Size distributions of polycyclic aromatic hydrocarbons in aerosol emissions from biofuel combustion

Chandra Venkataraman*, Geeta Negi, Satya Brata Sardar, Ritu Rastogi

Centre for Environmental Science and Engineering, Indian Institute of Technology, Bombay, Powai, Mumbai-400 076, India

Received 18 April 2001; accepted 29 August 2001

Abstract

Size-resolved emission factors of polycyclic aromatic hydrocarbons (PAH), from biofuel combustion in cooking stoves in India, were measured using a dilution sampler providing sufficient dilution and quenching for aerosol stabilisation. Emission factors of PAH (total of ten compounds) from combustion of biofuels like wood, briquette and dung cake, respectively, ranged 2.0–3.2, 2.8–3.0 and 3.1–5.5 mg kg⁻¹, per mass fuel burnt and 1.9–3.3, 4.1–4.8 and 5.9–10.6 mg kWh⁻¹, per useful heat input. Dung cake and briquette fuels were significantly more polluting than wood. The PAH profiles showed a predominance of fluoranthene, pyrene and benz(a)anthracene from all biofuels. Ratios of the semi-volatile PAH (3–4 ring species) to the non-volatile PAH (5-ring and larger), were significantly higher from briquette and dung cake, than from wood combustion. The PAH size distributions from all stove–fuel systems were unimodal with mass median aerodynamic diameters (MMADs) in the 0.40–1.01 μm range, for both semi-volatile and non-volatile PAH. Average MMADs of the PAH size distributions from biofuel combustion were about 5–10 times larger than in emissions from fossil fuel combustion like from automobiles, with potential implications for lung deposition and health risk. The PAH/PM ratio showed no simple trend with average particle diameter, suggesting that PAH were associated with the aerosol through multiple mechanisms including adsorption and absorption. This hypothesis must be further examined through surface characterisation methods like PAH photoemission or through particle and PAH formation models for biofuel combustion systems. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Biomass; Wood; Dung-cake; Cooking stoves; Emission factor
1. Introduction

Polycyclic aromatic hydrocarbons (PAH), associated with combustion and ambient aerosols, have been studied because of their carcinogenicity and pro-mutagenicity in animal and bacterial assay tests (Finlayson-Pitts & Pitts, 1986). Biofuels, including wood, dried animal-dung cake and crop waste, are used extensively for cooking energy in India and high concentrations of particulate PAH compounds have been reported in indoor environments, during biofuel combustion for cooking (Aggarwal, Raiyani, Patel, Shah, & Chatterjee, 1982; Raiyani et al., 1993a, b; Smith, 1987). Concentrations of benzo(a)pyrene measured over 15–30 min average sampling periods during wood and dung-cake combustion ranged 1.30–9.30 μg m\(^{-3}\) (Aggarwal et al., 1982). Concentrations of 16 particulate PAH measured over the cooking period of 45–60 min were 2.01, 3.46 and 3.56 μg m\(^{-3}\), respectively, from wood, wood/dung cake and dung-cake combustion (Raiyani et al., 1993a). Particulate PAH size distributions measured in these same indoor environments showed over 75% of the PAH mass in particles < 2 μm aerodynamic diameter (Raiyani et al., 1993b). There was a predominance of benzo(a)pyrene (35%) and dibenz(a,h)anthracene (15%), and of pyrene (18%) and benzo(a)pyrene (15%), respectively, in particles from wood and dung-cake combustion.

Higher incidence of cor pulmonale leading to cardiac disease, of chronic obstructive lung disease and of bronchitis, in biofuel users in India, has been documented by medical practitioners from various parts of India (Smith, 1987). In addition, higher occurrence active tuberculosis and blindness have been estimated for the Indian population of biofuel users (Misra, Retherford, & Smith, 1997a, b). While there is yet no direct evidence of higher lung cancer incidence in biofuel user populations in India, high daily doses of benzo(a)pyrene (Raiyani et al., 1993a) and an excess cancer risk of 5 × 10\(^{-3}\) (Smith, 1987) were estimated, using measured average PAH concentrations, in terms of benzo(a)pyrene equivalents, and average exposure times experienced by biofuel users.

Low-temperature biofuel combustion has the potential to result in higher emissions of PAH than high-temperature industrial sources. PAH emission factors from wood combustion in small-scale space-heating stoves exceeded those from furnace-oil combustion by a factor of 100 on an energy equivalent basis (Smith, 1987). Further, the mutagenicity emission factor, expressed as the number of revertants in a microbial assay test, from wood combustion exceeded that from furnace-oil combustion, modestly (factor of about 5) on a per kg of fuel basis, but substantially (factor of over 120) on a per energy equivalent basis (Smith, 1987).

Limited measurements of PAH emission factors have been made from small-scale, cooking-fuel combustion in an environmental chamber experiment (Gupta, Saksena, Shankar, & Joshi, 1998), and using duct sampling from a hood (Oanh, Reutergardh, & Dung, 1999). The reported emission factors of benzo(a)pyrene in respirable particles (< 5 μm aerodynamic diameter) from wood and root-fuel combustion ranged 5.3–13.2 mg kg\(^{-1}\) (Gupta et al., 1998). Total emission factors of 18 PAH in particulate-phase (by filtration) and vapour-phase (adsorption into an XAD-2 resin) ranged 25–100 mg kg\(^{-1}\) and were found to be highest from wood, followed by coal briquettes and charcoal (Oanh et al., 1999).

Emission factors of PAH from combustion of commonly used biofuels are required as the source term for estimating indoor concentrations under the widely varying building and ventilation conditions, and quantity of fuel burnt (Saksena, 1999). Subsequent risk estimation would
be based on the exposure time and the PAH dose received. The PAH size distribution would govern the fraction deposited in the lung (Venkataraman, & Raymond, 1998), needed to refine dose. Therefore, the measurement of size-resolved PAH emission factors is important for dose and risk assessment from exposure to stove smoke.

The PAH size-distribution which is governed by gas-particle partitioning (Finizio, Donald, Harner, & Bidleman, 1997; Venkataraman, Thomas, & Kulkarni, 1999) would be highly sensitive to the time-temperature history of the post-combustion gases. Thus, the need for size-resolved emission factor measurement using a sampling system providing sufficient quenching and stabilisation of the aerosol, is extremely pertinent in regard to PAH. Therefore, the objectives of this study were the following:

(i) Measuring, for the first time, size-resolved PAH emission factors from biofuel combustion, using a dilution sampler for aerosol stabilisation.

(ii) Relating these to fuel and combustion parameters to identify cleaner burning options.

2. Experimental methods

Particle samples were collected using a dilution sampler (Fig. 1), designed to provide sufficient dilution, cooling and residence time to achieve the quenching typical of indoor environments, constructed of stainless-steel (SS-304), with Teflon gaskets to avoid organic species contamination (Venkataraman & Rao, 2001). The burn cycle experiments, pollutant sampling and data inversion methods are described in detail in the earlier work. Fuels used in this study, included wood (Acacia nilotica, local names keekar or babul), dung cake and biofuel briquettes. Stoves used (Fig. 2) included three one-pot stoves (metal-S1, fired-clay-S2, traditional mud-S3) and one two-pot fired-clay stove, S4. Stoves S1, S2 and S4 were designed for improved thermal efficiency, while S3 was a traditional, U-shaped, mud stove.

The burn-cycle, involved heating 1.5 kg of water, from room temperature to its boiling point, and simmering for 5 min. This includes emissions from both low- and high-power phases, likely to be encountered in real cooking practice, giving a realistic emission factor. The time for accomplishment of the burn-cycle corresponded to the particle sampling times. The stove-fuel-pot assembly (fitted with a thermometer) was placed on a platform balance, to record weight (and hence fuel burn rate) every minute.

A micro-orifice uniform deposit impactor (MOUDI Model 110, MSP Corporation, USA), operating at a flow rate of 30.1 min$^{-1}$ was used to obtain a size fractionated particle sample. The MOUDI has 50% cut-point aerodynamic diameters of 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.097 and 0.056 μm on stages one through ten, and collects particles $<0.056$ μm on a 37-mm quartz-fibre after-filter. Flow-rate and leak checks were conducted on the impactor before and after each experiment and the 9th stage stagnation pressure was monitored using a pressure gauge. Aluminium foils of 47 mm diameter were coated with silicone by solvent evaporation of 20 μl of 4% silicone in cyclohexane solution, which was spread using a clean glass rod. All substrates were conditioned, before and after sample collection, for 8-h at about 25°C and 50% RH (in an air-conditioned room), and weighed on a microbalance (Model BP211D, Sartorius, Germany) accurate to 10 μg.
The adsorption artefact associated with sampling of semivolatile organic compounds (Grosjean, 1983) would not be of importance here, because the substrate used is an aluminium foil (with low adsorption area) and not a fibre filter. The low sampling flow rate and the use of an impactor (significant particle area not exposed to air flow) would result in negligible blow-off losses of semi-volatile PAH species, reported during high-volume sampling on glass fibre filters (Coutant, Brown, Chuang, Riggin, & Lewis, 1988). Estimates of volatilisation losses from a single jet impactor (Zhang & McMurry, 1991) showed collection efficiencies over 98% for species of vapour pressures lower than $10^{-9}$ atm, applicable to the PAH compounds considered here.

The particle-laden foils (from the MOUDI) from burn-cycle experiments were subjected to PAH analysis. HPLC grade solvents (Spectrochem Ltd., Mumbai, India) were used for extraction and chromatography. The particle-laden foil or filter was cut into pieces, placed in an amber coloured vial to exclude radiation and covered with 5 ml of dichloromethane (DCM). Extraction was done by sonication at 27°C for 30 min in an ultrasonic bath (Model No. 3210, Branson...
Inc., USA). The DCM extract was filtered through a 0.2 μm Teflon membrane filter (No. FGLP 02500, Millipore, USA) and evaporated to complete dryness under nitrogen. The residue was taken up in 200–1000 μl of acetonitrile for injection, depending on the total mass collected. Pre-cleanup methods using liquid–liquid extraction had high precision, but low recoveries of 60–80% for several PAH compounds, and were therefore not used (Negi, 1999).

A high performance liquid chromatograph (Series 200 LC pump, Perkin-Elmer, USA) was used, with a wavelength programmable fluorescence spectrophotometer (Model No. LS 30, Perkin-Elmer, USA). The program of time-varying excitation and emission wavelengths was optimised for highest sensitivity based on compound retention time (Table 1). A reversed-phase PAH separation column (ChromSpher, 4.6 mm × 100 mm, Perkin-Elmer, USA) was used, with gradient elution of the mobile phase at 1.5 ml min⁻¹ with a composition change from 75% acetonitrile in water to 100% acetonitrile in 16 min. The mobile phase solvents were degassed by helium bubbling to avoid quenching of the fluorescence signal by oxygen.
Table 1
Details of PAH analysis and recovery results

<table>
<thead>
<tr>
<th>PAH compounda</th>
<th>Elution time (min)</th>
<th>Excitation/emission wavelength (nm)</th>
<th>Lower quantifiable limits (pg)</th>
<th>System blanks (ng)</th>
<th>Recovery (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>FLT</td>
<td>3.61</td>
<td>280/450</td>
<td>8.3</td>
<td>NDb</td>
<td>93c</td>
</tr>
<tr>
<td>PYR</td>
<td>4.12</td>
<td>265/385</td>
<td>2.5</td>
<td>0.10</td>
<td>90</td>
</tr>
<tr>
<td>BAA</td>
<td>6.21</td>
<td>265/385</td>
<td>2.0</td>
<td>ND</td>
<td>94</td>
</tr>
<tr>
<td>CHR</td>
<td>6.88</td>
<td>265/385</td>
<td>1.8</td>
<td>0.04</td>
<td>96</td>
</tr>
<tr>
<td>BBF</td>
<td>9.26</td>
<td>297/430</td>
<td>4.0</td>
<td>ND</td>
<td>97</td>
</tr>
<tr>
<td>BKF</td>
<td>10.67</td>
<td>297/430</td>
<td>0.7</td>
<td>ND</td>
<td>95</td>
</tr>
<tr>
<td>BAP</td>
<td>11.59</td>
<td>297/430</td>
<td>1.3</td>
<td>0.02</td>
<td>96</td>
</tr>
<tr>
<td>DBA</td>
<td>14.18</td>
<td>297/430</td>
<td>3.1</td>
<td>ND</td>
<td>96</td>
</tr>
<tr>
<td>BGP</td>
<td>14.58</td>
<td>297/430</td>
<td>2.4</td>
<td>0.08</td>
<td>92</td>
</tr>
<tr>
<td>INP</td>
<td>15.82</td>
<td>305/500</td>
<td>19.4</td>
<td>ND</td>
<td>99</td>
</tr>
</tbody>
</table>

aAbbreviations: FLT (fluoranthene), PYR (pyrene), BAA (benzo[a]anthracene), CHR (chrysene), BBF (benzo[b]fluoran-
thene), BKF (benzo[k]fluoranthene), BAP (benzo[a]pyrene), DBA (dibenzanthracene), BGP (benzo[ghi]perylene), INP (indeno[c,d]pyrene).
bND-not detected.
cStandard deviations in four repeats were below 0.05%.

A 16 PAH mixture in acetonitrile (SRM 1647c and d, NIST, USA) was used to calibrate the instrument and linear calibration plots (linear detector response) were obtained over the concentration range of interest. Lower quantifiable limits for various PAH, expressed as five times the response factor, ranged 0.7–19.4 pg (Table 1). Repeat injections of the same standard dilution gave coefficients of variation within 3% for all compounds. System blanks on foils and filters averaged from below detection limit to 0.10 ng, for individual PAH, and were < 0.1% of the PAH amounts collected on stages 5–9 of the MOUDI where they predominated. PAH recovery by the extraction method was measured by spiking one half of a particle-laden foil with 20 µl of 10 × dilution of the standard containing known amounts of each PAH. Analysis of both halves, and a mass balance calculation, assuming identical PAH concentrations in particle matter and identical extraction recoveries on both halves, gave average recoveries of 90–99% (Table 1).

The PAH mass measured on each stage was converted to average concentration (mg m⁻³) in the duct by the volume of air pulled through the MOUDI during the time of burn (Table 2). These were converted to mass of PAH (mg) emitted multiplying by the total volume of air (m³) exhausted through the duct (Venkataraman & Rao, 2001) and to the emission factor (mg kg⁻¹) dividing by the fuel burnt. All size distributions are based on ten data points of PAH emission factor from the measured PAH mass on a given impactor stage vs. MMAD of that stage. The PAH size distributions were smoothed using the Twomey inversion algorithm and were inspected to establish unimodality. A least squares best-fit of a log-normal to the inverted data was used to calculate mass median aerodynamic diameter (MMAD) and geometric standard deviation (GSD) with R² of over 0.95 in all cases. The estimated uncertainty in the PAH emission factor (from uncertainties in the MOUDI flow rate, in the duct extraction rate and fuel mass measurement) is about 10%.
Table 2
Summary of emission factors and size distributions of PAH

<table>
<thead>
<tr>
<th>Fuels</th>
<th>Stoves</th>
<th>Number of repeat experiments</th>
<th>Time of burn (min)</th>
<th>Total Emission factor</th>
<th>Semi-volatile species</th>
<th>Non-volatile species</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>$E$(PAH) (mg kg$^{-1}$)</td>
<td>$E_Q$(PAH) (mg (kWh)$^{-1}$)</td>
<td>Emission factor (mg kg$^{-1}$)</td>
<td>MMAD$^d$ (µm)</td>
</tr>
<tr>
<td>Fuelwood</td>
<td>S1</td>
<td>4</td>
<td>26 ± 6$^c$</td>
<td>2.3</td>
<td>1.5</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>S2</td>
<td>4</td>
<td>25 ± 5</td>
<td>2.0</td>
<td>1.3</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>S3</td>
<td>4</td>
<td>30 ± 3</td>
<td>2.3</td>
<td>1.5</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>S4</td>
<td>4</td>
<td>24 ± 4</td>
<td>3.2</td>
<td>1.9</td>
<td>0.8</td>
</tr>
<tr>
<td>Dung-cake</td>
<td>S1</td>
<td>4</td>
<td>31 ± 5</td>
<td>5.5</td>
<td>4.2</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>S2</td>
<td>4</td>
<td>21 ± 5</td>
<td>3.8</td>
<td>2.8</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>S3</td>
<td>4</td>
<td>23 ± 4</td>
<td>3.1</td>
<td>2.3</td>
<td>0.7</td>
</tr>
<tr>
<td></td>
<td>S4</td>
<td>4</td>
<td>25 ± 2</td>
<td>3.9</td>
<td>2.8</td>
<td>0.8</td>
</tr>
<tr>
<td>Briquette</td>
<td>S2</td>
<td>4</td>
<td>19 ± 2</td>
<td>2.8</td>
<td>2.5</td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td>S3</td>
<td>4</td>
<td>20 ± 3</td>
<td>3.0</td>
<td>2.3</td>
<td>0.7</td>
</tr>
</tbody>
</table>

$a$Semi-volatile species include FLT, PYR, BAA and CHR.

$b$Non-volatile species include BBF, BKF, BAP, DBA, BGP and INP.

$^c$Uncertainties are one standard deviation from the mean of four samples.

$^d$Mass median aerodynamic diameter.

$^e$Geometric standard deviation.
3. Results and discussion

3.1. Effect of the fuel-stove system on PAH emission factors

In the following discussion we present PAH emission factors and size distributions from the MOUDI from the PAH mass measured on all ten stages and the after filter. PAH with atomic mass of 202 amu and higher (fluoranthene and higher molecular weight compounds) were analysed as these are likely to predominate in particulate emissions under typical indoor atmospheric conditions. Emission factors of total PAH correlated reasonably with those of particulate matter from each stove–fuel system (Fig. 3).

Emission factors of total PAH ranged $2.0–3.2$, $2.8–3.0$ and $3.1–5.5$ mg kg$^{-1}$, respectively, from combustion of wood, briquette and dung cake (Table 2) and were in agreement with previous studies of similar combustion systems (Gupta et al., 1998; Oanh et al., 1999). These were lower than PAH emission factors from open burning of agricultural biomass in wind tunnel experiments, which ranged $5–683$ mg kg$^{-1}$ (Jenkins, Jones, Turn, & Williams, 1996). Also shown in the table are emission factors in terms of unit useful heat input, $E_{U}(\text{PAH})$ (mg kWh$^{-1}$), the emissions to which the stove user would be exposed for accomplishment of a given cooking task. These are estimated using the mg kg$^{-1}$ value along with measured fuel calorific content and thermal efficiency of the stove–fuel system (Venkataraman & Rao, 2001). Emission factors of total PAH ranged $1.9–3.3$, $4.1–4.8$ and $5.9–10.6$ mg kWh$^{-1}$, respectively, from combustion of wood, briquette and dung cake (Table 2). Based on the mg kWh$^{-1}$ emission factor, dung cake and briquette were significantly more polluting than wood, with regard to PAH ($F = 164.2; p < 0.05$), a trend observed for particulate matter emissions as well (Venkataraman & Rao, 2001). All fuels had a similar content of fixed carbon, volatile matter and CHO (based on ultimate analysis) on a moisture and ash free basis (Venkataraman & Rao, 2001). Nitrogen was marginally higher in dung cake and briquette (2.5% and 1.3%, respectively) than wood (0.3%). However, dung cake and briquette fuels had a significantly higher ash content (31%...
Fig. 4. PAH profiles for wood (a), dung-cake (b) and biofuel briquette (c) all showed a predominance of fluoranthene, pyrene and benz(a)anthracene from all biofuels.

and 19%, respectively) than wood (3%). Since these fuels also had a lower density than wood, the fuel and residual ash filled the combustion chamber and blocked the primary air inlet holes resulting in low oxygen transfer, smouldering combustion and high emissions.

The relative abundance of various PAH species in the particulate emissions is important with regard to carcinogenic or mutagenic risk and also for use as source signatures in models for source apportionment, where convenient inorganic markers may not be available. Individual PAH emission factors were normalised to those of the total PAH to obtain these profiles (Fig. 4a–c). The PAH profiles showed a predominance of fluoranthene, pyrene and
Fig. 5. Emission factors of PAH from dung-cake combustion decreased with increasing power of stove operation (a), combustion temperature (b) and increasing stove thermal efficiency (c).
benz(a)anthracene from all biofuels. The predominance of the lower molecular weight PAH compounds is consistent with previous studies of wood combustion (Kamens, Fulcher, & Guo, 1986; Oanh et al., 1999), and in contrast to fossil fuel combustion, for example vehicular emissions, in which higher molecular weight PAH predominate (Venkataraman, Lyons, & Friedlander, 1994). However, previous studies have measured a predominance of benzo(a)pyrene from wood combustion (Kamens et al., 1986; Raiyani et al., 1993b), which was not seen here. Ratios of the semi-volatile PAH (3–4 ring species) to the non-volatile PAH (5-ring and larger), were significantly higher from combustion of briquette and dung cake (ranging 2.4–8.3), than from wood (ranging 1.4–2.0) (Table 2).

Comparison between stoves for a given fuel, showed PAH emissions from dung cake to be significantly higher from S4 (the two-pot, improved mud-stove) than the other stoves (t-test...
significant at 95%). PAH emissions from wood and briquette combustion were significantly lower from S2 (the one-pot, improved fired-clay stove) than S3 (the one-pot, traditional mud-stove) (t-test significant for all fuels at 90%). This indicates no consistent difference between the traditional and improved stoves, with regard to PAH emissions, and no reduction in PAH emissions from the design improvements, which led to a greater thermal efficiency of the improved stoves.

For dung-cake combustion, trends in the PAH emissions factors were similar to those in CO and particulate matter emission factors (Venkataraman & Rao, 2001). These increased with decreasing combustion temperature, with decreasing power and with increasing thermal efficiency (Fig. 5a–c). However, PAH emission factors from wood and briquette combustion showed virtually no trend with any of these parameters. We expect more complete combustion and lower emissions at higher burn rates and power (from the consequently higher combustion temperature). However, previous studies of space-heating woodstoves (Allen, 1981; Nilson, 1981; McCrillis, Watts, & Warren, 1992) showed that higher burn rates and power may actually
produce higher emission factors of the particulate organic matter (POM) fraction and PAH, indicating complex formation mechanisms of these pollutants. This could explain the lack of correlation between emission factors of PAH and combustion parameters for wood and briquette combustion.

3.2. PAH size distributions

Similar to the PM size distributions, the PAH size distributions from all stove-fuel systems were unimodal with mass median aerodynamic diameters (MMADs) in the submicron range, for both semi-volatile PAH (3–4 ring species) and non-volatile PAH (5-ring and larger). For example, from stove S2, the combustion of wood (Fig. 6a and b), dung cake (Fig. 7a and b) and biofuel briquette (Fig. 8a and b) resulted in PAH size distributions of 0.40–1.01 µm MMADs. These MMADs are about a factor of 5–10 higher than those previously measured in vehicular emissions, which showed a predominance of all PAH, irrespective of molecular weight, in the 0.075–0.12 µm aerodynamic diameter range (Venkataraman et al., 1994). This
would have implications for the efficiency and region of deposition of these compounds in the human lung (Venkataraman & Raymond, 1998).

Mass ratios of PAH to particulate matter, measured on stages 2–10 of the MOUDI, showed no simple dependence on average particle diameter (Fig. 9). PAH are formed, along with soot, from hydrocarbon polymerisation and condensation reactions in fuel-rich regions of the flame, during combustion of the volatilised components of liquid and solid fuels (Toqan, Farmayan, Beer, Howard, & Teare 1984; Nelson & Tyler, 1986) and would be associated with aerosols through adsorption and absorption. If PAH were primarily associated with the surface of the aerosol, then the PAH/PM mass ratio would show a $1/D_p$ dependence (assuming sphericity of particles), and would lie about a line of slope $-1$ on a log–log axis (Fig. 9). The measurements show no explainable trend of the PAH/PM ratio with average particle diameter, corresponding to the 50% cut-point for the given MOUDI stage. This suggests that the PAH were associated with the aerosol through multiple mechanisms including adsorption and absorption. This hypothesis must be further examined through surface characterization methods like PAH photoemission or through particle and PAH formation models for biofuel combustion systems.

4. Conclusions

Emission factors of PAH (total of ten compounds) from combustion of wood, briquette and dung cake, based on useful heat input (mg kWh$^{-1}$), indicate that dung cake and briquette were significantly more polluting than wood, with regard to PAH, a trend observed for particulate matter emissions as well. The PAH profiles showed a predominance of fluoranthene, pyrene and benz(a)anthracene from all biofuels. Ratios of the semi-volatile PAH (3–4 ring species) to the non-volatile PAH (5-ring and larger), were significantly higher from briquette and dung-cake, than from wood combustion. The PAH size distributions from all stove-fuel systems were unimodal with mass median aerodynamic diameters (MMADs) in the 0.40–1.01 μm range, for both semi-volatile and non-volatile PAH. Average MMADs of the PAH size distributions from
biofuel combustion were about 5–10 times larger than in emissions from fossil fuel combustion, as that from automobiles, with potential implications for lung deposition and health risk. The PAH/PM ratio showed no simple trend with an average particle diameter, suggesting that PAH were associated with the aerosol through multiple mechanisms including adsorption and absorption. This hypothesis must be further examined through surface characterization methods like PAH photoemission or through particle and PAH formation models for biofuel combustion systems.

Acknowledgements

This work was supported by a research grant from the Centre for Indoor Air Research, MD, USA (Contract No. 97-12A). We thank Gazala Parween and Tarun Gupta for help with experiments and data analysis. Appropriate Rural Technology Institute, Pune, provided the stove models. Colleagues whose insights have guided this work include Veena Joshi (Swiss Development Co-operation, New Delhi), V.V.N. Kishore (Tata Energy Research Institute, New Delhi), Sangeeta Kohli and M.R. Ravi (Indian Institute of Technology, New Delhi), Priyadarshini Karve (Appropriate Rural Technology Institute, Pune) and Kirk R. Smith (University of California, Berkeley).

References


