a similar trend to the experimental results.

Keywords: cookstove, firewood, charcoal, particulate emissions, PAH

**Highlights**

Emissions of PAH and particulate emissions from an African cookstove are measured.

Emissions from firewood and charcoal fuels are compared and both fine particulates and PAH were significantly higher for firewood.

A computer model has been developed to understand the main features of the PAH emissions.

The modelled PAH levels follow a similar trend to the experimental results.

## 1. Introduction

Atmospheric aerosols produced by combustion processes have an impact on both air quality [1-4] and on the climate [5, 6]. The global impact of pollution by ambient fine particulate matter (PM$_{2.5}$) in 2015 resulted in ~4.2 million deaths, representing ~4.2% of disability-adjusted life years [7]. This burden occurs predominantly in Asian and African countries where many people still rely on combustion of solid fuels for cooking and heating. Indoor air quality can be particularly poor due to lack of ventilation around cooking areas, which has a proportionately high impact on women.

The solid fuels used are mostly biomass and charcoal, predominantly burnt in traditional three-stone fires or cookstoves with high-levels of incomplete combustion. Particulate matter from combustion is commonly associated with carcinogenic polycyclic aromatic hydrocarbons PAH [8-12]. Improved cookstoves, designed to improve fuel efficiency, have been distributed across the developing world. Improved cookstoves may have lower PM and CO emission factors (EF) compared to traditional stoves [13,14]

The amount of fine particulate matter (PM$_{2.5}$) associated with cooking is increasing in Africa and also is a problem in India and China [10-15]. The major fuels are charcoal as well as wood and although charcoal is widely available in urban areas, it is expensive and thus many rural areas mainly use firewood. In Uganda, for example, ninety percent of the Ugandan population use charcoal or firewood-based fuel for cooking but the quantities used depend on the area of the country: the population in urban areas mainly use charcoal (70%) and some wood (15%), whilst poorer rural areas use more wood ( 86%) and less charcoal (11%) [16]. Rural areas predominately continue to use simple three stone fires, which can be made by the user. Generally the use of solid fuels for cooking in Africa has continued to increase alongside population growth [10] whilst in other areas, such as Europe it has tended to decline as shown in Fig 1
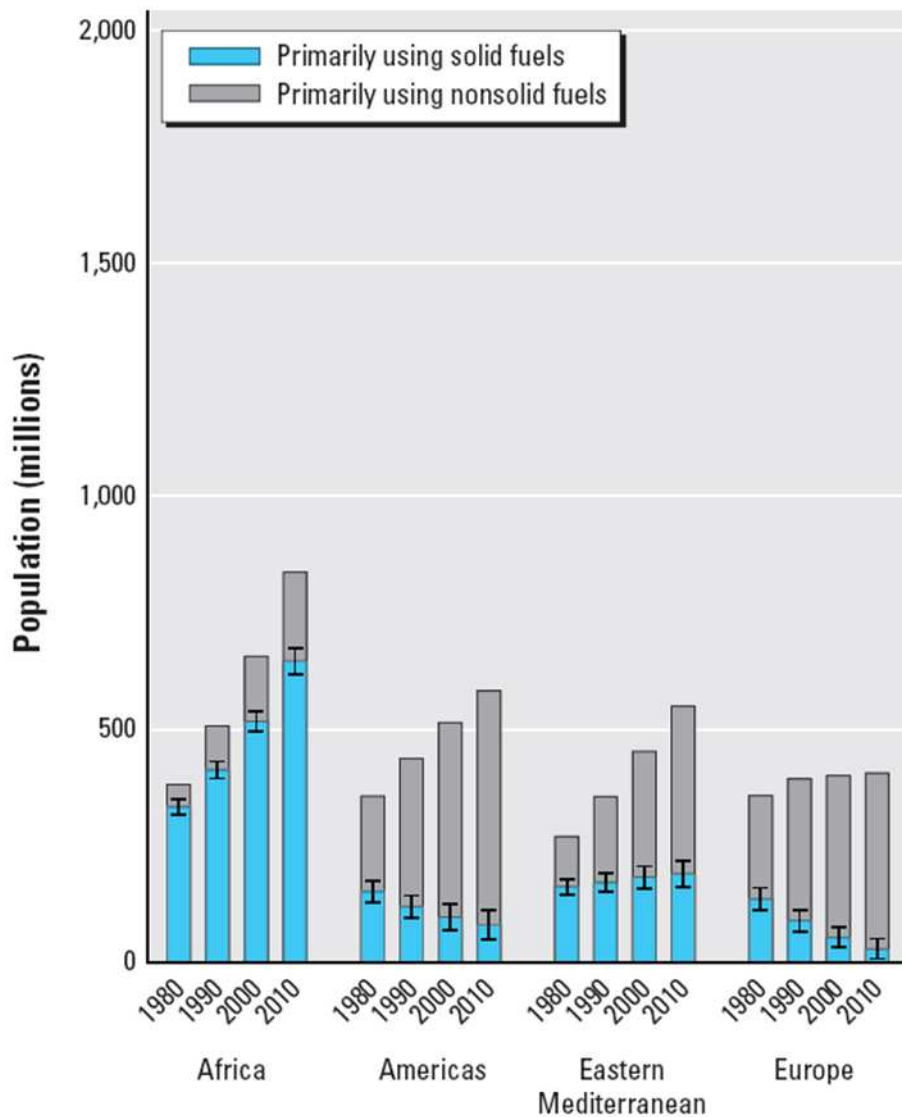
Fig. 1 Graph of population growth showing changes of primary cooking fuel, adapted from [8]

Many studies have been made of the gaseous emissions [13,14] and efficiencies for a variety of cookstoves, but few studies have been made of PAH emissions especially to make comparisons between wood and charcoal fuels, although there has been a recent study of the PAH emissions of a range of biomass fuels as well as coal, but nit charcoal [12]. In this paper the emissions of PAH and some gaseous species are compared for wood and charcoal using an improved cookstove widely distributed in Ghana. The levels of particulate species emitted are also determined. Fine particulates formed will act as carriers for adsorbed PAH species which pass deep into the lungs, potentially having carcinogenic and mutagenic effects whilst the particles themselves are associated with heart disease, cancer, lung diseases and other serious illnesses [7,8,15].

## 2. Experimental methods

### 2.1 Details of the Cookstove.

Many different designs of cookstoves have been used around the world ranging from the three stone method of heating to more energy efficient units. This study uses the Gyapa cookstove which is a widely used fuel-efficient stove designed for use with charcoal [17]. Almost 500,000 units have been sold across Africa mostly in Ghana. The unit consists of a metal casing with a 50 mm thick ceramic liner inside which reduces radial heat losses, and a raised grate designed with twenty five 15 mm air holes and a door to control the flow of primary air. .A photograph of this type of stove is shown in Fig. 2. The stove was placed in a ventilated Fire Box measuring 3m x 3m x 3 to give a controlled environment. Combustion occurs in the grate and the air flows through the bed of fuel. This gives a uniformity for the cooking process as well as providing a more uniform flow of air through the combustion zone. The cooking utensil is placed across the top on supports that are shown in the Fig. 2.



Fig. 2: Picture of the Gyapa Cookstove used showing the fuel feeding aperture and the exit holes for the combustion gases

The experimental study was conducted using firewood and charcoal and their Proximate and Ultimate compositions are given in Table 1. The compositions are typical of the fuels available in that region.

**Table 1.** Ultimate and Ultimate Analysis of the Fuels %db). Moisture is ar basis

|          | C     | H    | N    | S    | volatiles | Fixed carbon | ash  | moisture |
|----------|-------|------|------|------|-----------|--------------|------|----------|
| Charcoal | 76.72 | 1.73 | 0.59 | 0.04 | 16        | 70.7         | 13.4 | 3.1      |
| Firewood | 47.4  | 6.2  | 0.4  | 0.04 | 82.6      | 15.7         | 1.7  | 5.6      |

Db: dry basis; ar: as received basis

## 2.2 Measurement Methods.

Sampling of the combustion products was undertaken using a stainless steel sampling probe placed at 1m to the side of the stove in a position representing the head height of a seated cook. Flue gas composition was measured by online FTIR. Condensation of volatile species was prevented by use of a heated sample transfer line at 180ºC. PM $_{2.5}$ to PM $_{10}$ was estimated using a Dylos PM sensor. Total PM was measured gravimetrically at regular intervals throughout the combustion cycle using a filter holder heated at 70ºC in a way previously described [18]. Particulate samples were collected onto GF/F filter papers which were extracted into toluene using an ASE (accelerated solvent extractor), and subsequently analysed using a GCMS operated in SIM (Selected Ion Monitoring) mode. The estimated experimental error of the PAH emission data is 10%.

Emission factors were calculated according to the total capture method. In this method, all pollutant emissions from the cookstove are collected in the flue and the emission factor $EF_i$ is calculated as:

$$EF_i = \int_{t_0}^{t} \frac{C_i \times Q}{m} dt$$

where $C_i$ is the concentration of species i, Q is the flow rate in the flue and m the mass of fuel burned between times $t_0$ and t. This method is the most accurate since it relies on the direct measurement of the fuel burning rate and the flue gas flow rate [18].

## 3. Experimental Results

### 3.1. Particle and PAH measurements

The effects of the two fuels on the emissions were studied. Batch combustion in a fixed bed like a cookstove follows the stages of: initial ignition, flaming combustion and then smouldering. All emissions are highest for both fuels during the initial (cold) ignition phase but charcoal lumps burn for a long time without the need for the addition of fuel, that is, it is normally operating under smouldering conditions. In the case of firewood there is a frequent need to reload fuel, and hence a well-defined smouldering combustion stage is not achieved, measurements are taken effectively using an 'averaged value' over the flaming and smouldering phases.

Measurements were made of particulate $PM_{2.5}$, $PM_{10}$ and the PAH contents in the sampled gases. The $PM_{2.5}$ contains carbonaceous soot particles together with adsorbed PAH species, some particulate high molecular weight PAH and some condensed metal oxides with some very small fragments of char broken off during the combustion process. The $PM_{10}$ contains mainly fragments of char produced during the addition of additional fuel (refuelling) which occurred more frequently with the wood firing. Smaller particles are most harmful due to deeper penetration into the lungs. Table 2 shows the experimental data obtained 25min after ignition; in this time the structure of the stove reaches a steady temperature. It is clear that there is substantially more particles and more PAH are emitted with firewood combustion compared to charcoal, and that there is a greater proportion of small particles from the firewood. The measurement for the fine particles is consistent with wood firing generally. There is a greater correlation between the fine particles and PAH concentration that with the course particles.

**Table 2**. Comparison of Dylos average particulate concentrations and PAH concentration (taken 25 min after ignition- flaming combustion)

| | Fine Particles $PM_{2.5}$ ($\mu g/m^3$) | Coarse Particles $PM_{10}$ ($\mu g/m^3$) | Ratio of $PM_{2.5} : PM_{10}$ | Total PAH $ng/m^3$ |
|---|---|---|---|---|
| Charcoal | 7.1 | 2.5 | 2.9 | 7.9 |
| Firewood | 104.0 | 6.2 | 16.9 | 73.4 |
| Emission Ratio Firewood/Charcoal | 14.6 | 2.5 | - | 9.3 |

The PAH species identified are shown in Fig. 3 for a range of different phases of combustion for the two fuels and these were for charcoal (i) ignition, (ii) flaming and (iii) smouldering, and for wood (iv) ignition and (v) flaming; thus for each species there are five measurements. The PAH species observed ranged from fluorene to benzo(g,h,i)perylene. These were significantly higher for firewood compared to charcoal. PAH were barely detectable for charcoal once the smouldering combustion stage was reached. The distribution of the PAH species are in accordance with our recent paper [19] for samples which were collected from a wood stove using a heated filter maintained at a temperature of $70^{o}C$ as before [18]
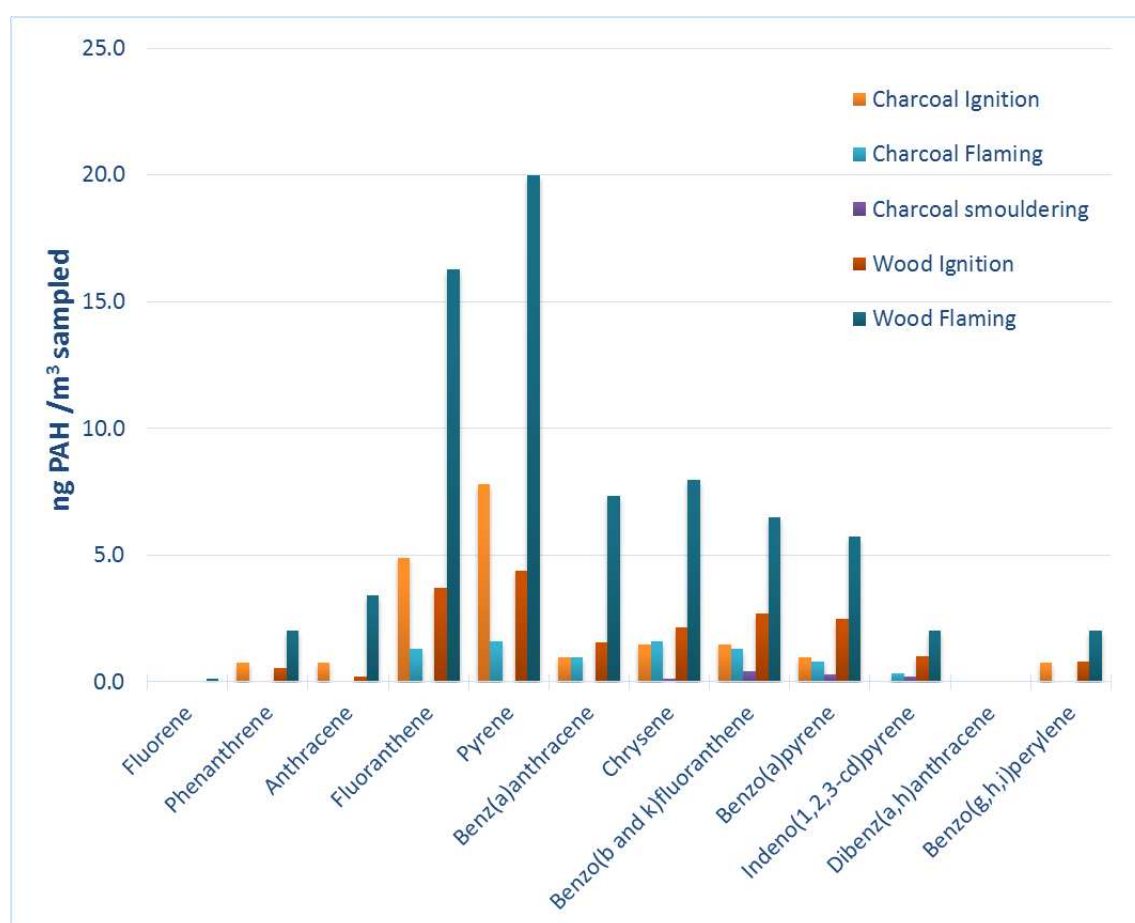


**Fig. 3** The distribution of the PAH species for charcoal and wood combustion collected on the filter during the different phases of combustion.

There is little published work on PAH emissions from cookstoves and none that can be directly compared with the results here. Ozgen et al. [20] have recently shown that for wood stoves that there is a considerable range of of PAH emissions depending on the combustion conditions

and the fuel. If CO is taken as an indicator of the degree of incomplete combustion, then the ratio of [CO]/[PAH] can be used to compare the results of the present work with other studies. If, as in [20], we consider the concentrations of the toxic PAH species benzo[a] and benzo[b]pyrene for flaming wood and this is divided by the concentration of CO from Table 3 it gives 230,000 ±25%. Ozgen et al. [20] show that for an open wood with a similar fuel the ratio has a value of 205,129 which is within a factor of 2 of the present data, which is good given the fact that we are comparing a cookstove against an enclosed wood-fired unit. Thus, Shen et al. [4] have measured emission factors for a range of PAH species for wood combustion which is consistent with the present data. In terms of isomer Rrtios as a source signature Shen et al [4] have recently compiled data for a number of wood fired appliaces but not for cookstoves, nor for the use of charcoal where ther is little information. They found that for wood ANT/(ANT+PHE) = 0.13, and FLA/(FLA+ ) = 0.53, where ANT = anthracene, PHE = phenanthrene, FLA = fluoranthene and PYR = pyrene. We obtained values of 0.4 and 0.44 respectively for the cookstove. The former higher value probably represents the inefficient combustion of this type of cookstove.

Measurements were made of the gaseous emissions from the smouldering charcoal and the averaged flaming firewood phase taken 45 min after ignition, and these are shown in Table 3. It can be seen that species associated with incomplete combustion - i.e CO, methane and ammonia, are substantially higher for the firewood. The proportion of gaseous emission from firewood relative to charcoal emission is ~15 times higher for the firewood, which correlates well with the observed increases in fine particles between the two fuels. This is consistent with the emission factor data of Bhattacharya et al. [21].

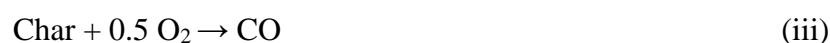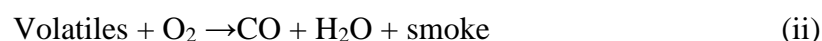**Table 3**. Average gaseous emissions (taken 45 min after ignition)

|  | CO ppm | NO ppm | $CH_4$ ppm | $NH_3$ ppm |
|---|---|---|---|---|
| Charcoal | 168 | trace | 11 | trace |
| Firewood | 2773 | 48 | 161 | 75 |
| Emission Ratio Firewood/Charcoal | 16.5 | - | 14.6 | - |

## 4. Discussion

4.1 Combustion Mechanism

The combustion of solid fuels such as wood and charcoal can be summarised [19] by the overall global reaction below:

$$Fuel \rightarrow volatiles + tar + char \qquad (i)$$

$$Volatiles + O_2 \rightarrow CO + H_2O + smoke \qquad (ii)$$

$$Char + 0.5\,O_2 \rightarrow CO \qquad (iii)$$

$$CO + 0.5\,O_2 \rightarrow CO_2 \qquad (iv)$$

$$Volatiles\ and\ tar \rightarrow smoke \qquad (v)$$

Any low molecular weight PAH, such as naphthalene, formed is included within the 'volatiles' group whilst the high molecular weight PAH is contained in the 'tar' together with any other high molecular weight pyrolysis products such as sugar derivatives. The major difference between the charcoal and the firewood is that the former has been thermally treated resulting in a reduction of the volatiles matter (VM) content of the fuel.

The formation of PAH species was computed using the Chemkin plug flow reactor model [22] and the POLIMI 1201 chemical kinetic model [23]. The empirical formula for wood was taken to be $C_4H_6O_3$ which is derived from the chemical composition of the wood given in Table 1. On the same basis charcoal has an empirical formula of $C_{6.5}H_2O_1$. In the calculation the diameter of the cookstove was 25cm and the vertical gas velocity of 2 cm/s was used. The results are not sensitive to this value and doubling the value only changes the computed results by about 10%.

In the case of dry wood, the pyrolysis gases on the basis of the empirical formula are taken to be $CO_2+CO+\{C_2H_6\}$ where $\{C_2H_6\}$ represents the average molecular formula of the pyrolysis hydrocarbon gases. Thus, by assuming a reasonable amount of air entrainment the gases to be combusted are $\{C_2H_6\} + 0.8\,N_2 + 0.1\,O_2$. This is designated as Case A. However the actual pyrolysis products from wood are not aliphatic hydrocarbons but consist of products from cellulose and the aromatic products from lignin as previously discussed [24]. The output from the Chemkin model of the behaviour of the reaction products showed that ethane pyrolysed immediately to give a pool of C/H/O/species which rearranged to a number of

species including acetylene which then form benzene and PAH species via the HACA route [24]. Methane is also formed in this way [25].

The simulated concentrations of selected key species at the exit of the combustor 9cm above the bed are given in Table 4. This distance was chosen to represent the distance a cool cooking utensil is above the burning bed. At this position it is assumed that the reaction products are quenched and no further reaction takes place.  But the wood pyrolysis volatiles are not all aliphatic species and should not be represented by ethane, since aromatic species are also formed. To approximate a more realistic set of pyrolysis products a mixture of {0.5 $C_2H_6$ + 0.25 $C6H5OCH3$} was used, the former representing the cellulose products and the latter those from lignin. The results are designated Case B and are shown in Fig 4 and Table 4. In Fig 4, which is the direct Chemkin output, the compositions of naphthalene; anthracene; acenaphylene; pyrene and the soot precursors, Bin 1A and Bin1 B are shown as they pass from the fuel bed through to the surface of the cooking utensil where the temperature drops and the reactions are quenched. Bin 1A and Bin 1B are precursors of soot formation and have been used to model the formation of particulate soot [26]. Of the PAH species the relative concentrations are as might be expected except that here the volatile naphthalene and anthracene are not collected on the 70$^o$C filter. The concentrations at the exit point are given in Table 4, and this includes the values for CO and methane. In Table 4 the values for Case A are also given. And the effect on the PAH and soot precursors are lower than for the realistic Case B.
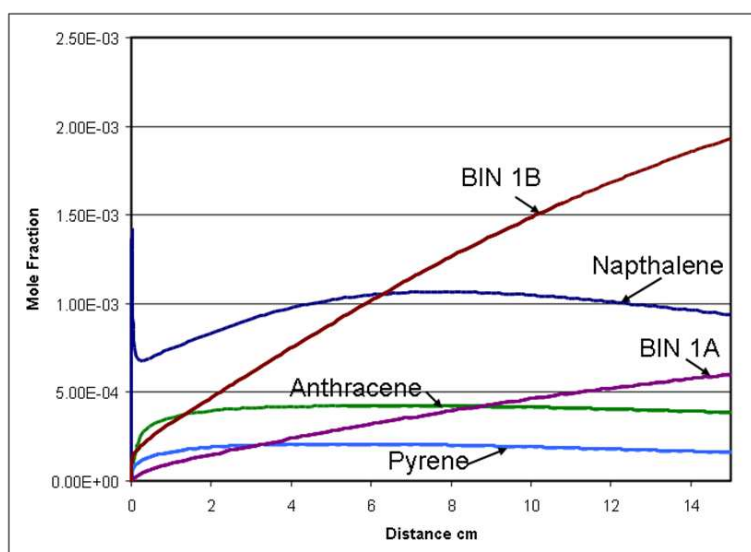


**Fig. 4.** Chemkin output for Case B: naphthalene, anthracene, pyrene, and soot precursors Bin 1A and Bin1 B.

**Table 4**. Computed species (mol fraction) at a vertical distance of 9cm from the combustion bed.

| | Case A, wood, non-aromatic products assumption | Case B, 1300 ligno, products assumed to be similar to real pyrolysis products | Case C, charcoal |
|---|---|---|---|
| $CH_4$ | 3.2 E-2 | 2.8 E-2 | 3.7E-2 |
| benzene | 1.36 E-4 | 1.7 E-3 | 6.1 E-4 |
| naphthalene, $C_{10}H_8$ | 6.19 E-4 | 8.6 E-4 | 1.5 E-4 |
| anthracene, $C_{14}H_{10}$ | 1.67 E-4 | 3.6 E-4 | 2.0E-5 |
| Pyrene, $C_{16}H_{10}$ | 7.57 E-5 | 1.4 E-4 | 9.4 E-6 |
| Smoke precursor: Bin 1 A | 1.17 E -4 | 6.6 E-4 | 2.7E-5 |
| Smoke precursor: Bin 1 B | 5.0 E-4 | 2.1E-3 | 2.7E-4 |
| CO | 7.1 E-2 | 9.5 E-3 | 3.2 E-2 |

In the case of charcoal, the devolatilisation products calculated in the same way, are 0.1 $CH_4$, 0.8 $N_2$, 0.1 $O_2$. Computations were undertaken for some of the key PAH components including benzene, $CH_4$ and CO. The results are designated Case C and are shown in Table 4. Interestingly the PAH products from the charcoal are of a much lower concentration than for firewood, which is observed experimentally.  The data for $CH_4$, CO and NOx emissions obtained are comparable with previous experimental data [26].

The concentration of CO has been widely used as an indicator of the level of poor mixing ('unburnedness') in combustion systems, although generally it is applied to turbulent combustion systems where it is also used as a measure of combustion efficiency. Here we have a system with a low degree of turbulence but the methodology is still applicable. It is known that PAH emissions are also an indicator of incomplete mixing and there is approximately a

linear relationship between PAH and CO. This is also true for $CH_4$. Generally PAH concentrations in the literature are approximately 10 times smaller than the CO and $CH_4$. There is a strong correlation between the ratios of emission of CO, $CH_4$ and fine particles when comparing the two fuels.

The relevance of this work to health is complicated by the range of toxicities of different PAH species. So far no 'particular source or component that uniquely determine the toxicity of the $PM_{2.5}$ mixture, and therefore the evidence does not support the development and application of source-specific relative risk functions for burden estimation' [8]. Whilst PAH has been associated with the particulate toxicity because of its involvement in carbonaceous particle growth the amount associated with individual particles is dependent on the time temperature history of the particle [16].

5.  **Conclusions**

The use of wood as a fuel results in higher emissions of PAH than charcoal by a factor of nine, under the operating conditions used here. This is explained by the PAH and the smoke emission being dependent on the volatile matter contents of the fuels, that for wood being much higher than that of charcoal, due to prevalence of pyrolysis reactions.

The ratio of $PM_{2.5}$: $PM_{10}$ is much greater for wood than it is for charcoal. This is consistent with previous studies which show emissions of fine particles of smoke is proportional to the volatile matter in the fuel. A correlation between higher fine particles and PAH emissions is indicated.

Methane emissions are fifteen times higher for wood than for charcoal. Again this is due to difference in the volatile emissions.

Emissions of NOx from firewood and charcoal are similar and these both depend on the fuel–N content of the fuel. However, emission factors on a thermal basis for NOx using wood is slightly higher than for charcoal.

The computer model developed from analysis of the combustion mechanism supports the observed link between the differences in volatile matter of the fuels with the PAH and particulate emission.

## Acknowledgements

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