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Peer reviewed|Thesis/dissertation

University of California,
Irvine

Emissions from Solid Fuels in Traditional Indian Cookstoves

Dissertation

submitted in partial satisfaction of the requirements
for the degree of

Doctor of Philosophy

in Public Health

by

Robert Michael Weltman

Dissertation Committee:
Professor Rufus Edwards, Chair
Professor Jun Wu
Professor Michael Kleinman
Professor Sergey Nizkorodov

2023

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Dedication

To

Uncle Donald Sarvas
who called me Doctor Robert before I could even dream of it
and in recognition of the endless love and support of my peers, friends, and family

an oath

Whoso saves the life of one person, it shall be as if they had saved the life of all mankind

Quran 5:32

and a joke

Before you start up a ladder count the rungs

Yiddish proverb

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- Edwards, R., Princevac, M., Weltman, R., Ghasemian, M., Arora, N. K., & Bond, T. (2017). Modeling emission rates and exposures from outdoor cooking. *Atmospheric Environment*, 164, 50–60. <https://doi.org/10.1016/j.atmosenv.2017.05.029>.
- Fleming, L. T., Weltman, R., Yadav, A., Edwards, R. D., Arora, N. K., Pillarisetti, A., Meinardi, S., Smith, K. R., Blake, D. R., & Nizkorodov, S. A. (2018). Emissions from village cookstoves in Haryana, India, and their potential impacts on air quality. *Atmospheric Chemistry and Physics*, 18(20), 15169–15182. <https://doi.org/10.5194/acp-18-15169-2018>.
- Weltman, R. M., Edwards, R. D., Fleming, L. T., Yadav, A., Weyant, C. L., Rooney, B., Seinfeld, J. H., Arora, N. K., Bond, T. C., Nizkorodov, S. A., & Smith, K. R. (2021). Emissions measurements from household solid fuel use in Haryana, India: implications for climate and health co-benefits. *Environmental Science & Technology*, 55(5), 3201–3209. <https://doi.org/10.1021/acs.est.0c05143>.

Abstract of the Dissertation

Emissions from Solid Fuels in Traditional Indian Cookstoves

by

Robert Michael Weltman

Doctor of Philosophy in Public Health

University of California, Irvine, 2023

Professor Rufus Edward, Chair

Emissions from solid fuel use play a role both in climate forcing of the atmosphere and in the health of the global population through exposures in homes and contributions of solid fuel use to ambient and regional air pollution. Assessments of the overall impacts from emissions from household solid fuels used in cookstoves at a population level have been hampered by large variations in reported emission factors measured using different approaches with inconsistent conclusions between fuel types. In this thesis, emissions from traditional Indian cookstoves are evaluated for animal dung and brushwood, two of the most commonly utilized fuels in India, using both uncontrolled in-home measurements and minimally-directed cooking tasks performed in rural Indian kitchens.

Following the validation of a moisture probe for use with dung fuels, emissions measurements from both the minimally directed and uncontrolled cooking tests demonstrated a negative correlation between fine particulate ($PM_{2.5}$) emission factors and global forcing using 100-year global warming potentials. While the United Nations Clean Development Mechanism and voluntary carbon markets have featured emissions from cookstoves with a value of approximately 10 million dollars per year in 2020, these measurements that for the first time also include formation of secondary organic aerosol

imply cookstoves are climate neutral at ~10 grams of primary PM_{2.5} per kilogram of dry fuel, which falls squarely in the middle of the range of emissions typically reported in global inventories (5-12 g/kg dry fuel).

Emissions from solid fuels in cookstoves also cause major health burdens in global populations. In 2016, emissions from solid fuels were estimated to cause 2.6 million deaths and 77 million DALYS on a global basis, greater than Malaria, HIV and TB combined. Current global burden of disease approaches, however, do not incorporate the approximately 500 million people who live in homes that primarily cook outdoors. To refine estimates of the health burdens caused by solid fuel use, models were developed to estimate personal exposure contributions from outdoor emissions to allow the incorporation of outdoor cooking into Global Burden of Disease models. Notably, emissions from outdoor cooking can be much higher than those of indoor cooking while still contributing to personal exposures at levels less than the World Health Organization Air Quality Guidelines (WHO AQG) and interim targets due to the greater dispersion of pollutants in outdoor environments.

While particulate matter emissions from solid fuel use are comprised of a wide variety of compounds, many unique to different fuel types, polycyclic aromatic hydrocarbons (PAHs) have been recognized as some of the more toxic constituents of particulate matter emissions, and are associated with cancer endpoints. In these emissions measurements, the ratio of high- to low-temperature combustion was predictive of synthesis of 2-3 ring particulate bound PAHs. Increased elemental carbon (EC) emissions, however, were better predicted by increases in modified combustion efficiency (MCE), since EC emissions reflect the balance between increased PAH synthesis and soot formation which reduces PAH concentrations.

To estimate overall health implications of emissions from these stoves a modified box-model was used to estimate personal exposures to mixed fuels used in the Chulha, which would result in an average of

approximately 2.7 cancers per 100,000 individuals cooking with these fuels. To alleviate these health burdens a suite of interventions are necessary, however, the effectiveness of different interventions approaches has not been systematically evaluated. Meta-analysis of randomized control trials of interventions indicated that use of simple chimneys and hoods were often more effective than the installation of unvented advanced combustion cookstoves in homes.

1.0 Introduction

1.1 Rational

Use of non-woody biomass for fuel, such as dung and crop residues, is prevalent in many areas of the world with approximately 3 billion people living in homes where solid fuels are the primary fuels for cooking⁴. In India a large portion of rural households primarily cook outdoors (9.9% of rural homes and 7.5% of households overall)⁵, and emissions frequently result in elevated levels of aerosols in households⁶, neighborhoods⁴, and urban areas^{3, 4, 7}. The WHO attributes 3.8 million people dying annually from exposure to household air pollution, and the greatest number of individuals dying is attributed to Southeast Asia, where the Chulha is the principal stove used⁸. The Chulha is a traditional U-shaped cookstove usually made with baked clay where fuels are loaded in the front and cookware placed on top. The Angithi is another traditional stove typically used to simmer milk or animal feed and is a bowl shape also made of baked clay where animal dung patties are loaded around the bowl and an ember placed in the center before the large pot is placed over the dung patties limiting airflow. Both stoves utilize small amounts of fire starters, such as plastic bags, to begin combustion. Pictures of the traditional stoves can be found in the supplementary information (Figures S1 and S2).

While there have been considerable efforts to develop protocols and perform laboratory-based testing of stoves using wood fuels, there has been much less (1) testing of unprocessed brushwood or animal dung and (2) development of stoves suited to burning these fuels. Dung from a variety of animals (cows, water buffalo, yak, camel) is used in many parts of the world to cook food and heat homes, particularly in high altitude areas above the treeline and in arid environments where biomass is scarce, but also in agricultural areas of India and Nepal. Emissions from dung burning and properties of dung fuels, however, are not well documented⁹, with only a handful of studies on emission factors from India¹⁰⁻¹⁴.

Limited data indicate that emissions of particulate matter (PM), carbon monoxide (CO), and PAHs from dung were considerably higher than those for fuel wood or woody briquettes¹⁰, and that advanced

combustion stoves may not deliver the benefits anticipated when using these fuels^{11, 12}. Of particular concern to human health are fine respirable particles that can be inhaled and pass through the throat into or beyond ciliated airways to the gas exchange regions of the lung. PM_{2.5}, defined in practice as particles collected by methods with a size-selective cut point to collect particles with a median aerodynamic diameter of 2.5 microns or lower, has most frequently been used as the measure for particulate air pollution. Several reasons exist for the preference for using PM_{2.5}, including availability of inexpensive equipment to make these measurements, reliability using gravimetric methods¹⁵, and well-studied dose-response for several known health endpoints¹⁶. In practice however, approximately 99% of cookstove emissions are smaller than 1 µm, with a mass median diameter of approximately 100 nm¹⁷.

Cleaner cookstoves can have direct health benefits through reductions in pollutant exposures in homes, through reduced downstream ambient pollution (by preventing formation of secondary air pollutants including ozone and secondary organic aerosol), and through reductions in emissions affecting climate, including black carbon and short-lived climate forcing compounds^{18, 19}. Because emissions of particulate and volatile species are dependent on combustion conditions, there is a need to evaluate whether combustion conditions during either in-home or laboratory testing are representative of typical household cooking activities. This additionally highlights the need to develop methods that allow collection and characterization of household emissions measurements that are representative of combustion conditions during typical household cooking activities²⁰.

Evaluating the climate and health benefits of cookstoves can help prioritize policies that maximize co-benefits for near-term climate, human health, agriculture, and the cryosphere²¹. In addition, climate financial incentives, based on emission reduction credits, provides a mechanism to reduce up-front installation costs for clean cooking solutions, allowing them to be competitive with cost-effective health interventions^{22, 23}. While a number of studies have estimated climate and health implications of cookstoves²⁴⁻²⁷, they have been hampered by a lack of emissions data from stoves during normal usage.

Furthermore, few detailed co-benefit analyses have been based on actual measurements of stove performance in-field²⁸. A growing body of evidence has demonstrated substantial differences between laboratory testing and in-field observations²⁹⁻³⁴. There are therefore significant concerns whether climate and health co-benefits estimated from controlled emissions testing represent the reality in homes.

Much of this thesis pertains to work that was conducted in collaboration with INCLIN, the International Clinical Epidemiology Network (New Delhi, India), within the SOMAARTH Demographic Development and Environment Surveillance Site (DDESS) in Palwal district, Haryana covering 51 villages (population approximately 200,000). Most households in the area rely on brushwood and dung as primary household fuels. The entire region is prone to low, ground-level inversions in the winter, resulting in especially high PM concentrations between November and February.

1.2 Moisture assessment of dung fuels

Developing cleaner alternatives for dung fuels is essential to reduce associated health burdens. A first step in this process is to improve the ease with which emissions from this fuel can be measured.

Because emissions measurements are calculated and often normalized on the basis of the dry mass of fuel used, measurement of the moisture content of biomass fuels is critical for estimating emission factors and for accounting for differences between stove performance results from standardized tests, such as the water boiling test¹. Moisture probe measurements have been used systematically for assessment of the moisture content of wood and brushwood fuels by measuring the conductivity between two sharp probes that are inserted into wood. Before this work, protocols stated that the moisture probe should not be used to measure the moisture content of dung, crop residues, or other non-wood fuels, greatly complicating the ease and logistics of measuring emissions. This chapter evaluates if simple moisture probes can be used, with an appropriate calibration, to assess the moisture content of dung patties in relation to oven drying, a standard method for estimating fuel moisture

content, over the range of typical dung moistures used in Indian cooking. The null hypothesis being tested is that there is no difference between probe testing and the gold standard oven drying method, with the alternative hypothesis being tested that there is a linear relationship between these two testing methods.

1.3 In-home and minimally-directed stove testing

Recent chemical characterization of fuel-specific particulate matter emissions¹⁸, and emissions of 76 volatile organic compounds (VOCs)¹⁹ from minimally directed cooking tests in India demonstrated that use of dung patties leads to approximately three times more secondary organic aerosol and ozone formation compared to wood. These studies also found that stove type significantly influenced VOCs, such as benzene and previously unidentified nitrogen-containing organic compounds, in the particulate emissions. This chapter pertains to both in-home and minimally-directed stove testing that was done in Haryana, India and seeks to assess emissions. The null hypotheses being tested are that minimally directed cooking tests, by performing similar cooking tasks using local fuels and fuel mixtures, generate the same emissions and estimates of climate and health co-benefits in these communities as in-home testing.

1.4 Modeling emission rates from outdoor cooking

In many areas of the world cooking occurs outdoors, especially in tropical regions, with an estimated 533 million living in homes that primarily cook outdoors³⁵. Although the number of houses using solid fuel has been estimated³⁶ as a development indicator, for global burden of disease estimates³⁷ the proportion of households cooking outside has not been disaggregated. This is important as the exposure and health implications of stoves operated outdoors are likely to be significantly different to those operated indoors in kitchens, both in the pollutant dispersal and in the amount of time spent in the near vicinity of the stove. Among solid fuel users in Andhra Pradesh in Southern India, kitchen and living room concentrations of PM₄ were higher for homes with enclosed indoor kitchens compared to homes with

outdoor kitchens, which resulted in differences in exposure for both cooks and non-cooks³⁸. Outdoor concentrations of PM₁₀ during cooking in Bangladesh were also substantially lower than those in kitchens and living areas³⁹.

Health impacts of emissions from household fuel use are most closely linked to the exposures they cause. Models that link emissions from cookstoves with indoor concentrations of fine particulate matter (PM_{2.5})⁴⁰ have been useful in evaluating which stove types would meet WHO air quality guidelines in indoor environments⁴¹. These models revealed that even the most recent generation of unvented forced draft biomass cookstoves are still far from reaching emissions levels in controlled laboratory tests that would meet WHO guidelines or interim targets indoors⁴¹. Since the focus was on emissions into indoor environments, outdoor cooking was not considered as part of the WHO indoor air quality guidelines. Based on these estimates, standards for indoor emissions have been incorporated into the International Workshop Agreement (IWA) on tiers of performance¹ as part of the ISO framework as a precursor to International Standards.

In this chapter the importance and abundance of outdoor cooking across the globe is demonstrated. A model is developed for the emission rates from outdoor cooking that would be required to reduce the personal exposure contributions from cookstoves to levels equivalent to the WHO AQG and interim targets, which are based on scientific evidence relating to air pollution and its health consequences. The modeling is performed using distributions of cooking times and in-home emissions rates from Haryana, India as a growing body of evidence consistently indicates laboratory testing using the water boiling test does not reflect emissions during daily cooking activities both in emission rates and particle optical properties^{9, 31, 42-45}. In addition, emissions rates from the forced draft Philips HD4012 stove are modeled to demonstrate the degree to which current more advanced stoves achieve health-based targets such as

¹ <http://cleancookstoves.org/technology-and-fuels/standards/iwa-tiers-of-performance.html>

those proposed by the WHO or individual countries. Finally, since emissions from neighboring houses contribute to elevated ambient concentrations of PM, the distance where individual stove emissions will drop down to $1 \mu\text{g}/\text{m}^3$, and the AQG of $10\mu\text{g}/\text{m}^3$ were modeled. The null hypothesis being tested is that indoor and outdoor cooking produce the same indoor concentrations of fine particulate matter.

1.5 Particulate polycyclic aromatic hydrocarbons

Although some polar compounds play key roles in the health effects of air pollution⁴⁶, polycyclic aromatic hydrocarbons (PAHs) are involved in both malignant and non-malignant diseases of the airways⁴⁷. Mutagenic PAHs, including Benzo[a]pyrene (BaP), have also been shown causing dose-related destruction of ovarian follicles in both developing and developed ovaries, thus affecting reproductive health⁴⁸. Approximations for the atmospheric lifetime of BaP are on the order of several days and modeling suggests that BaP emitted in East Asia is can be transported widely across the region, even reaching the west coast of the United States⁴⁹.

Emission measurements of PAHs in laboratory studies have been reported as not representative of emissions during typical household use⁵⁰, and thus should not be used as inputs for source apportionment, exposure and risk assessment, and chemical fate and transport models. Summed PAH emissions from the Chulha reported here were on the lower end of emissions for traditional Indian cookstoves when compared to other in-field measurements, highlighting the need for research to focus on specific stove-fuel combinations. One common set of PAHs in emission measurements is known as 16PAHs (or when summed as $\Sigma 16\text{PAHs}$) based on its component mix, developed in the 1970s to assess PAHs that are ubiquitous in environmental samples⁵¹. The 16PAHS are acenaphthene, acenaphthylene, anthracene, fluoranthene, fluorene, naphthalene, phenanthrene, pyrene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, benzo[a]pyrene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-cd]pyrene. Reported ΣPAHs , occasionally measuring more than

the suite of 16-PAHs presented here, for household wood combustion span many orders of magnitude (from less than 0.1 mg/kg to over 1000 mg/kg, with summed Σ 16PAHs from \sim 10 – 100 mg/kg), leading to large uncertainties and limited generalizability of findings⁵². In a previous study of wood and dung burning in India, relying on an electric heater to combust the material, emissions of Σ 16PAHs were highest in dung cakes (60 mg/kg) with slightly lower emissions from fuelwoods (44 mg/kg)⁵³. A similar study, relying on a combustion–dilution chamber, reported a 2 order of magnitude span of PAH emission factors for Indian fuelwood (0.4 – 34 mg/kg) and a factor of 2 difference for dung wood (48-98 mg/kg)⁵⁴. Laboratory measurements using standardized burns have reported similar emissions of PAHs from dung and fuel woods^{54, 55}, however in-field measurements of multiple different cookstoves in India using fuel wood and dung cakes showed that stove type and cooking practices strongly influence emissions and that emissions can be highly variable between homes with standard deviations >50% of emission values⁵⁶, as a result of fire tending and other factors that are not well represented by laboratory burns⁵⁷. At the moment, I am aware of only one study that reported in-home measurements of particulate PAHs from a variety of traditional Indian household cooking stoves during household cooking, and no measurements for Angithi stoves, and this study reported particulate PAH emissions for the same suite of PAHs between 70-384 mg/kg for dung cakes and 59-348 mg/kg for fuel wood⁵⁶.

The global burden of disease (GBD) estimates suggest 2.6% of ill-health is directly attributable to indoor smoke from solid fuels, but this estimate is restricted to ALRI (acute lower respiratory infections), lung cancer, stroke and IHD (ischemic heart disease) from fine particulate matter⁵⁸. Because this methodology does not include all potential negative health outcomes due to solid fuel combustion it has the potential to underestimate the total burden of disease. This chapter also extends previous work on modeling indoor and outdoor cooking emissions and provides a quantitative estimate of cancer risk

from field measurements of hydrocarbons from mixed-fuel use (dung and wood combined) on a traditional Indian cookstove – the Chulha.

1.6 Meta-analysis of stove interventions

Household air pollution (HAP) is a major contributor to air pollution exposures and resultant health impacts⁶. HAP contribute to air pollution exposures of the primary cooks as well as other household members. Evaluation of the potential for different interventions to reduce disease burdens from exposure to household air pollution is therefore an intergenerational priority. The use of meta-analysis with a literature-based evaluation of the effectiveness of replacing traditional stoves with more modern ones is an effective tool for evaluating potential stove replacement interventions. Based on previous work modelling outdoor cooking, interventions to vent the smoke out of homes using chimneys can also be effective in reducing exposures and can be used to predict potential reductions in disease burdens of these interventions. The null hypothesis is that the levels of household air pollution are the same before and after a stove intervention – meaning either changing the stove or stove location.

2.0 Probe-based measurements of moisture in dung fuel for emissions measurements

Chapter 2 is reproduced in part from, "Probe-based measurements of moisture in dung fuel for emissions measurements" in *Energy for Sustainable Development Volume 35 pages 1-6* published by Elsevier¹.

Gautam, S., Edwards, R., Yadav, A., Weltman, R., Pillarsetti, A., Arora, N.K., Smith, K.R., 2016. Probe-based measurements of moisture in dung fuel for emissions measurements. *Energy Sustain. Dev.* 35, 1e6. Copyright 2016 International Energy Initiative.

2.1 Introduction

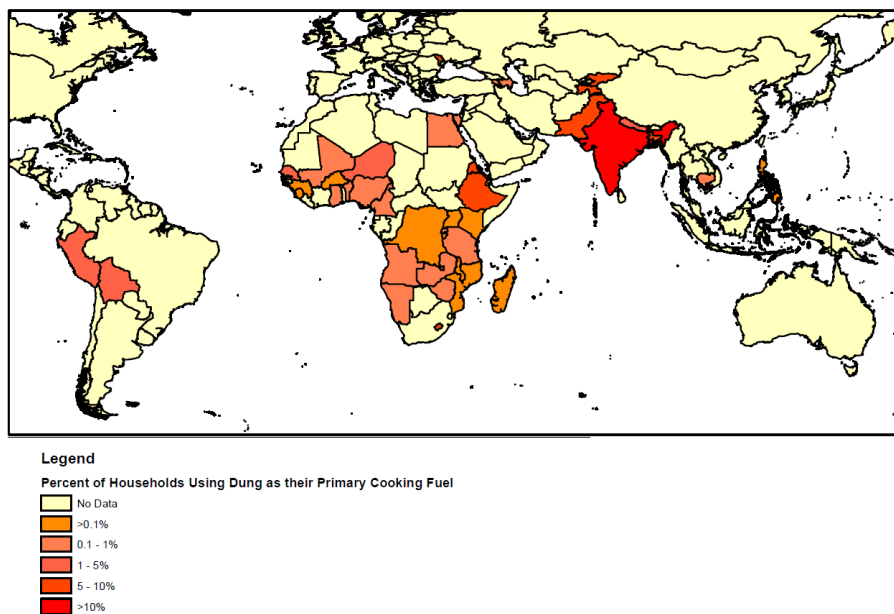


Figure 2.1: Global distribution of dung fuel use.

Figure 2.1 shows the global distribution of dung fuel use in stoves as the primary cooking fuel compiled using the US AID Demographic and Health Surveys (DHS)² Program "STATcompiler" Version 1.5.2 and plotted in ArcGIS, ArcMap 10.3.1. The majority of dung use is in South Asia, with India reporting the largest percentage of total energy use arising from dung combustion and the largest population using dung fuels for cooking. Dung use is also prevalent in Africa, but with much lower frequency, and at high altitudes in Peru. Although dung represents a small fraction of the total energy use in countries where dung use is

² <http://www.dhsprogram.com/>

prevalent, the number of people globally using dung for fuel is large and the emissions are high relative to other fuels. Based on population data for 2014 compiled in the World Development Indicators database³, approximately 178 million individuals live in homes where cooking is primarily done with dung. This chapter is an evaluation of whether simple moisture probes can be used, with an appropriate calibration, to assess the moisture content of dung patties in relation to oven drying, a standard method for estimating fuel moisture content, over the range of dung moistures typically found and used in villages in India where dung is a fuel for cooking.

2.2 Material and methods

2.2.1 Study area

The study was conducted within the SOMAARTH Demographic Development and Environment Surveillance Site (DDESS) in Palwal district, Haryana covering 51 villages (population approximately 200,000) (Figure 2.2, image from <https://commons.wikimedia.org/wiki/File:HaryanaPalwal.png#file>). The cow and buffalo dung patties for the comparison of moisture probe measurement techniques with oven drying methods were collected in Khatela village from August – September 2015. The plastic bags, dung patties, and wood were locally sourced from the village where measurements were done. The uncontrolled field measurements, where dung was collected during cooking, were performed in Manpur, Gehlab, Banchari and Mitrol villages.

³World Development Indicators database, World Bank, 11 April 2016
<http://databank.worldbank.org/data/download/POP.pdf>

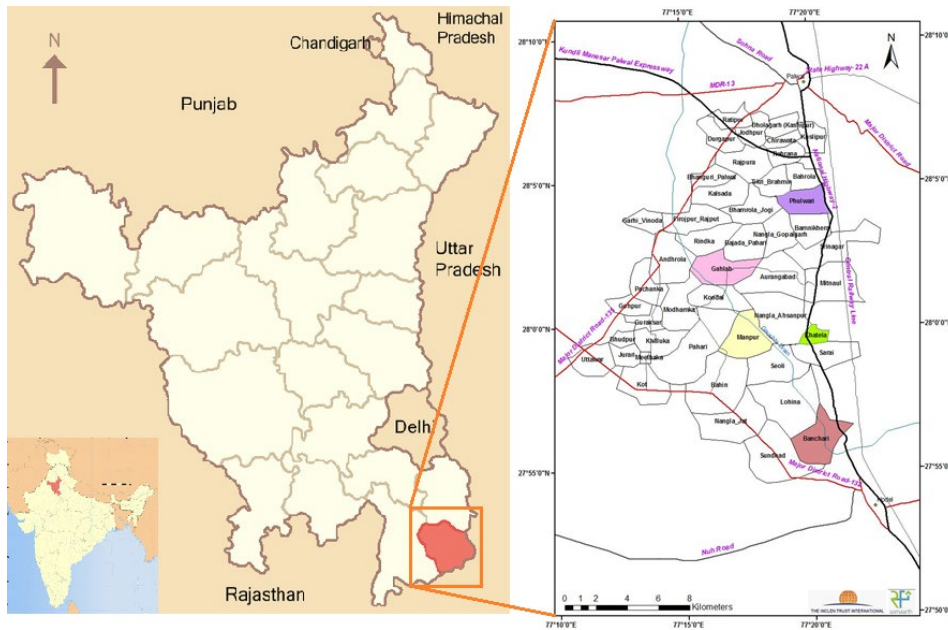


Figure 2.2: Palwal study site with villages where dung patties were collected.

2.2.2 Comparison of moisture probe measurements with oven drying method

The average of 5 individual moisture probe measurements from each of 35 dung patties were compared to moisture measured via oven drying at $103^{\circ} \pm 2^{\circ}\text{C}$. All dung patties were obtained from real homes where dung was collected and manufactured into patties and dried and stored for later use for cooking. Probe-based moisture measurements were conducted immediately in the field using a single digital moisture meter (Model: 50270, SONIN Inc, China) by inserting the probes to their full length into the dung patty (Fig 2.3a). Dung patties were selected to range in moisture content from 5% to 65%, with 5 patties in each 10-point increment, based on initial screening with the moisture probe of 5 measurements across the patty; on the edges, in the center and between the edge and center. After transportation to the field center., a sample of approximately 100 g was then broken off from each dung patty from the edge inward toward the center, and five individual moisture readings were recorded on the broken off portion; on the edges, in the center and between the edge and center. The samples were then wrapped in aluminum foil and kept in a plastic zip-lock bag to minimize moisture and weight loss over time. Measurements for dung

with moisture content of 55%-65% as assessed by the digital moisture meter were repeated to confirm results.

Measuring fuel moisture content by oven drying was performed by weighing each sample before placing on an aluminum tray in a temperature controlled convection oven (Oracle Equipments, India) at $103^{\circ} \pm 2^{\circ}\text{C}$ until mass loss stopped on repeated weighing (24 h approximately) (Figs 2.3b&c).



Figure 2.3ab&c: Moisture measurements: (a) moisture meter with sample, (b) sample arrangement in convection oven, (c) convection oven.

2.2.3 Analysis of elemental composition and energy content

Analysis of elemental composition and energy content were performed by Intertek India Private Limited utilizing ISO 1350 parts 1-4. Ash, along with nitrogen, sulfur carbon, and moisture %, were all calculated as a % of the total weight of each fuel. Approximately 0.2-0.25 g of each sample was burnt in a flow of oxygen and the products were absorbed by suitable reagents and masses determined gravimetrically. Ash was weighed after thermal decomposition in the presence of excess oxygen. Oxides of nitrogen were retained by red lead, oxides of sulfur were retained by lead chromate, soda-asbestos was used in combination with anhydrous magnesium perchlorate to absorb water and carbon dioxide, and concentrated sulfuric acid contained in a bubbler was utilized to indicate the flow of oxygen and prevent

any back flow to the absorbents.

2.3 Results

Table 2.1 shows the means, standard deviations and coefficient of variations (COVs) for moisture content in each moisture-content bin measured with the moisture probe and by oven drying. Values are binned by their moisture content as measured on the probe from the field measurements. Moisture meter measurements were made 5 times on each of the 5 samples in each bin, and oven drying measurements were made for each sample in each bin. The upper bound of our moisture contents, 55-65% as read by the probe, was repeated with 5 additional samples as displayed in table 2.1. In general, between dung patties in bins below 55% measured by the probe, the COV appears to be somewhat lower and more consistent due to the 5 repeated measurements of each dung patty compared to oven drying. Above 55%, the oven drying showed considerable variability in moisture content among the patties selected, which was not reflected in the probe measurements. The discrepancy between the probe and the oven drying were consistent when this bin was repeated to confirm results, indicating the probe measurements are not consistent with the oven drying for values above 55% measured by the probe.

Table 2.1: Variability in moisture contents across dung patties in each bin measured with the moisture probe and by oven drying.

Moisture content, wet basis (%)		05-15%	15-25%	25-35%	35-45%	45-55%	55-65%	55-65%
Probe based	Mean	13.6	23.0	27.5	36.5	52.4	63.4	61.3
	SD	1.7	1.4	1.5	2.4	2.8	0.6	1.1
	COV	0.12	0.06	0.06	0.07	0.05	0.01	0.02
Oven drying	Mean	6.6	9.3	8.6	12.5	15.0	36.1	44.1
	SD	0.4	0.9	2.6	1.0	1.9	11.6	10.0
	COV	0.06	0.09	0.30	0.08	0.13	0.32	0.23

Figure 2.4 shows the reduction in 95% confidence intervals (95% CI) as successive probe measurements are made in individual patties in each bin. In general, the 95% CI increases at higher dung moisture content but are generally consistent at lower moisture contents, with the exception of the 15-25% bin, where the probe measurements were very consistent. It is also apparent that a substantial reduction in the 95% CI is achieved when increasing the number of probe measurements in each patty from 3 to 5, although the difference between 4 and 5 is less apparent. As a result, 5 probe measurements of each patty achieved a good balance between ease of measurement and reduction in 95% CI around the mean.

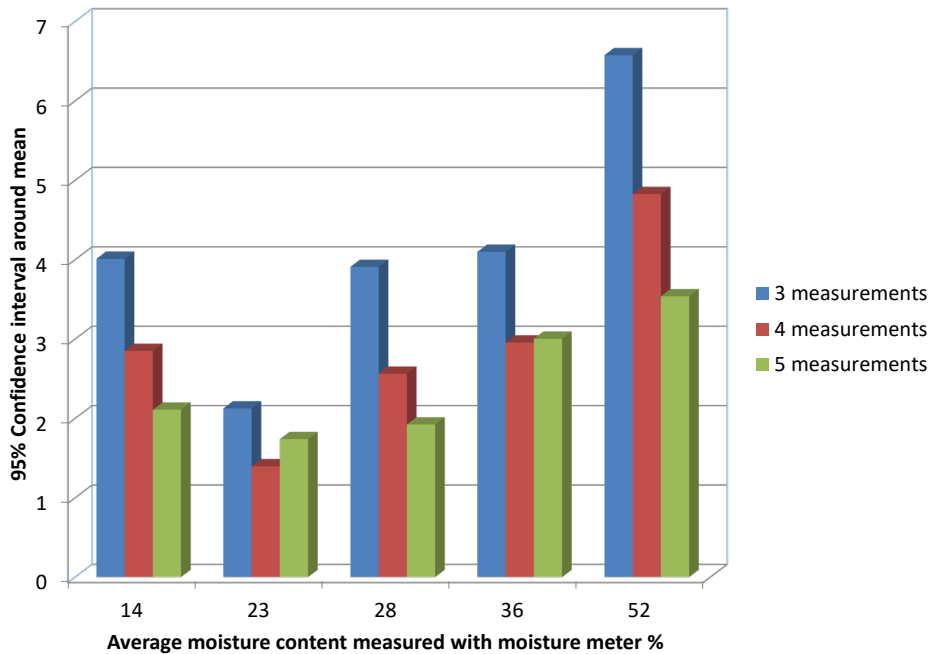


Figure 2.4: Reduction in 95% confidence interval around mean with repeated probe measurements.

Figure 2.5 shows the relationship between the average of 5 moisture probe measurements and the oven drying method to assess moisture content. It shows that moisture probe measurements increase linearly with oven-based moisture content on a wet basis between moisture probe measurements of 5 and 55%, but above 55% the relationship with oven moisture content becomes unreliable. The relationship

between moisture probe measurement below 55% and oven based moisture content on a wet basis is shown in (Equation 2.1).

Equation 2.1: Oven based moisture content as a function of moisture probe readings.

Oven Based Moisture Content (wet basis) = 0.21 * Moisture Probe Measurement + 3.86 $r^2 = 0.76$; $p < 0.001$

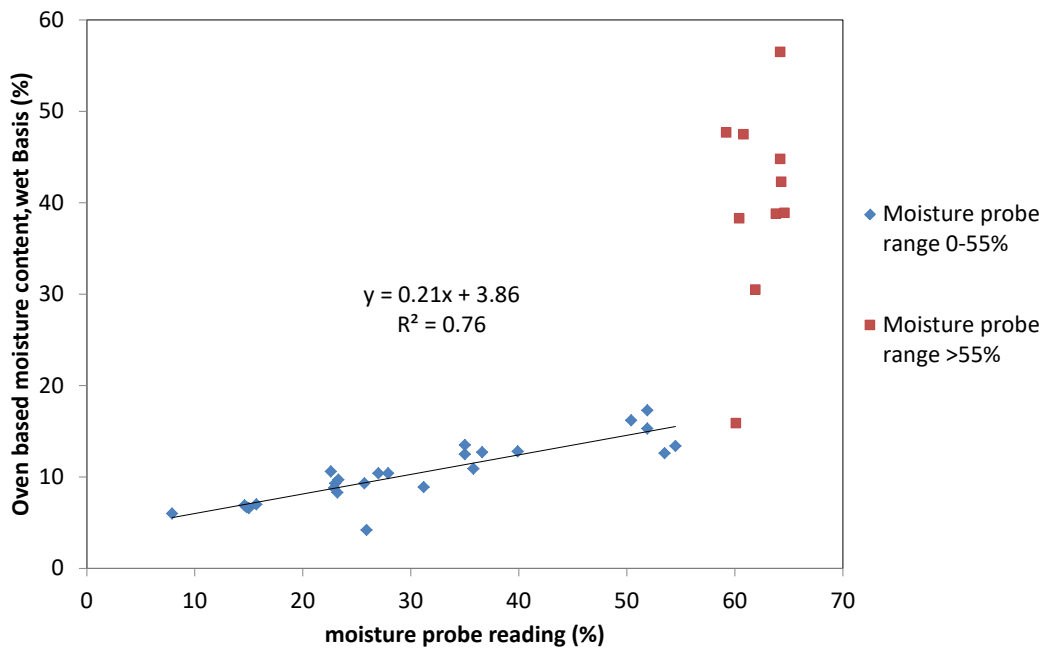
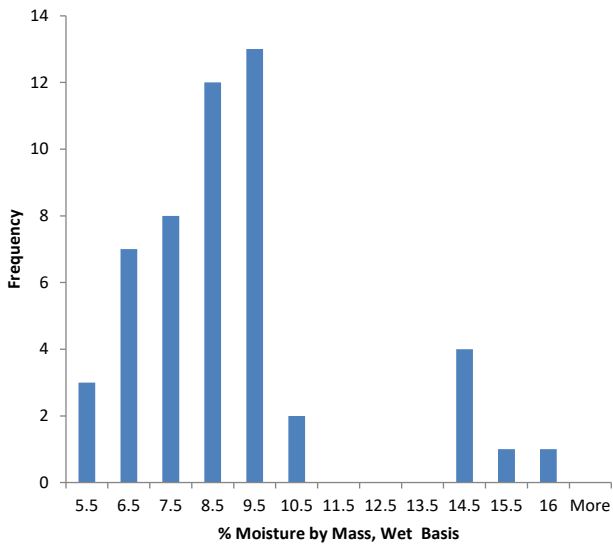
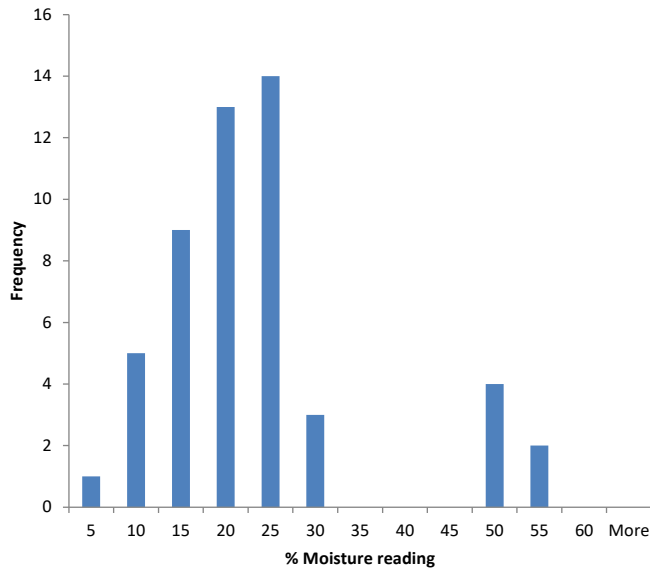


Figure 2.5: Comparison between the average of 5 moisture probe measurements and oven drying methods to assess moisture content.

Figures 2.6a&b show histograms of the average of 3 additional moisture probe measurements of 3 dung patties in 17 homes in 4 Haryana villages measured in 2014 and the corresponding histogram of estimated oven-based moisture content using equation 2.1. All moisture probe readings are less than or equal to the 55% cut off point, with a median of 20%, demonstrating the moisture probe can be used to assess the moisture content in these homes using the relationship shown in figure 2.5. The two highest moisture probe measurements were 51 and 55, which indicates that care must be taken to ensure values do not

exceed the 55% cut off, however, in controlled testing women cooks from the region reported dung patties above 40% were hard to use for cooking tasks and drier patties were preferred.

a) Probe measurements



b) Equivalent oven based moisture content

Figure 2.6a&b: Frequency histogram of 3 moisture probe measurements (a) of dung fuel with conversion to oven-based moisture content using equation 2.1 above (b).

Table 2.2 shows the ash, gross calorific value, sulfur, nitrogen and carbon content of brushwood (shrubs and twigs), fuelwood (stem wood), and dung used in Haryana, India. As expected, dung had a lower calorific value and greater ash content compared to the other fuels. In addition, dung was higher in sulfur relative to the other fuels, but had similar nitrogen content. Fuelwood and brushwood did not differ substantially, although moisture content was more variable in brushwood.

Table 2.2: Ash, gross calorific value, sulfur, nitrogen and carbon content of brushwood fuelwood and dung used in Haryana, India.

SAMPLE	n	MOISTURE	ASH.	GROSS	SULPHUR	CARBON	NITROGEN
		% WT	% WT	CALORIFIC VALUE CAL/GM	% WT	% WT	% WT
Brushwood	5	12.7 (± 7.1)	3.8 (± 1.8)	4243 (± 435)	0.11 (± 0.1)	17.5 (± 2.1)	1.2 (± 0.3)
Fuelwood	3	8.2 (± 0.5)	3.5 (± 0.7)	3898 (± 136)	0.09 (± 0.03)	19.1 (± 1)	1.1 (± 0.4)
Dung	5	10.1 (± 0.7)	29.4 (± 4.7)	2958 (± 351)	0.25 (± 0.02)	15.7 (± 1)	1.3 (± 0.1)

2.4 Discussion

Measurement of the moisture content of biomass fuels is critical for measurement of emission factors and accounting for differences in stove performance results from standardized tests such as the water boiling test⁴. The type, size and moisture content of fuel have a large effect on the outcome of stove performance tests. During the measurement of emission factors, moisture, carbon, and the energy content of the fuel are used to estimate emissions in grams of carbon per kg dry fuel and/or per MJ. Moisture probe measurements are more convenient than oven-drying because multiple measurements can be rapidly performed on site as the fuel is being selected for use. The measurements may also be adjusted for different species and calibrated for different ambient temperatures. However, the current protocols for the water boiling test 4.2.3⁴ state that the moisture probe used to measure wood moisture

⁴ <http://cleancookstoves.org/technology-and-fuels/testing/protocols.html>

content cannot measure the moisture content of dung, crop residues, or other non-wood fuels. As a result, the oven method to determine moisture content must be used, but this presents logistical difficulties in obtaining access to drying ovens and accurate balances in remote locations or to transporting fuels for later testing. The analysis presented here demonstrates that a commonly used moisture probe, when calibrated against oven based methods, can be used to assess moisture content of dung patties over the range of dung moisture typically found and used in villages for cooking purposes. This is an advance in reducing the complexity and cost of making field measurements of emissions in real homes and also improves the ability of testers to rapidly assess moisture content during laboratory testing using the water boiling test or other protocols. Perhaps more importantly, the moisture meter can rapidly assess the moisture of each individual piece of fuel used in the water boiling test, or assess moisture in multiple patties that are used in cooking a meal. The scope and range of such measurements would not be feasible in field studies of many homes due to cost and logistical constraints of performing many oven drying tests. In situations where fuel moisture contents vary widely between individual pieces of fuel, such as in dung patties used in Indian villages, assessment of moisture in multiple patties used to cook a meal therefore allows more accurate assessment of the average moisture content of the whole fuel mixture used for cooking.

The calibration against oven drying here provides a best estimate when oven drying and weighing facilities are not feasible, however, this study is not intended to completely delineate the relationship between moisture probe measurements and oven drying. In particular, the study should be replicated in other geographies both in India and beyond to evaluate the generalizability of these relationships. This is particularly true if the dung patty manufacturing process differs substantially from the process in India or if animal diets are substantially different across evaluation sites. In addition, the relationship likely varies depending on the make and model of moisture probe, and testing should be performed on a broader range of devices to build up a database of coefficients.

Since many studies rely on literature based values for carbon and energy content of wood and dung fuels to compute emissions factors as g carbon per MJ or per kg dry fuel, Table 2.2 shows these parameters for fuelwood, brushwood (shrub) and dung fuel in Haryana India. Overall, the carbon contents were relatively similar between fuels, with dung having the lowest percentage (15.7 ± 1), followed by brushwood (17.5 ± 2.1) and wood (19.1 ± 1). Not surprisingly dung had the lowest energy content (2958 ± 351 cal/g), followed by brushwood (3898 ± 136 cal/g) and wood (4243 ± 435 cal/g). Perhaps more surprising was the increased sulfur content of dung fuels, which when combined with the lower energy content will lead to greater emissions of sulfur based compounds relative to brushwood and fuelwood. As with the moisture analysis, dung composition should be replicated seasonally to examine the impact of animal diets on composition, and also in other geographies both in India and beyond to evaluate the generalizability of these relationships.

3.0 Emissions measurements and implications for climate and health co-benefits

Adapted from: Weltman, Robert M., et al. "Emissions measurements from household solid fuel use in Haryana, India: implications for climate and health co-benefits." Environmental Science & Technology 55.5 (2021): 3201-3209.

3.1 Introduction

This chapter compares uncontrolled, in-home measurements of fuel consumption and emission factors of carbon dioxide (CO₂), carbon monoxide (CO), elemental carbon (EC), organic carbon (OC), and PM_{2.5} during daily cooking events in three villages in Haryana, India, with minimally directed cooking tests in an adjacent village kitchen and also separately to previous laboratory and fuel-based measurements of emissions in India. Evaluations of the climate impact of total emissions from these same stoves including secondary pollutants is presented. A non-traditional cookstove, the Philips HD4012 fan stove, was also evaluated during uncontrolled, in-home cooking to compare to previous laboratory measurements. The results demonstrate that minimally directed cooking tests, by performing similar cooking tasks using local fuels and fuel mixtures, generate representative emissions and estimates of climate and health co-benefits in these communities.

3.2 Methods

3.2.1 Sample selection

For the uncontrolled testing, village homes were identified within the SOMAARTH demographic site in Manpur, Gehlab, Banchari and Mitrol^{59, 60}. Sampling occurred during both morning and evening cooking periods. Cookstoves included Chulhas (traditional Indian mud cookstoves used for cooking), Angithis/haros (two names for similar traditional Indian mud cookstoves, used primarily to cook animal feed, differing only in that haros are fixed in place while Angithis are portable), and the Philips HD4012 fan stoves (a modern, fan-driven, top-loading partial-gasifier stove). No fuels were provided to the

homes as fuels were left up to the individual cooks to provide and homes were selected as a convenience sample in the village.

3.2.2 Minimally directed cooking tasks

A local cook was hired and instructed to prepare a meal with either rice or chapatti (an Indian flatbread) as starch, vegetables, and dahl based on market availability of ingredients for 4 people. Each meal was prepared by the same local cook who determined fuel loading and fire-tending from a load of fuel preselected for fuel type (dung or brushwood or both mixed together) and moisture content (wet or dry). When fuels were mixed, the ratios of dung to wood were chosen by the cook. The cook was also instructed to cook typical village meals rather than specialty meals. No other instructions regarding cooking were given to the cook in order to maximize the cook's ability to cook in their typical fashion. The minimally directed testing were also done in SOMAARTH and were conducted in an outdoor kitchen in the village of Khatela, Palwal, Haryana, India. Palwal District has ~170000 homes in which 39% use wood as their primary cooking fuel, followed by dung (25%), and crop residues (7%)⁶¹. In SOMAARTH, the percent of households using biomass and agricultural residue as their primary fuel for cooking has been estimated at 96.6%⁶².

3.2.3 Fuel assessment

For both the minimally directed and uncontrolled cooking tests, the total mass of each fuel type utilized was calculated by weighing the total fuel of each type before and after each cooking event using a postal scale (Model PE10, Pelouze, China). Fuel moistures were assessed using a 9-volt digital moisture meter for both wood and dung patties (Model: 50270, SONIN Inc., China). Moisture measurements for dung patties were adjusted in accordance with Gautam et al. 2016¹. For the uncontrolled in-home testing fuel selection, meal-type, fuel loading, and fire-tending were determined by the individual cooks (n=5). Two homes were measured twice during uncontrolled testing for a total of 7 meals.

3.2.4 Sampling and analysis

For all testing emissions were sampled and analyzed for CO₂, CO, and PM_{2.5} using established methods⁶³. In brief, three-pronged metal probes were hung above each stove and emissions sampled using PCXR8 pumps (SKC Inc. Universal, Pennsylvania, USA). Simultaneous measurements were conducted in the kitchen yard for determination of background concentrations for subtraction during analysis. Flows were evaluated via a flow meter calibrator (Defender 530, BGI Mesa Labs, Lakewood, CO) during the in-home testing and a flowmeter (TSI 4140, Shoreview, MN) during controlled testing before and after each cooking event. Pumps were turned on before cooking began so that entire cooking events were captured and turned off when cooking was completed. Johnson et al. reported less than a 1% difference between modified combustion efficiency (MCE, the ratio of emitted moles of CO₂ to CO₂ and CO) between sampling hoods and the three-pronged probes used in this study⁶³. Similarly, Zhang et al. also reported no significant changes in emission ratios between flue gas and hood samples⁶⁴. Concentrations of CO₂ and CO were analyzed for all samples using a Q-Trak 7575 (TSI, Shoreview, MN), and adjusted for background ambient concentrations¹⁸.

Size selection of aerosols to collect PM_{2.5}, EC, and OC was achieved using a SCC 1.062 (Triplex) personal sampling cyclone (Triplex, BGI Incorporated, Waltham, MA). Polytetrafluoroethylene (PTFE) filters (PTFE Filter with PMP support ring, 2.0 µm, 47 mm, SKC Inc., Fullerton, CA) were pre and post-weighed on a Cahn-28 electrobalance with a repeatability of ±1.0 µg after equilibrating for a minimum of 24 hours in a humidity and temperature-controlled environment. Five field blanks were collected, by opening filters in the field site and resealing, which had an average mass difference of 0.4±3.1 µg, equivalent to less than 0.1% of average mass deposition of emissions samples and 0.2% of background samples. All sample filters, background and emissions, had a minimum of 109 µg collected material, above the limit detection for the method calculated at 9.3 µg or three-times the standard deviation of the measurement of the field blanks. Quartz filters were collected and analyzed for EC and OC with a Sunset Laboratory

OC/EC analyzer using established methods⁶⁵.

Emission factors (EFs) for gases and PM_{2.5} were determined using the carbon-balance method⁶⁶. In brief, ERs and EFs were determined by multiplying the carbon fraction of each pollutant emissions by the total emitted carbon during the burn. The carbon content of the fuels were taken to be 33.4% for buffalo dung and 45.4% for brushwood fuels based on Smith et al⁶⁶. Carbon in ash was estimated as 2.9% and 80.9% of the mass of char for dung and brushwood, respectively⁶⁶.

Climate impacts were estimated using 100-Year global warming commitments potentials (GWP₁₀₀) for each chemical species as tonnes of carbon dioxide equivalent (tCO_{2e}) per kilogram dry fuel incorporating the fraction of non-renewable harvesting of fuels and are listed as global warming commitments (GWC₁₀₀)⁶⁷. Species included in estimating climate impacts were CO₂, CO, EC, and OC emission factors. In order to convert PM_{2.5} emission factors from water boiling tests (WBTs)⁶⁶ into EC and OC, EFs assumptions on the relationships between organic matter, organic carbon, elemental carbon and PM_{2.5} were utilized in a similar manner to Grieshop et al.²⁷. Elemental carbon was estimated as 21% of PM_{2.5} mass, organic matter estimated as the remaining 79%, and organic carbon estimated as organic matter divided by 1.9 based on the values suggested for fireplace combustion of pine or oak in Roden and Bond⁶⁸. The fraction of the fuel that is from non-renewable biomass were assumed to be zero for dung and taken as 19% for wood based on a reported value for Haryana⁶⁷. GWC₁₀₀ estimations are also limited by assumptions made on GWP₁₀₀ values and the fraction of the fuel that is from non-renewable biomass. By assuming that organic matter is 1.9 times organic carbon it may either over or underestimate the contribution of organic carbon to GWP₁₀₀, as this relationship has been shown to vary between ~1 and 3 depending on the source and age of the aerosol⁶⁸. In order to account for the effects of secondary organic aerosols on climate impacts, total PM_{2.5} SOA was calculated from primary PM_{2.5} emissions. Based on previous modeling work, 1.64 grams of organic carbon SOA were added when calculating GWP for each gram of primary PM_{2.5}⁴. Additional information on equations used for climate impacts can be found

in the supporting information in Weltman et al.²

Statistical analyses were performed with R version 3.3.1 and figures produced in either Microsoft Excel 2010 or R version 3.3.1.

3.3 Results

3.3.1 Emissions

Table 3.1 lists the geometric mean EFs for PM_{2.5}, EC, and OC in grams per kilogram dry fuel and fuel consumption rates for the uncontrolled in-home and minimally directed tests of mixed-fuel use in the Chulha. Values for the uncontrolled and marginally-directed tests are listed as geometric mean (standard deviation). Results for mixed fuels are presented since this was the typical practice in village homes, although use of mixed fuels complicates comparisons with controlled testing, as the majority of results from WBT tests typically use only one fuel type. Overall PM_{2.5} emission rates from minimally directed cooking were on the upper end of the range of uncontrolled emission factors, but no statistically significant differences ($p < 0.05$) were observed for particulate EFs (PM_{2.5}, organic or elemental carbon) or fuel consumption rates between the uncontrolled and minimally directed testing.

While CO₂ emission factors in g/kg dry fuel were significantly higher in uncontrolled testing compared to minimally directed cooking tests ($p < 0.01$), they were not significantly different on a carbon basis (g/kg Carbon), due in part to differences in the ratio of dung: wood in the mixed fuels. Because wood and dung have different carbon contents as a percent of dry weight, as the ratio of dung: wood changes, the total carbon per kilogram of dry fuel is also altered. Thus, relatively minor compositional changes in the ratio of dung: wood lead to differences in g/kg dry fuel not observed when analyzing on a per kilogram of carbon basis. Fuel consumption rates in terms of dry fuel burned g/min were also more highly variable in uncontrolled testing likely due to household size and specific cooking demands, which may

also partially explain why the standard deviations for PM, EC, and OC emissions factors and fuel consumption were higher in uncontrolled testing.

Table 3.1: Geometric mean MCEs and EF consumption rates for the uncontrolled in-home and controlled tests of mixed-fuel use in the Chulha with standard deviations in parentheses alongside differences in the arithmetic mean values and P values from Welch’s two-sided t tests.

	Uncontrolled (n=7)	Minimally directed (n=13)	Difference in Arithmetic Mean	P(T<=t) Two-Tail
MCE	89.2% (1.1%)	86.4% (2.5%)	2.80%	<0.01
PM _{2.5} EF g/kg Dry Fuel	8.7 (7.6)	12.3 (2.5) [‡]	-1.6	0.61
EC EF g/kg Dry Fuel	0.4 (0.5) [‡]	0.6 (0.2)	-0.1	0.69
OC EF g/kg Dry Fuel	3.9 (5.7) [‡]	5.6 (0.9)	0.3	0.91
Dry Fuel Burned g/min	23.5 (9.6)	25.8 (2.3)	-2	0.54

3.3.2 Differences between in-home measurements and WBTs

Table 3.2 and figure 3.1 show a comparison between geometric mean EFs for total suspended particles (TSP) from select WBTs of traditional cookstoves for various fuel types (Smith et al., 2000) and minimally directed in-home field tests for PM_{2.5}⁶⁹. Geometric mean EFs for TSP by both stove and fuel type for both WBTs and minimally directed tests are presented alongside factor differences. The “All Chulha” category includes the cow dung, wood, and mixed-fuel tests. These EFs were generated based on WBTs of traditional Chulha cookstoves⁶⁶. Testing is labeled as MCC for minimally directed cooking tests and WBT for water boiling tests. Data for this figure are presented in Table 3.2. There were 12 mixed-fuel Chulha MCC tests. Differences observed between partially controlled testing and WBT are thus slightly conservative because TSP includes both PM_{2.5} and particles with larger aerodynamic diameters. Previous measurements of mass fractions for burning of biomass in traditional stoves showed emission factors for TSP 17% higher than PM_{2.5} for firewood and 20% higher for dung cakes⁷⁰.

Table 3.2: Factor differences between water boiling tests and in-home field testing of particulate emission factors.

Fuel/Stove Type	Cow Dung in Chulha		Wood in Chulha		All Chulha Tests		Cow Dung in Haro	
	n	Geomean	n	Geomean	n	Geomean	n	Geomean
India measurements from Smith et al. 2000 (g TSP/kg dry fuel)	3	2.2	3	0.6	6	1.2	3	0.5
Minimally directed in Haryana (g PM _{2.5} /kg dry fuel)	15	18.2	14	6.3	41	10.8	10	32.3
Approximate Factor Difference	9		9		9		65	

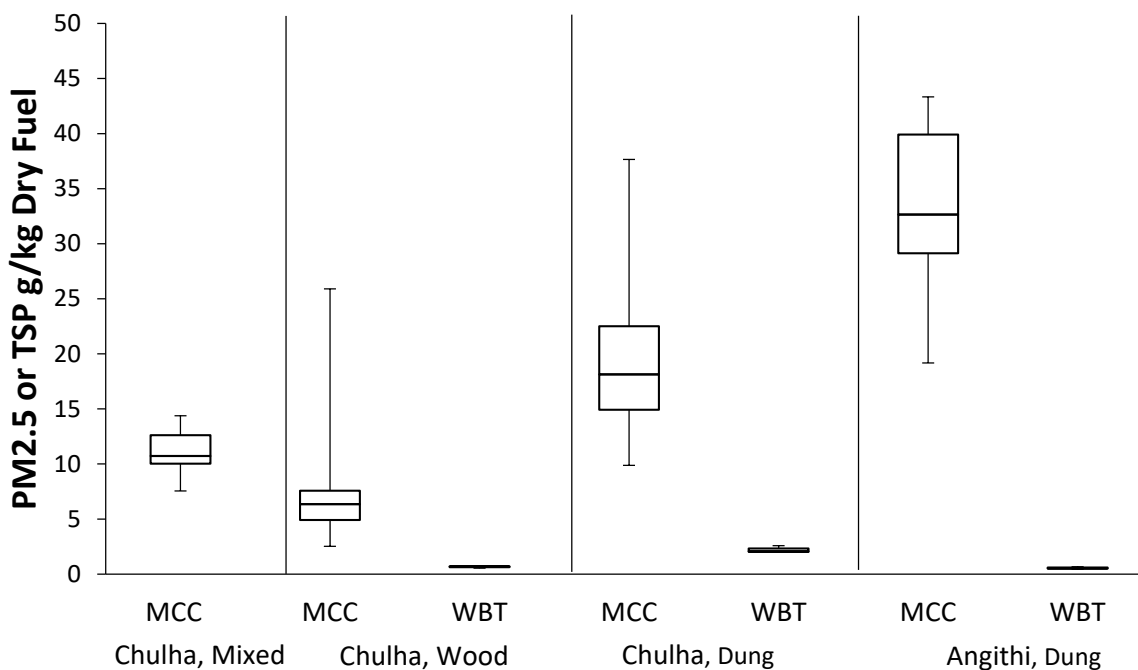


Figure 3.1: Particulate emission factors for both in-home minimally directed cooking tests.

TSP emission factors per kilogram of dry fuel derived from the WBT were a factor ~9 lower than the Chulha across all fuel combinations when compared with minimally directed cooking. Use of mixed fuels or wood resulted in significantly lower PM emissions compared to use of solely dung. Pandey et al. also found an underestimation of PM emission factors by WBT compared to prescribed cooking tests in a rural Indian home by a factor of 2-8³². Similarly, both Johnson et al. and Roden et al. reported a factor of 2-4 difference in particulate matter emissions between laboratory WBTs and field experiments for traditional stoves^{31, 33}. Although factor differences were larger when comparing the Smith inventory and the current minimally directed cooking study, both Johnson et al. and Roden et al. report emission factors of >2 g PM_{2.5}/kg dry fuel for WBTs, which would result in a factor of ~2-3 difference when comparing to the minimally directed cooking. The range of factors observed, suggests that predicting field emissions based on laboratory tests is unlikely to lead to reliable estimates.

Average reported field EFs for Rajasthan fuel wood and Bihar dung were 10.5 (95% confidence interval 7.7-13.4) and 22.6 (14.9-32.9) g kg⁻¹ for the burn cycles, however wood from Punjab had PM EFs ranging from 3-15 g kg⁻¹ (depending on combustion phase) and dung from Uttar Pradesh had PM EFs ranging from 5-28 g kg⁻¹³². The low number of samples for each location (n≤4) precluded detecting any statistical differences between fuel wood types³². Emissions from the Angithi/haro, which is typically used for slow simmering of milk or animal fodder using smoldering dung patties, were 65 times higher in uncontrolled testing compared to WBTs, which may indicate that the test protocol used to perform a WBT created highly uncharacteristic combustion conditions.

Table 3.3 shows a comparison of emissions from the wood-burning Philips stoves in the laboratory and from uncontrolled in-field testing. Laboratory⁷¹ testing is listed as average values for triplicate (or more) measurements of dry wood/wet wood, with the wet wood value as the second entry. EFs are listed as averages in g/kg dry fuel and ERs are listed as averages in g/minute for CO and mg/min for PM.

Emissions of PM_{2.5} per kg dry fuel for the Philips stoves in the current study were substantially higher than those measured during laboratory tests of both wet and dry wood by Jetter et al.⁷¹. Laboratory-based testing of cookstoves utilizing the WBT employed three separate phases of testing; a cold start, a hot start and a simmering phase (Water Boiling Test version 4.2.3). Emissions of PM_{2.5} per kg dry fuel were substantially higher in the current tests compared to the laboratory by factors of 2.4-9.0.

Uncontrolled cooking tests and the wet wood WBTs had similar mean moisture contents (22.7% in the uncontrolled cooking versus 22.1 to 23% in the WBTs) although the variability in uncontrolled in-home testing was much larger as the standard error was 25.6% of the mean for uncontrolled cooking versus 3.1 to 12.3% for WBTs). ER and EF differences between uncontrolled cooking and laboratory testing were smallest for comparisons of the cold-start with wet wood (factors of 0.8-2.4), although significant differences in MCE were observed across all three phases of laboratory testing when comparing to the uncontrolled cooking via Welch’s two-sided t tests (p<0.05).

Table 3.3: Comparison of average emissions from the Philips stove in laboratory and uncontrolled testing.

Philips Stove	n	MCE	PM _{2.5} EF	PM _{2.5} ER	CO EF	CO ER
Uncontrolled Cooking (current study)	7	0.95	3.2	22.3	42.9	0.3
WBT Simmer	3	0.99/0.98	0.5/0.5	2.8/3.3	10.8/21.3	0.1/0.1
Factor differences for Simmering			6.6/6.4	7.9/6.7	4.0/2.0	4.5/2.0
p-Values		0.015/0.033	0.019/0.019	0.031/0.034	0.024/0.081	0.008/0.038
WBT Cold Start	3	0.99/0.98	0.5/1.4	7.3/19.3	10.4/25.8	0.2/0.4
Factor differences for Cold Start			6.9/2.4	3.0/1.2	4.1/1.7	1.7/0.8
p-Values		0.014/0.049	0.018/0.103	0.074/0.790	0.019/0.186	0.069/0.474
WBT Hot Start	3	1.00/0.99	0.4/0.6	6.0/10.5	2.4/11.0	0.0/0.2
Factor differences for Hot Start			9.0/5.2	3.7/2.1	17.9/3.9	6.7/1.5
p-Values		0.007/0.013	0.016/0.027	0.057/0.243	0.007/0.021	0.004/0.134

3.3.3 Comparison to laboratory fuel-burning

PM_{2.5} emission factors from in-laboratory burning of fuel in non-cooking settings by Saud et al. determined using a modified dilution sampler for dung cake and fuel-wood collected from Delhi, Uttar Pradesh, Punjab, Haryana, Uttarakhand and Bihar of $16.3 \pm 2.3 \text{ g kg}^{-1}$ and $4.3 \pm 1.1 \text{ g kg}^{-1}$ for dung and fuel-wood, respectively⁷² were similar to uncontrolled field measurements in Haryana using the same fuels (18.2 ± 7.1 and $6.3 \pm 5.7 \text{ g kg}^{-1}$ for dung and fuel-wood respectively) for the Chulha, but were not reflective of the mixed fuel use typical of homes in the region, and of emissions from Phillips and Angithi stoves showing that fuel tests need to reflect the way in which the fuel is burned in real stoves.

3.4 Discussion

3.4.1 Emission data

Although laboratory testing serves a critical function in evaluating stove design, the use of the results to draw wide conclusions about environmental and health co-benefits of cookstoves can provide misleading information of the relative benefits, as they do not reflect emissions from regular use in real homes. Minimally directed cooking tests in our study villages resulted in emissions that were more reflective of actual usage in real homes than laboratory testing. Emission factors from minimally directed cooking were close to those from uncontrolled tests in these villages and overlapping with those measured by Johnson et al. (2019)³⁴. Previous research has mostly indicated that emission factors for non-CO₂ species increase relative to CO₂ in cookstoves when fuel moisture is increased as a result of increased products of incomplete combustion^{73, 74}, although this effect is not universally true for all stove testing^{68, 75}. Selection of high and low moisture dung patties and/or wood for minimally directed cooking tests, however, did not lead to significant difference in emissions rates although verbal complaints about high moisture patties and compensatory behavior was expressed by the cook during cooking. Matching the moisture content of fuels for testing minimally directed cooking tasks to those

used on a regular basis for that cooking task would likely generate emissions estimates that more closely match those from uncontrolled cooking, and shows promise for testing approaches that would provide more realistic estimates of climate and health co-benefits.

While the minimally directed cooking tests in these villages in Haryana show promise in producing more representative emission estimates, there are a number of limitations. The sample size was limited in our study, villages and a larger number of samples from a wider set of locations would be required for wider applicability, both in India and further afield. In real homes, stove types, usage, and stove maintenance vary. Each of these parameters has significant impacts on combustion conditions, which in turn will change emissions. Use of minimally directed cooking tasks does not inherently capture the wide range of stove types, maintenance, chimney heights, draft characteristics, and variations in operation and tending seen in homes. Further, consideration should also be made for the range of fuels used during different seasonal periods of the year, and the degree of stove-stacking present in homes. Given the widespread presence of stove and fuel stacking in different parts of the world, estimating environmental and health implications of cookstoves by simply comparing results from water boiling tests from one stove to another assuming total replacement will lead to misplaced expectations for stove programs. In addition, incorporating stove stacking into current international emission guidelines for stoves, emission inventories and climate and health co-benefit estimates is a priority. Use of minimally directed cooking tests allows for multiple stoves to be used according to user preferences and may generate more representative measurements of emissions in homes.

Geometric mean fuel consumption rates for the Philips stove during uncontrolled tests in these 3 villages in Haryana utilizing only wood were 6.9 ± 1.4 g/min, which were closer to those seen in the simmering phase of the water boiling test, and were considerably lower than those seen in the cold start and hot start (5.7/6.4 for the simmering using dry/wet wood, 15.5/14.0 for the cold start and 17.5/16.8 g/min for the hot start phase, respectively)⁷¹. Thus, similar to cooking in Michoacan Mexico³⁰, the

majority of cooking involved low-power tasks, and high-power tasks represent a small fraction of total stove usage. For the Phillips in this study, a burn cycle for dry wood where approximately 11% of the fuel was consumed in the cold-start phase and 89% in the simmering phase would achieve equivalent fuel-consumption rates to that seen during uncontrolled cooking, suggesting that task-based emission factors can provide more representative, realistic expectations of climate and health co-benefits for programs that provide alternative stoves.

3.4.2 Climate and health co-benefits

Figure 3.2 shows climate warming potentials and particulate EFs for Chulha stoves (data labeled “Chulha”) in minimally directed cooking tests (data labeled “MCC”) and uncontrolled tests (data labeled “U”) in village kitchens using wood, mixed fuels, and dung. Minimally directed cooking results for the Angithi stove burning dung (labeled “MCC Angithi Dung”) were also included as well as literature values for in-home emissions of 22 traditional Indian Chulhas (labeled “Johnson et al. 2019”), utilizing wood as their primary fuel³⁴. Error bars indicate the standard error of measurements. Particulate matter is expressed as both primary emissions only (top horizontal axis) and as total emissions including SOA mass (bottom horizontal axis). In order to account for the effects of secondary organic aerosol (SOA) on both climate and health co-benefits, SOA was calculated as a function of primary PM_{2.5} emissions. SOA mass was assumed to be 164% of the primary PM_{2.5} emissions’ mass, based on previous secondary organic PM_{2.5} mass concentrations predicted by CMAQ simulations for September 2015 at SOMAARTH headquarters⁴. SOA mass was assumed to be OC for estimating climate benefits. A negative correlation between PM_{2.5} emission factors and GWC₁₀₀ ($R^2 = 0.99$) implies cookstoves are climate neutral for emissions at 9.8 grams of primary PM_{2.5} per kilogram of dry fuel (26 g/kg of PM_{2.5} including SOA). Similarly, for GWC₂₀, cookstoves would be climate neutral at 8.8 grams of primary PM_{2.5} per kilogram of dry fuel (23 g/kg of PM_{2.5} including SOA). In field emissions factors of traditional unvented biomass stoves from inventories average around 7.4 g/kg dry fuel, with a typical range of 5 to 12 g/kg dry fuel⁷⁶,

implying that these stoves may be slightly warming or cooling with values close to neutral at both 20 and 100-year time horizons. Improvements in combustion efficiency through transition to cleaner burning cookstoves will tend to increase warming commitments from these stoves as the organic carbon emissions decrease. Estimates of warming or cooling are not sensitive to the ratio of PM_{2.5} to organic carbon, as previous uncontrolled measurements have found robust linear relationships between ratios of PM_{2.5}:OC, with slopes of 1.29 to 1.35 for a variety of biomass stoves across 174 measurements representing a wide range of fuel types, stove types, flues, altitudes, and cooking locations⁷⁷. Although there are issues with time horizons when using GWP to compare the effects of short-lived and long-lived atmospheric species on climate⁷⁸⁻⁸⁰, warming commitments from these stoves on both time frames are dominated by contributions of short lived climate forcing species OC and SOA.

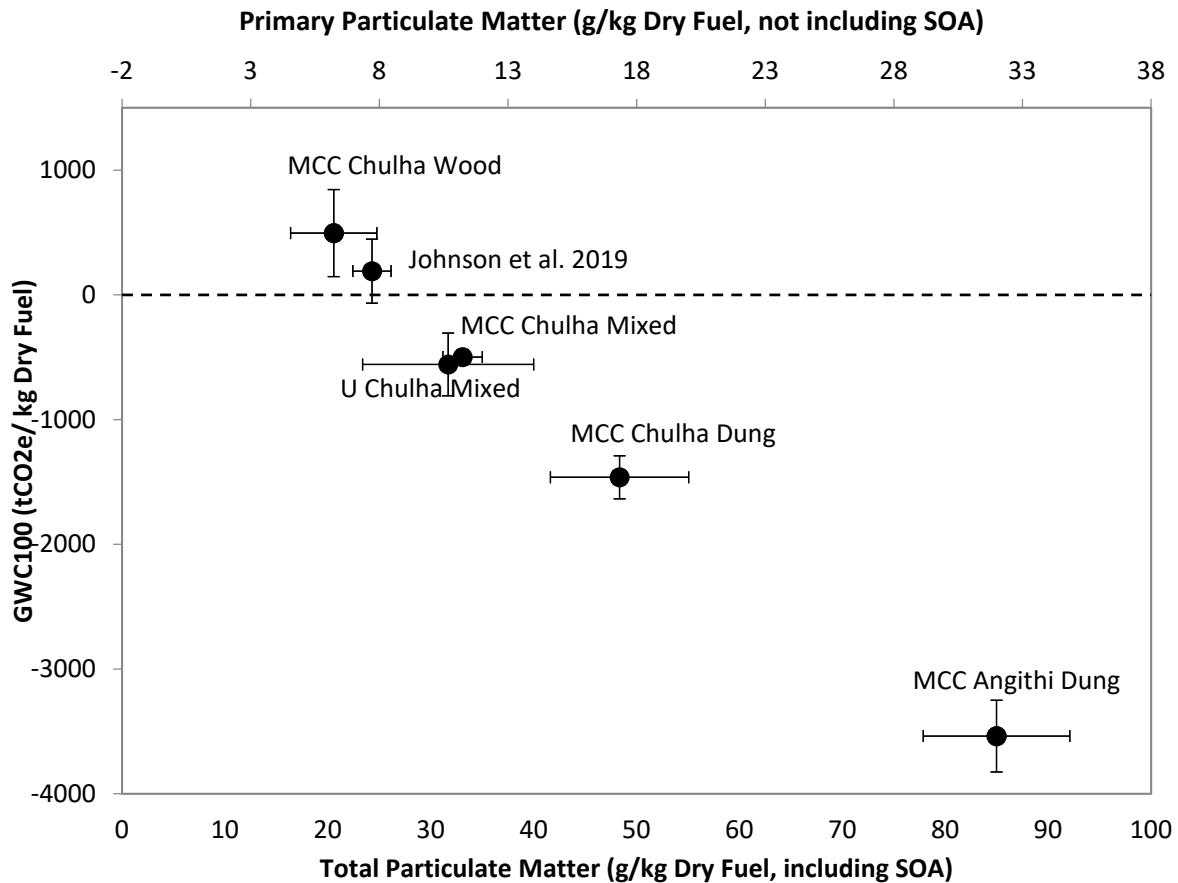


Figure 3.2: A plot of GWC100 versus particulate matter EFs for wood, dung, and mixed fuels.

The fraction of non-renewable biomass harvested and secondary organic aerosol generated in the atmosphere differ between agro-climatic regions, and thus the relative impacts of stoves will vary across regions. Current estimates of the fraction of non-renewable woodfuels have large geographic variations. For example in 2009, while India had seen a net gain in afforestation in recent years, 23-24% of India's woodfuel and 29.6% of Asia and Oceania's woodfuel was harvested unsustainably⁶⁷. Using Asia and Oceania's average fraction of non-renewable biomass of 29.6% would raise the PM intercept for climate-neutral emissions to 10.4 g/kg dry fuel for a 100-year horizon and 8.9 g/kg for a 20-year horizon. Using this cut-off, some uncontrolled field tests of biomass burning stoves in Nepal, Cambodia, and Tibet would imply a net cooling⁸¹⁻⁸⁴. Assuming that SOA formation processes are similar between different regions, the difference between the 29% regional estimates of the fraction of non-renewable harvesting and the 19% for Haryana result in only a modest difference in the intercept between warming and cooling emissions. Large fractions of global emissions, therefore, would be close to climate neutral based on these estimates. Clearly, however, in fuelwood harvesting hotspots the intercept where primary emissions are warming would be higher, which highlights that the climate implications of stoves will depend on the specific communities in which the stoves are distributed. Similarly, where households use different mixtures of fuels the intercept for climate neutral emissions will also vary from those presented here based on the specific fuel mixture present and fraction of renewable harvesting of each fuel. In spite of these limitations, these findings show that when SOA and other climate forcing particulate species are included in estimates, along with regional estimates of non-renewable harvesting, emissions from stoves using biomass fuels are likely to be much less climate warming than previously thought, and some may be climate cooling. Although beyond the scope of the current chapter, this has large implications both for methods to estimate carbon offsets, and for the viability of climate offsets from solid biomass cookstoves, as improved combustion will lead to less PM_{2.5} emissions primarily as a result of reduced OC. While these results cannot capture the full range of emissions, SOA

formation conditions and harvesting from different agroclimatic regions, these findings highlight the importance of calculating global warming from cookstoves including a full suite of climate forcing species including SOA formed after emission into the atmosphere, and also including realistic estimates of the fraction of non-renewable harvesting of biomass.

4.0 Modeling emission rates and exposures from outdoor cooking

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4.1 Introduction

Outdoor cooking was not incorporated into WHO indoor air quality guidelines by definition. Although one of the tiers of performance as part of the ISO framework is for overall emissions from cookstoves, outdoor cooking is not addressed explicitly and overall emissions are not linked to health based air quality guidelines. The tiers for each performance indicator were developed by choosing values of performance for the upper and lower tier boundaries, and then selecting intermediate values. One end of the spectrum is emissions from a three stone fire and the other is emissions from a forced draft stove during a water boiling test. To provide a consistent health based framework for standards relating to overall emissions from cookstoves, there is an urgent need to measure exposures and associated emissions from cooking outdoors. As an interim approach, and to guide future studies, a mechanistic model can outline the plausible health implications from cooking outdoors and other frequently encountered cooking and housing configurations.

4.2 Methods

4.2.1 Prevalence of outdoor cooking

The prevalence of both outdoor cooking (as the most common location for daily use) and solid fuel as the primary cooking fuel in the home were separately plotted in ArcGIS, ArcMap 10.5 over the March 2017 “World Countries” layer package provided by the Environmental Systems Research Institute (ESRI). Both maps were color-coded according to the heat maps included in the legends.

Data were derived from the most recent Multiple Indicator Cluster Surveys (MICS) and Demographic and Health Surveys (DHS) for each country other than China and Mexico. When a country had DHS and MICS surveys published after the year 2000 the latest available survey before 2017 was preferentially utilized in plotting. Solid fuels were defined as charcoal, coconut parts, paraffin, wood, straw, shrubs, grass, sawdust, dung, and agricultural crop residue. Chinese statistics were provided by Dr. Xiaoli Duan from the Ministry of Environmental Protection (MEP) of People's Republic of China⁸⁵. Statistics for Mexico were provided by the “Centro de Investigaciones en Ecosistemas (CIECO)” at the National Autonomous University of Mexico⁸⁶. Countries where outdoor cooking and solid fuel cooking were presumed to be very rare were included in the lowest range of both charts.

When data for solid fuel were unavailable in both the DHS and MICS survey, the estimates from the WHO were utilized⁸⁷. The DHS and MICS survey were used preferentially because the WHO data utilized a multilevel model, rather than survey data, for all but five countries. The WHO data are 39% international multi-country surveys, 18% national census data, 20% from national surveys, “such as household, employment, living conditions, or expenditure surveys”, and the remaining 23% data points are from “other sources, including environmental and poverty assessments, MDG reports, and statistical figures provided on the websites of national statistics bureaus”⁸⁷. Population estimates are based on the World Bank estimates accessed in May 2017.

4.2.2 Modeling emissions rates from outdoor cooking

For outdoor cooking a Gaussian based inverse dispersion model was nested in YASAIw, an Excel-based Monte Carlo simulation tool, to determine emission rates (mg/min) from outdoor cooking that would result in an incremental increase in exposures equivalent to WHO AQG or interim targets. Thus similar to the Box model for indoor emissions⁴¹, the annual mean WHO AQG (10 $\mu\text{g}/\text{m}^3$) or interim targets (IT1 35 $\mu\text{g}/\text{m}^3$; IT2 25 $\mu\text{g}/\text{m}^3$; and IT3 15 $\mu\text{g}/\text{m}^3$) are assumed to be the exposures for which the resultant emissions rates are estimated. Emissions rates that would result in these exposure increases for the

cook were estimated based on cooking two meals during a 24-hour period, which is typical of this region in India. Thus, exposure concentrations [E] for each emissions rate (mg/min) were calculated in equation 4.1 as:

Equation 4.1: Exposure concentrations for outdoor cooking.

$$[E] = AQG / \left(\frac{\text{cooking duration}}{1440} \right)$$

Since exposures from outdoor cooking are a function of the times that cooks are in close proximity to the stoves during the day, a Monte Carlo approach was used to randomly select cooking times from a normal distribution of 51 measurements of cooking times for rice or chapatti meals for four adults in a village setting in Haryana India, where the time in minutes reflects the time the cook was next to the stove when lit. The total time next to the lit stove during the cooking of a meal was on average 69 ± 16.5 min. Thus the time spent cooking next to the lit stove to cook 2 daily meals was on average 138 min (~ 2.3 h), which is similar to the mean of 2.4 ± 1.1 h women cooks (16-60 years old) spent in the kitchen in Andhra Pradesh while cooking ($n=299$)³⁸.

Use of the Monte Carlo approach allows rapid, independent simulation of the duration of a morning and evening cooking event to generate a distribution of 20,000 simulations of emissions rates from outdoor cooking that would result in the incremental increase in exposure equivalent to the WHO AQG or interim target values during a 24-hour period.

Since outdoor stoves in India are commonly placed on the ground and are about 30 cm high, while in other locations stoves may be placed on tables, or platforms and are waist-high, two scenarios were modeled to span the range of these cooking arrangements. The first scenario assumed the stove emission occurred 30 cm above the ground and the receptor (cook) squatting with the breathing zone approximately 1 m above the ground. In the second scenario the stove emission was at 0.9 m high and

the receptor (cook) standing with the breathing zone approximately 1.5 m high. Although these scenarios do not represent a large difference between stove and receptor height, they were chosen to represent the most common cooking behaviors. Table 4.1 shows the other input parameters for the dispersion based model.

Table 4.1: Input parameters for Gaussian-based inverse dispersion model.

Parameter	Value	Parameter	Value
Wind speed [m/s]	0.5	Exit velocity [m/s]	0.5
Receptor distance [m]	1	Smoke temp [K]	700
Stove diameter [m]	0.2	Ambient temp [K]	303

Wind speed in Table 4.1 corresponds to the wind speed in the breathing zone of the cook at a height of 1m when squatting and 1.5m when standing. Based on a logarithmic profile wind speed near the ground would be expected to be lower than wind speed in meteorological data usually measured at a height of 10 m. Thus, for wind speeds of 0.5 and 1.5 m/s in the breathing zone of the cook, the equivalent velocity at 10m would be approximately 0.9 and 2.6 m/s, respectively.

Similar to the box model used to derive emission rates for stoves to meet indoor air quality guidelines and interim targets⁴¹, background ambient concentrations were not incorporated into the model as the objective was to determine emission rates that would result in an incremental increase in exposures equivalent to WHO AQG or interim targets. These concentrations can be readily incorporated based on the local context, however, since background ambient concentrations vary widely, they are not used to derive the emissions rates for stoves for more general application. Similarly, distributions for wind speed could be incorporated, but to be conservative in the estimates of emission rates, a constant wind speed

of 0.5 m/s was used to reflect the low wind speeds present in densely built up village settings in India. Low wind speeds would decrease particulate dispersion and are thus conservative as they result in lower estimates of emissions rates that would increase exposures equivalent to the AQG. Exposures were also conservatively estimated based on the center of the plume, rather than what is typical where people try to avoid being directly downwind of the stove. While people tend to sit outside the plume, and wind speeds are frequently higher than 0.5 m/s, emissions rates were estimated to be protective of health by minimizing the dispersion of the plumes by wind, and represent a worst case scenario of poorly ventilated outdoor spaces.

4.2.3 Gaussian dispersion model

Gaussian dispersion models were used A) in an inverse mode to estimate emissions rates from outdoor cooking that would result in an incremental increase in exposures equivalent to WHO AQG or interim targets, and B) to estimate distance for the plume to disperse as an indicator of the impact on neighborhood pollution levels.

The Gaussian dispersion model was employed in equation 4.2 to estimate the concentration at the receptor located at (x, y, z) :

Equation 4.2: Concentration at receptor site.

$$C(x, y, z) = \frac{Q}{2\pi\sigma_z\sigma_y U} \left(\exp\left[-\frac{1}{2}\left(\frac{z-z_e}{\sigma_z}\right)^2\right] + \exp\left[-\frac{1}{2}\left(\frac{z+z_e}{\sigma_z}\right)^2\right] \right) \times \exp\left(-\frac{y^2}{2\sigma_y^2}\right)$$

Where Q is the pollutant emission rate, U is the near surface wind speed, z_e is the effective source height, σ_y and σ_z are a measure of plume spread in the horizontal and vertical directions, respectively⁸⁸.

The plume parametrizations for σ_y and σ_z calculation were selected to be suitable for modeling near source concentration^{89, 90}.

The effective source height, Z_e , is given in equation 4.3 by:

Equation 4.3: Effective source height.

$$z_e = z_s + z_p$$

Where z_s is the physical source height and z_p is the plume rise due to the buoyancy and initial momentum associated with the smoke exit velocity computed in equation 4.4 as follows:

Equation 4.4: Plume rise.

$$z_p = \left[8.3l_m^2x + 4.2l_bx^2 \right]^{\frac{1}{3}}$$

Where x is the distance downwind of the source, l_m and l_b are the momentum and buoyancy length scales, shown in equations 4.5 and 4.6 respectively:

Equation 4.5: Momentum scales.

$$l_m \approx \frac{w_o r_o}{U}$$

Equation 4.6: Buoyancy length scales.

$$l_b \approx \frac{w_o r_o^2 g \Delta\theta}{U^3 \theta_a}$$

Where w_o is the smoke exit velocity, r_o is the stove radius where smoke exits, U is the ambient wind speed at stove top, g is the gravitational acceleration, $\Delta\theta$ is the temperature excess and θ_a is the ambient potential temperature⁹¹. As the common cooking time in Haryana is early morning and early

evening, the atmospheric stratification is neutral. Unstable stratification typically occurs in the middle of the day, when the heat flux on the ground is high and $dT/dz < 0$. Stable stratification occurs at night when the ground temperature is lower than atmosphere and $dT/dz > 0$. During sunset and sunrise the heat flux vector direction on the ground switches to a neutral condition approaching zero. Thus plume rise parameterization assumes that the plume rises in a neutral boundary layer, and a logarithmic wind velocity profile was used. Wind speed at any given height is given in equation 4.7:

Equation 4.7: Wind speed at different heights.

$$U(z) = \frac{u_*}{\kappa} \ln\left(\frac{z + z_0}{z_0}\right)$$

Where U is the wind speed at height of z above the ground, u_* is the friction velocity calculated based on the reference velocity at the reference height, κ is the Von Karman's constant and z_0 is the ground roughness height. A roughness height of 0.03 m was applied based low vegetation or hard packed yards around village homes which is typical for village scenarios in Haryana India, based on Blocken 2015⁹².

4.2.4 Emissions sampling

To estimate the distance for the plume to disperse as an indicator of the impact on neighborhood pollution levels, particulate emissions rates were measured during normal daily cooking tasks in homes in three villages within the SOMAARTH demographic site in Haryana India with 16 homes using Chulhas, seven using Angithi, and 13 using forced draft Philips stoves. Sampling consisted of two meals one in the morning and one in the evening. The Philips forced draft advanced combustion stove was designed primarily to burn woody biomass and utilizes a battery-powered blower to maintain constant flow into the combustion chamber.

Emissions for each stove test were collected directly above the stove using a three-pronged aluminum sampling probe⁹³. A simultaneously collected background sample was used to correct emission factors for dilution with background concentrations. 37mm Teflon filters were inserted in-line to determine PM

emissions. Filters were equilibrated for 48 h at $45\pm 3\%$ relative humidity and $20\pm 2^\circ\text{C}$ before taking pre and post weights on an electro-microbalance (Cahn Model 29, Thermo Electron Corp., USA). No mass adjustments were necessary based on field blanks. Flows were evaluated via a flowmeter (Mesa Labs Defender 530) before and after each cooking event. Pumps were turned on before cooking began so that entire cooking events were captured and turned off at completion of the burn cycle.

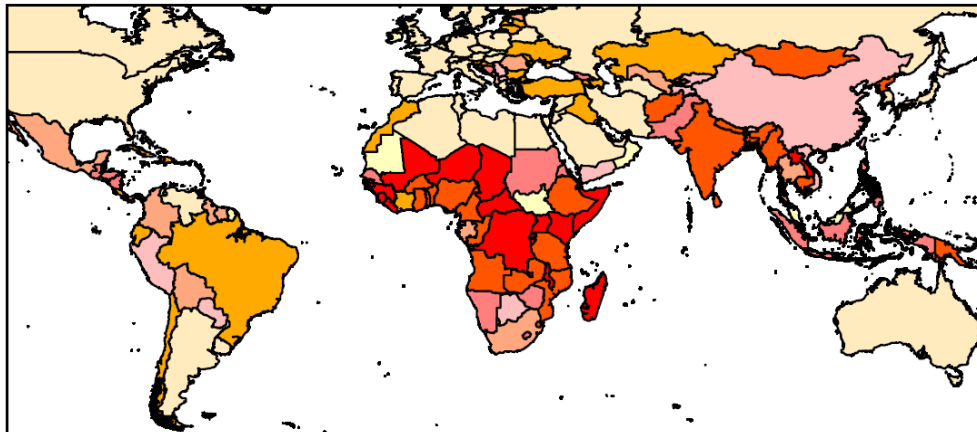
Emission factors were determined using the carbon balance method, which accounts for the fate of the fuel carbon in the emitted species⁹⁴, and has been used frequently for similar studies⁹⁵⁻⁹⁷. Briefly, the ratios of each emission species (as carbon) in the sample to the total carbon in the sample were multiplied by the total emitted carbon to derive emission factors. Carbon content for PM was derived from analysis of elemental and organic carbon of particulate matter on quartz filters performed at the University of Illinois. Fuelwood weights were measured using a digital hanging scale with a 10 g resolution (American Weigh SR-20) over the course of the cooking event. Ash was weighed in a metal pan after taring on an Accuteck Digital Postal Scale (W-8580-110-Black) with a 2.8 g resolution. Total fuel carbon was determined by weighing the fuelwood consumption and adjusting for water content and carbon diverted to ash. Water content was measured with a digital moisture meter (Model: 50270, SONIN Inc, China) and ash was measured after completion of cooking. Moisture measurements for dung patties were adjusted based on oven based drying methods¹.

4.3 Results

4.3.1 Outdoor cooking prevalence

Figure 4.1 shows the prevalence of solid fuel use and figure 4.2 the global prevalence of outdoor cooking. The 95% confidence interval reported by the WHO for the percentage of the world population utilizing solid fuels as their main cooking fuels in 2010 was 37-44%. Utilizing the DHS and MICS data combined with WHO data suggests that 42% of the 2017 world population (2.94 billion individuals)

utilize solid fuels as their main cooking fuels. Outdoor cooking, as expected, appears to occur more frequently in equatorial and subtropical regions where ambient temperatures are higher. Although outdoor cooking is less frequent than indoor cooking it still remains an important fraction of global cooking supplying an estimated population of 533 million people.



Legend

Prevalence of Solid Fuel as Primary Cooking Fuel

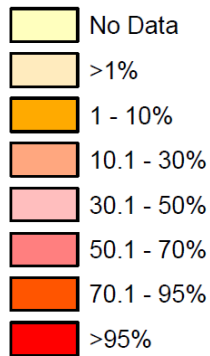
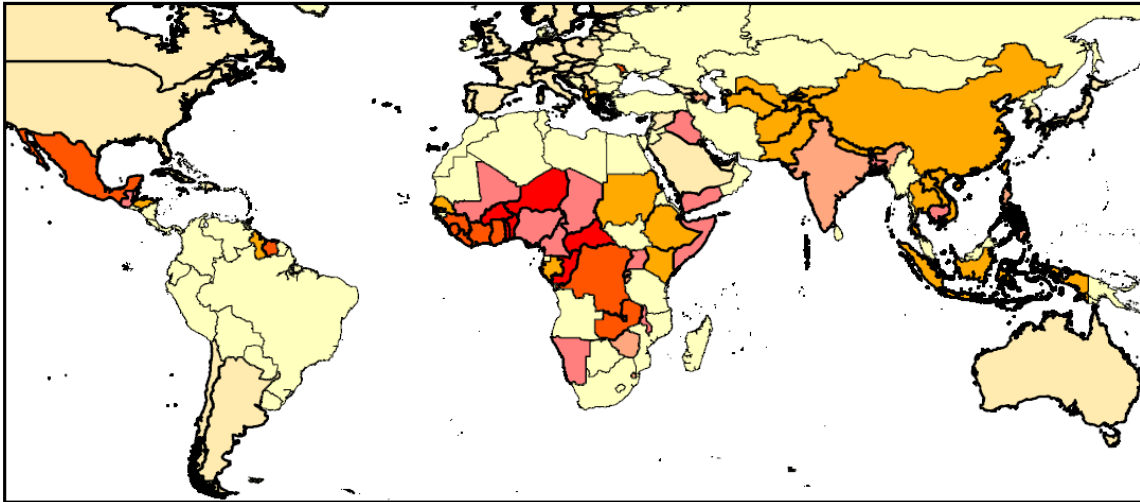


Figure 4.1: Prevalence of solid fuel use for primary energy provision for cooking.



Legend

Prevalence of Outdoor as Primary Cooking Location

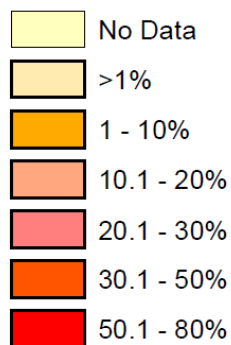


Figure 4.2: Prevalence of outdoor cooking as the primary cooking location for the household.

The available data for outdoor cooking include 85 countries and represent almost two-thirds of the world's population (4.82 billion individuals, 65.5%) of which approximately 11% (533 million individuals) live in homes where cooking is done primarily outdoors.

MICS survey data, including surveys that were not utilized as a more-recent DHS survey was available, cover 939 million individuals (~13% of world population) of which approximately 14% live in homes where cooking is done primarily outdoors. The DHS survey data, including surveys that were not utilized as a more-recent MICS survey were available, cover 2.92 billion individuals (~40% of world population) of which approximately 18% live in homes where cooking is done primarily outdoors. Within the 21

countries where both DHS and MICS survey data were available (629 million individuals), DHS data indicate 25% and MICS data indicate approximately 22% live in homes where cooking is done primarily outdoors. Although survey questions ask whether cooking is usually done in the house, in a separate building, or outdoors, seasonal changes in meteorology such as the monsoon rains may change the cooking locations for during periods of the year depending on the precipitation condition suited for outdoor cooking in a given geographic region, which is not captured in survey data.

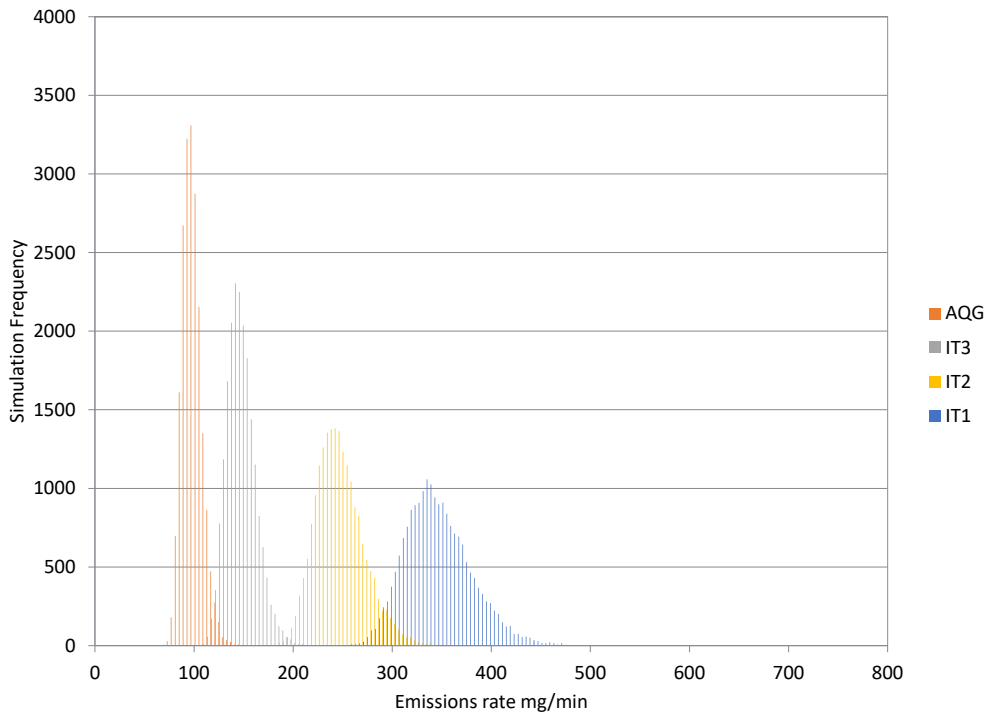
The available data for solid fuel cooking include 154 countries and represent data for over 6 billion individuals (~82% the world's population), of which approximately 49% live in homes where they cook primarily with solid fuels. Including the 36 high income countries in which WHO assumes solid fuel use to be negligible translates to a 41.5% global prevalence of global solid fuel use covering 96% of the global population. Within the 28 countries where both DHS and MICS survey data were available for solid fuel use (821 million individuals), DHS data indicate approximately 62% and MICS data indicate approximately 73% live in homes live in homes using primarily solid fuels. The remaining 980 million individuals in 57 countries were covered by the WHO data, of which 150 million (15%) individuals live in homes using primarily solid fuels. Estimates for populations living in homes that cook primarily with solid fuels differ by 3% in countries that had both DHS and WHO data (66% and 62% respectively); 8% in countries that had both MICS and WHO data (65% and 57% respectively), and <1% between the combined MICS and DHS data set used for mapping in the current study and WHO data (49% vs 48% respectively).

4.3.2 Modeling emission rates from outdoor cooking

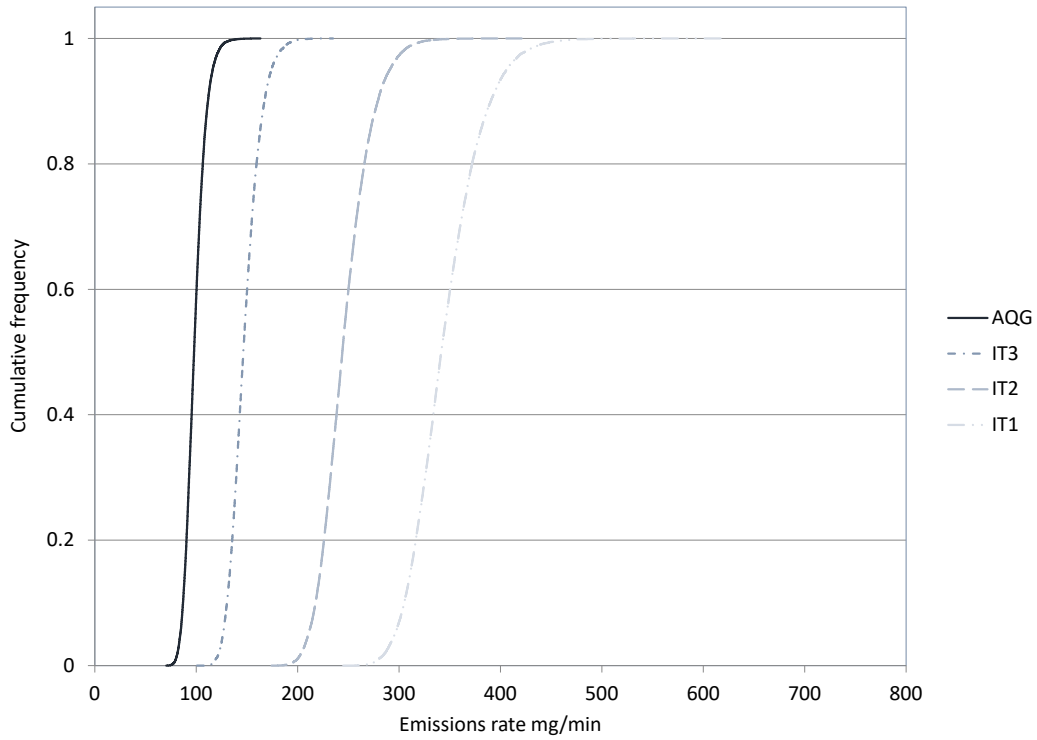
The models for outdoor cooking while squatting and while standing were the product of random selection from the distribution of cooking times for a morning and evening cooking event over 24 h with 20,000 simulated runs. The model then calculated the emission rate from the stove (mg/min) for each of the

simulated runs that would result in an incremental increase in personal exposure equivalent to the WHO AQG or interim targets during a 24-hour period. The output distributions in Figure 4.3 and Figure 4.4 therefore represent the distribution of emissions rates from a stove that would result in incremental increases in personal exposure equivalent to the WHO AQG or interim targets during a 24-hour period. Simulations are done based on the cook standing that would result in incremental increases in personal exposure equivalent to the WHO AQG or interim targets during a 24-hour period. Simulations are presented as either histograms(a) or cumulative frequency plots(b). Table 4.2 shows summary statistics of the output distributions of emissions rates that are relevant to standards and guidelines for stoves that emit pollutants outdoors.

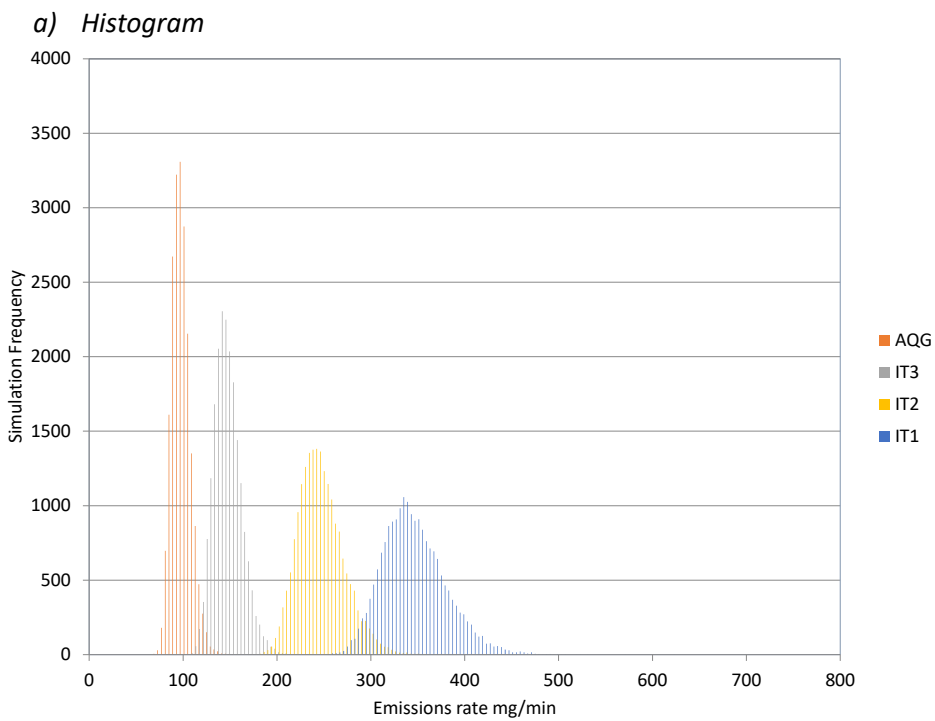
a) Histogram



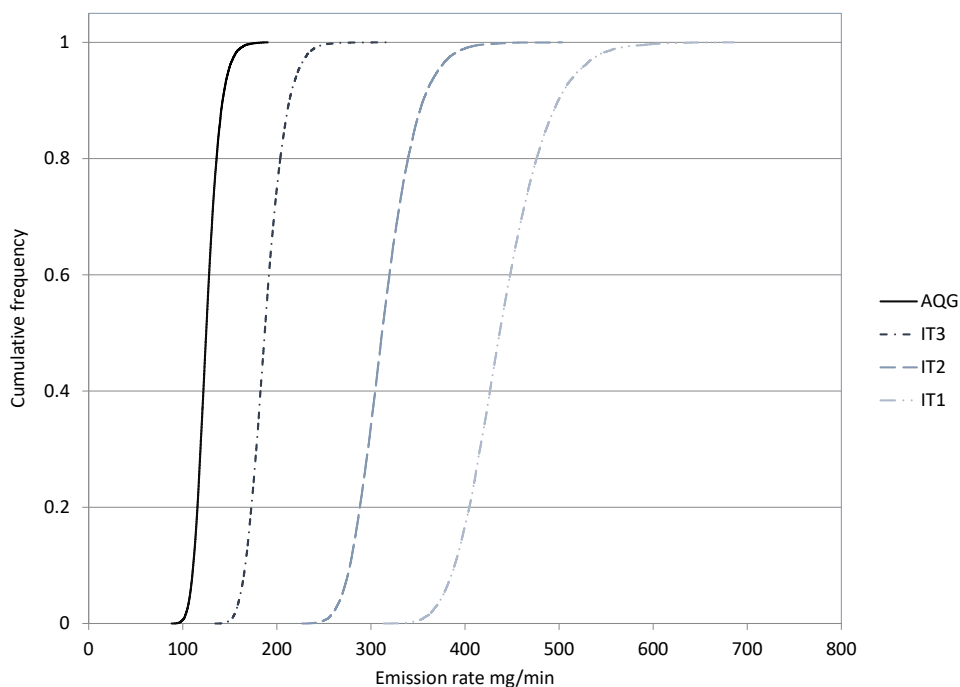
b) Cumulative frequency



Figures 4.3a&b: Monte Carlo simulation of emissions rates from outdoor cooking while standing.



a) Cumulative frequency



Figures 4.4a&b: Monte Carlo simulation of emissions rates from outdoor cooking while squatting.

Table 4.2: Emissions rates from outdoor cooking to increase exposures equivalent to air quality guidelines and interim targets.

Exposure concentration	stove/cook height (m)	Emission rate(mg/min)								
		Mean	St Dev	5%	10%	25%	50%	75%	90%	95%
AQG	0.3,1	126	13	108	111	117	125	134	142	148
IT3	0.3,1	189	19	161	166	176	187	200	213	222
IT2	0.3,1	315	31	269	278	293	312	334	355	371
IT1	0.3,1	441	44	377	389	411	437	468	499	520
AQG	0.9,1.5	99	10	84	87	92	98	105	112	116
IT3	0.9,1.5	148	15	126	130	138	146	157	167	174
IT2	0.9,1.5	247	24	211	218	229	244	261	279	291
IT1	0.9,1.5	345	34	296	305	321	342	366	390	406

4.4 Discussion

In households cooking with solid fuels the location where the cooking occurs significantly impacts the exposures to the cook and to other family members, and thus the potential for adverse health impacts from exposure to the smoke. Cooking outdoors results in significantly different exposures to the cook and to family members compared to indoor cooking largely because the smoke disperses more readily

outdoors compared to unvented indoor cooking³⁸. Exposures for family members that are not actively cooking are also reduced since smoke does not transfer within the household as is seen when kitchens are connected to other rooms in the household³⁸. The location where cooking occurs therefore impacts which emissions rates are likely to pose a health issue. Of current IWA performance guidelines for stoves, only the indoor emission tier derived using a box model for indoor kitchens is based on health endpoints^{40, 41}. While there are tier guidelines for overall emissions from a stove, these are not related to health. The analysis presented here is a first step in modeling emission rates relevant to health endpoints for one of the more common cooking arrangements found globally. Globally outdoor cooking with solid fuels in cookstoves is a significant fraction of the total solid fuel use in cookstoves supplying an estimated population of 533 million people, or approximately 18% of the 2.94 billion individuals who cook primarily with solid fuels. Understanding the exposure patterns of these individuals is therefore important in understanding the overall impacts of the use of solid fuels for cooking globally. In addition, modeling emissions rates should be expanded from a single room house to cover some of the more common housing configurations and kitchens.

Emission rates modeled for outdoor cooking (Table 4.2) are substantially higher than IWA emission rates for indoor cooking because the models estimate impact of emissions on personal exposure concentrations rather than microenvironment concentrations, and because the smoke disperses more readily outdoors compared to indoor environments. In contrast emissions remain in indoor environment for longer periods until ventilation rates reduce concentrations back to ambient levels. Emission rates for WHO indoor air quality guidelines use a Monte Carlo probability analysis of the number of homes that meet guidelines and interim targets, and focus explicitly on cookstove emissions and the resulting indoor air pollutant concentrations in kitchens or single room houses⁴¹. IWA emissions rates rely on a more simplified assumption of 30 m³ room and a rate of 15 air exchanges per hour. WHO guidance allows for small single room homes where fires are lit most of the day and cooks and infants are exposed for extended periods.

Typically however personal exposure concentrations are lower indoor kitchen concentrations due to the time spent away from the proximity of the stove ⁹⁸. Use of personal exposure concentrations as a basis for estimating emissions rates to meet AQG or interim targets would allow for substantially higher emissions rates from stoves. In smaller homes where the stove is not separated from the main living areas, the contribution of the cookstove to exposures will be substantial as mothers with young infants frequently spend a large fraction of their day inside at home. For kitchens separated from the living area by a wall or partition the contribution to exposure reflects the time spent in proximity to the stove during cooking and while preparing food. In contrast the dynamics for outdoor cooking tend to be somewhat different, and exposure contributions reflect the time in close proximity to the stove. In this case a focus on the contribution of time near the stove to personal exposures, rather than ambient concentrations is more relevant, as pollutant concentrations change quite rapidly with distance from the stove even at low wind speed. Guidance for the WHO AQG notes that to be related to health, air pollutant concentrations should be representative of exposures⁹⁹, and thus this approach is taken for outdoor cooking.

Another key consideration for linking emissions sources with exposures and air quality guidelines is that exposures reflect emissions from a variety of sources needed to meet daily cooking and lighting needs, including other household sources such as tobacco smoke, mosquito coils and incense, and exposures in other indoor and outdoor environments, such as during transportation. The models presented here therefore represent the contribution of stoves to personal exposures rather than the total exposure. While the supralinear nature of the dose response curves implies that the health implications of a given emission rate depend on the other sources that contribute to personal exposures, in practice ambient concentrations vary substantially by region¹⁰⁰ and contributions from other sources are too numerous and varied to be practical for modeling guidance in emissions rates from outdoor cooking. Thus in a manner similar to box models to evaluate emissions rates for indoor cooking⁴¹, the models presented here focus on the contributions of outdoor cooking to personal exposures and to neighborhood pollution levels.

Exposure contributions of the stove will also depend on the precise position of the cook relative to the emissions and shifts and changes in wind speed and direction. Exposure contributions are modeled in the center of the plume, although in practice people are likely to avoid standing or sitting in smoke where possible. Thus, the estimates of emissions rates in Table 4.2 are likely conservative, and more protective of health.

In indoor environments for stoves to meet WHO AQG guidelines, emissions rates are lower than the best performing solid-fuel biomass stoves, which make use of fans and/or gasify the solid fuel before combusting the resulting gases⁴¹. Since emissions performance from daily cooking is often worse than that observed in controlled water boiling tests, the fraction of stoves in indoor environments that meet AQG is likely even lower. In contrast, many more stoves would meet AQG when cooking outdoors. Since outdoor cooking represents a significant fraction of global cooking, emissions guidelines should better represent the different indoor and outdoor contexts in which stoves are being used, especially given that in these contexts the best performing solid fuel stoves have the potential to provide significant benefits.

That many more stoves would meet AQG when cooking outdoors also highlights the significant benefits of venting of stoves outdoors, or separating the cooking areas from the living areas as a separate room or with physical boundaries or partitions to reduce exposures to cooks and family members^{38, 101}.

Although ambient concentrations are elevated by the number of other homes in close proximity, contributing to a localized neighborhood pollution effect, the reductions in exposures to the cook and family members are substantial. To better quantify the impacts of outdoor cooking emissions rates on neighborhood pollution levels figure 4.5 shows outdoor cooking emissions rates and Gaussian dispersion modeling of the distance from the stove when emissions are diluted to an air concentration of $1 \mu\text{g}/\text{m}^3$ for 4 different wind speeds 0.5 m/s, 1.0 m/s, 1.5 m/s and 2 m/s. The relationship between emission rate and distance is close to linear consistent with neutral stratification.

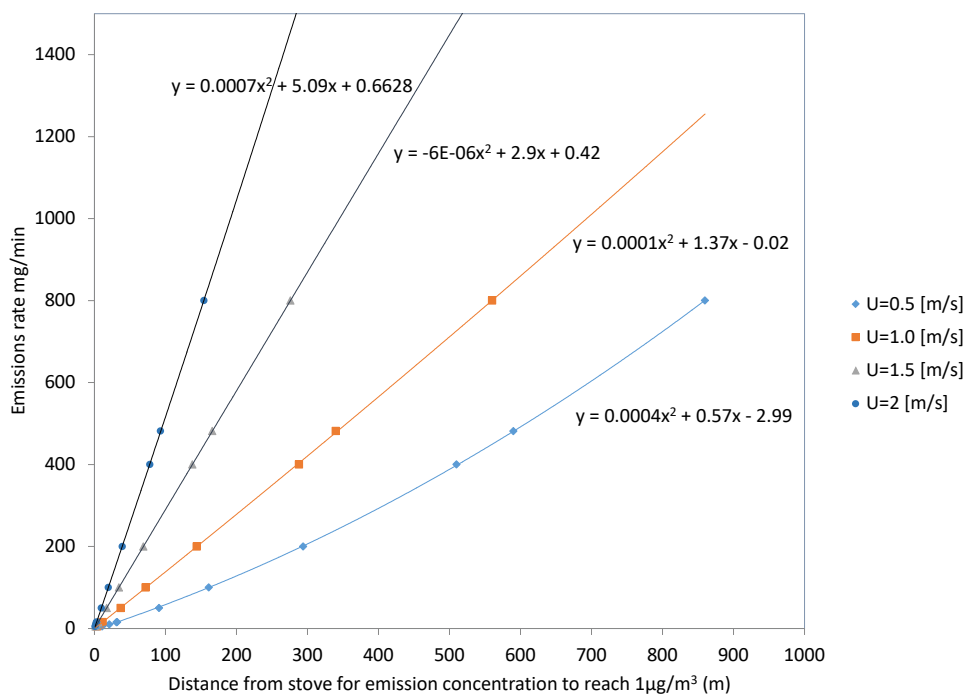


Figure 4.5: Gaussian dispersion modeling of distance from the stove before outdoor cooking emissions are diluted to a PM_{2.5} concentration of 1 µg/m³.

Table 4.3: Emissions rates, modified combustion efficiency, and distance before plume reaches 1 µg/m³ for stoves in Haryana India¹⁰².

Stove Type	Fuel	n	Time (min)	MCE	PM _{2.5} (mg/min)	U=0.5 [m/s]	U=1 [m/s]	U=1.5 [m/s]	U=2 [m/s]
Angithi/Haro	Dung	5	143 (± 52)	0.87 (± 0.02)	302	407	217	104	59
Chula	Dung + wood	12	169 (± 52)	0.91 (± 0.02)	424 (± 395)	163	78	37	21
					108				
Philips	Dung + wood	5	238 (± 139)	0.93 (± 0.02)	182 (± 192)	50	20	9	5
					27				
Philips	Wood only	7	211 (± 101)	0.95 (± 0.03)	59 (± 56)	42	15	7	4
					21				
					27 (± 22)				

PM_{2.5} emissions rates reported as geometric mean with arithmetic mean below and standard deviation in parentheses

Table 4.3 shows in field emissions rates and modified combustion efficiencies during normal cooking activities in village homes in Haryana India for traditional Chulha stoves, Angithi stoves used principally for animal fodder, and the Philips forced draft stove using wood and dung fuels. In addition, Table 4.3 shows Gaussian modeling of the distance for the plume to reach 1 µg/m³ as an indicator of the impact on neighborhood pollution levels. Angithi stoves outdoors would have to be 104 meters apart and

traditional Chulha 37 meters apart for the stove not to appreciably increase background concentrations creating a localized neighborhood pollution effect with a wind speed of 1.5 m/s. This distance is much greater than the typical distance between village homes in Haryana, with the result that significant neighborhood pollution impacts are seen. In contrast the Philips stove using wood only would only have to be 7 m apart for the stove not to appreciably increase neighborhood pollution levels with a wind speed of 1.5 m/s, and impacts on neighborhood pollution would be minimal.

Emissions rates from outdoor cooking that would increase exposures equivalent to air quality guidelines and interim targets would generally result in a neighborhood pollution impact in villages in Haryana, which are densely populated. In the more rural conditions prevalent in many parts of Central America and Africa this would not be the case due to the housing density. Thus, prioritization of emissions rates to reduce personal exposure impacts or neighborhood impacts will be a function of local conditions, and both models should be evaluated simultaneously. These models however highlight the importance of looking at personal exposures and the impacts of neighborhood pollution. Inclusion of the neighborhood impact of pollution should be addressed more formally both in guidelines on emissions rates from stoves that would be protective of health, and also in wider health impact evaluation efforts and burden of disease estimates. This is especially true as the burden of disease from household air pollution is likely underestimated since the impacts of neighborhood pollution are not captured by satellite models that estimate global burdens of ambient pollution¹⁰⁰ as the resolution is much larger than that of neighborhood pollution.

Although outdoor cooking thus contributes to both neighborhood concentrations and ambient background concentrations, from an individual stove perspective it is useful to evaluate what emissions rates cause direct impacts to neighbors. Figure 4.6 shows a comparison of emission rates from outdoor cooking for exposures to increase equivalent to AQG, with distance required for emissions plume concentrations to reach $10 \mu\text{g}/\text{m}^3$. For emissions rates that would reduce exposures of the cook

equivalent to the AQG (126 mg/min) and IT 3 (189 mg/min), the plume concentration would reduce to lower than the AQG before the plume reached neighbors downwind in these Haryana villages at wind speeds greater than 1 m/s (10m and 14m respectively). In contrast emission rates that would reduce exposures of the cook equivalent to IT 2 and IT 1 would result in emissions concentrations that were significantly above the AQG by the time the plume reached neighbors downwind (23m and 32m respectively), and thus would be expected to have a direct health implication. Clearly, however, the extent of impacts would again be dependent on housing density.

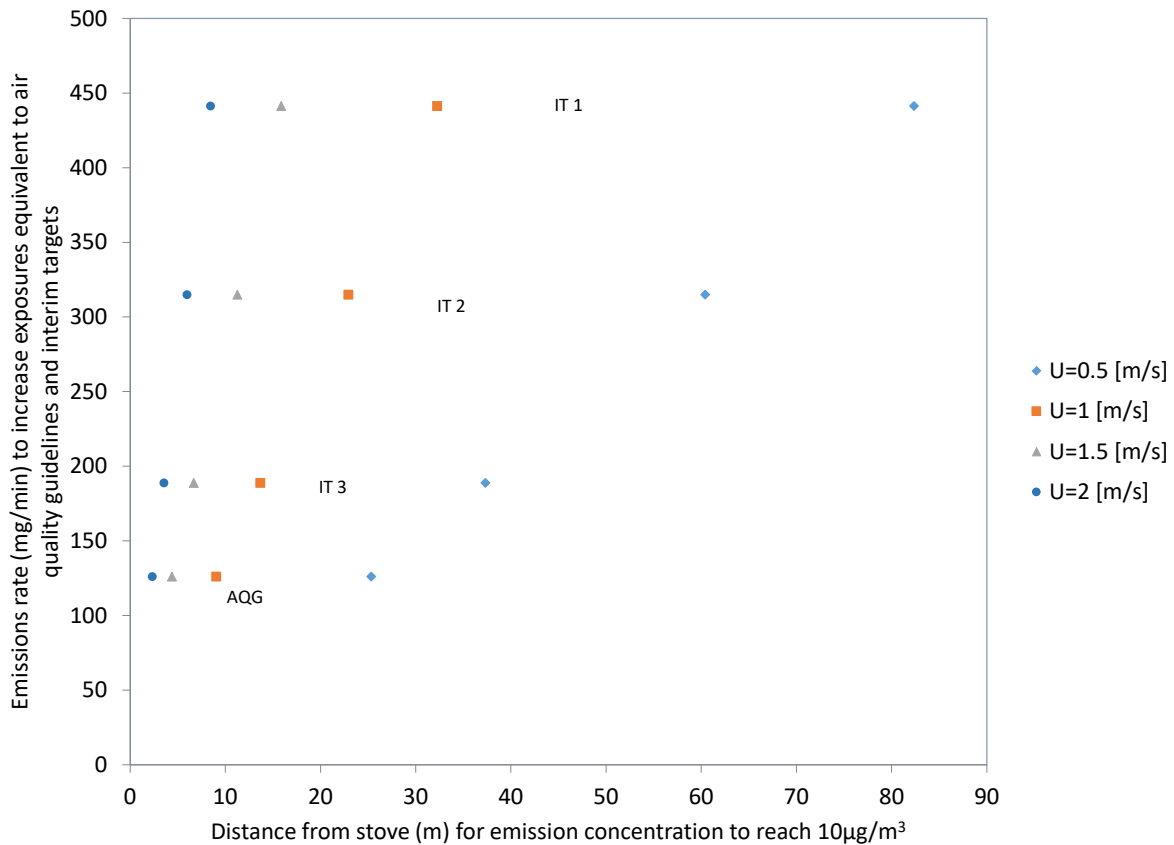


Figure 4.6: Emission rates from outdoor cooking for exposures to increase equivalent to AQG in comparison to distance required for emissions concentrations to reach 10 µg/m³.

While these examples serve to illustrate the impacts on neighborhood pollution of different emissions rates, there are many other factors in the real world that impact pollution dispersion such as building

orientation, barrier walls, variable wind speeds etc. The dispersion models here do not incorporate this variability, but rather use a set of relatively conservative assumptions to be more protective of health. Most critically these models use a set of measurements of emissions rates from real village homes during normal daily cooking activities and are not the result of controlled water boiling tests, as controlled water boiling tests are not representative of in field emissions and generally tend to underestimate emissions from open fire type stoves^{9, 31}. In addition, these measurements use a set of cooking times from real cooking of rice and chapatti meals in a village kitchen. Clearly, however the meals cooked do not represent the wide range of dishes cooked in village households in India, or seasonal changes in dishes, but rather represent the two basic meal types typical for that area for an average household size. Field measurements of exposures during cooking would improve modeled estimates of emissions rates and are a priority. Wind speeds used to estimate emissions rates and pollution dispersion are low to represent the dense building in Haryana villages. Pollution concentrations are estimated in the center of the plume representing the most elevated concentrations, although that is unlikely as people avoid standing directly in the smoke plume. Further, these models assume use of one stove at a time in a household, but it is possible that cooking animal fodder and cooking meals may occur simultaneously. However, these models are a first step in integrating neighborhood pollution impacts into emissions rates from outdoor cooking that are protective of health.

4.5 Conclusions

Emission rates modeled to meet AQG for outdoor cooking are substantially higher than emission rates for indoor cooking, because the models estimate impact of emissions on personal exposure concentrations rather than microenvironment concentrations, and because the smoke is able to disperse more readily outdoors compared to indoor environments. As a result, the best performing solid-fuel biomass stoves would meet AQG when cooking outdoors. Since outdoor cooking represents a significant fraction of global cooking, emissions guidelines should better represent the different contexts

in which stoves are being used, especially given that in these contexts the best performing solid fuel stoves have the potential to provide significant benefits.

Emissions rates from outdoor cooking that would increase exposures equivalent to air quality guidelines and interim targets may also result in neighborhood pollution impacts depending on housing density.

Thus prioritization of emissions rates to reduce personal exposure impacts or neighborhood impacts should be evaluated based on local conditions. Inclusion of the neighborhood impact of pollution should be addressed more formally both in guidelines on emissions rates from stoves that would be protective of health, and also in wider health impact evaluation efforts and burden of disease estimates.

5.0 PAH emissions from dung, wood and mixed fuels

5.1 Introduction

While studies have previously identified emissions of known human-carcinogenic hydrocarbons found in solid-fuel smoke in Indian homes^{103, 104}, such as 1,3-butadiene, benzene, and PAHs^{103, 105, 106}, only a few in field assessments of individual PAHs from traditional stoves in rural Indian household cookstove emissions have been performed^{50, 103, 107-109}. The overall impact of PAHs emitted from Chulhas in Southeast Asia is difficult to assess due to large variation in reported emission factors measured using different approaches with inconsistent conclusions between fuel types. This chapter provides quantitative assessments from in field measurements of a set of 16 particle bound PAHs, designated as High Priority Pollutants by the United States Environmental Protection Agency (US EPA), from wood, bovine dung, and mixed-fuel (a combination of wood and bovine dung patties) in traditional Indian Chulha and Angithi cookstoves.

Elemental Carbon or soot formation starts with the formation of gas-phase molecular species from fuel pyrolysis, followed by incipient ring formation, PAH growth, particle nucleation, heterogeneous surface reaction, and particle-particle interactions. Pyrolysis is a temperature dependent devolatilization independent of surrounding gas composition. Biomass fuels contain cellulose, lignin and hemicellulose in variable quantities which each have their own pyrolysis behavior. In the pathway to soot formation numerous experimental and numerical studies have validated that the formation of thermodynamically stable PAHs is critical in the transition from gaseous fuel molecules to solid soot particles. These reactions are intrinsically difficult to model or predict for more complex fuels, such as solid biomass in cookstoves, as they involve many intermediary compounds and multiple reaction mechanisms, and therefore are often evaluated experimentally on a per event basis¹¹⁰.

PAH formation from household solid fuel use is temperature driven, and the balance of two competing processes: chemical synthesis of PAHs enhanced by increasing temperature^{111, 112} and increasing oxidation of PAHs into elemental carbon at high temperatures¹¹³. Particle formation pathways generally transition solid fuel emissions from lower MW, smaller PAH compounds to larger, generally more mutagenic PAHs, and finally to coagulated soot particles that form EC¹¹⁴. During the early flaming combustion, when temperatures are lower, a large amount of low molecular weight (MW) PAH are synthesized through the hydrogen abstraction acetylene addition (HACA) pathway due to oxygen restriction¹¹⁵. In early flaming combustion large particles with diameters between 0.2 μm –1 μm are dominant containing higher concentrations of low-MW PAH¹¹⁵. As temperatures increase in later burning stages the particles within the range of 0.04 μm –0.2 μm are emitted in lower number concentrations but with higher concentration of higher MW PAH emissions¹¹⁵.

PAH emission factors from residential fuel combustion are highly variable, resulting in significant uncertainty with respect to the estimation of emissions of PAHs from this sector. In part, this variability would be expected as combustion conditions in household solid fuel use are highly variable. The influence of stove type and cooking practices on PAH emissions should not be surprising, as it is generally understood that PAHs can act as soot/elemental carbon precursors during incomplete combustion of biomass and that additional fused benzene rings are formed by reactions of smaller hydrocarbons (such as 1,3-butadiene and diatomic carbon or acetylene)¹¹⁴.

Factors that dominate emissions of volatile organic compounds (VOCs), which include PAHs as a subset, are controlled by pyrolysis of fuel biopolymers¹¹⁶, and may not correlate well with emissions of CO, CO₂, and NO_x produced mainly from flaming or non-pyrolysis smoldering processes. In household stoves, modified combustion efficiency (MCE, the ratio of CO₂ to the sum of CO and CO₂) alone could only explain about 46%, and 30% of the total variations in emission factors for parent PAHs, and a subset of oxygenated and nitrated PAHs¹¹⁷. Using the relative contributions from high and low temperature

combustion, as the temperature is known to drive differences in VOC profiles from distinct combustion events¹¹⁸, would therefore be expected to be a useful predictor of PAH emissions. The ratios of ethyne (C_2H_2) to furan (C_4H_4O) were shown to best predict the ratios of high- to low-temperature emissions of VOCs in various biomass burning profiles measured by PTR-MS¹¹⁸. In this study the temperature regime of the fire determined by the ethyne (C_2H_2) to furan (C_4H_4O) ratios were used to evaluate the contribution of different ring sizes as a % of the total summed PAH emissions for each fuel and stove combination.

PAH isomer ratios have been used for PAH source apportionment and assessing their chemical fate during transport. A number of studies have relied on PAH isomer ratios in ambient PM to apportion PAH to biomass burning, vehicular emissions, and other sources in either household or direct emissions^{106, 119} and in downwind atmospheric concentrations¹²⁰⁻¹²², as the ratios of individual species are considered fairly stable during atmospheric fate and transport^{119, 120}. However, ratios observed in emissions plume may differ from those in ambient air due to changes associated with the chemical aging and resulting transformation of the PAH compounds, such as due to vapor/particle partitioning, so comparisons between primary emissions, rather than between primary and secondary emissions, are most apt¹²³. Apportioning sources, therefore, relies on well constrained PAH isomer ratios comparing primary emissions¹²⁴⁻¹²⁶. PAH isomer ratios are often applied to PAHs that are close together in MW, such as fluoranthene and pyrene (FLA and PYR), as the transition to higher MW PAHs is associated with differences in combustion that can be considered intrinsic to the burn^{120, 121, 127-129}. PAH isomer ratios of FLA to the sum of FLA and PYR (FLA/FA+PYR) in the range of 0.5 ± 0.1 are often cited as characteristic of vehicle emissions, whereas wood combustion diagnostic ratios vary widely across stoves, ranging from ~ 0.1 to ~ 0.8 ¹²⁷. Parent PAH compounds have been widely used to detect PAHs from combustion, including to gauge the contribution from petroleum versus other combustion sources¹²⁸.

5.2 Methods

Samples were collected between August and September 2015 in a rural Indian village located at the SOMAARTH Demographic, Development, and Environmental Surveillance Site in Palwal District, Haryana, India, run by the International Clinical Epidemiological Network (INCLEN).

More information on the field measurements can be found elsewhere^{2, 18, 130}. Briefly, a local woman was recruited to cook traditional rice based, or chapatti based local Indian meals with market vegetables for four people (average household size) using the Chulha or simmering animal fodder with the Angithi. Molecular composition of particles were discussed by Fleming et al.¹⁸, gas phase VOC composition was reported by Fleming et al.¹³⁰, SOA formation from emitted VOC was discussed by Rooney et al.⁴, and Weltman et al.² discussed climate effects of the emissions and compared results for infield vs laboratory emission measurements for cookstoves.

5.2.1 Sample collection

More detail on sample preparation, collection and storage are reported in Fleming et al.¹³⁰. Briefly, emissions were sampled in the plume 1 m above the stove. Quartz filters were used to collect PM_{2.5} samples using 1.5 l/min flow (PCXR-8, SKC Inc.) through cyclone samplers (2.5 micron, URG corporation). Quartz filters were then briefly stored at -20°C in India until being transported to be stored in a freezer at -80°C. Flows were evaluated before and after sampling using a mass flowmeter (TSI 4140) and the average flow rate combined with the elapsed time was used to calculate the sampled air volume. Background samples were collected on quartz filters at the same flow rates, but no PAHs above the limits of detection were observed in these samples, and therefore background subtractions were not performed. Likewise, no PAHs above the limits of detection were observed in the solvent blanks used in this project.

Quartz filters were removed from storage at -80°C and a 1-cm square punches were taken with a SP-10 sample punch (Sunset Laboratory Inc. OR, USA) for elemental carbon/organic carbon (EC/OC) analysis. The remaining filter was then placed in individual 15-mL polypropylene centrifuge tubes (Corning, NY, USA) and stored at -80°C until extraction and analysis of PAHs within two years.

5.2.2 HPLC methods

PAHs were analyzed using established HPLC methods¹³¹. First, 5 mL aliquots of HPLC-grade acetonitrile (Fischer Scientific, NH, USA) were added to the centrifuge tubes containing the quartz. Each centrifuge tube was then placed in a water bath sonication device (FS20 Ultrasonic Cleaner, Fisher Scientific, NH, USA) in ice water, but was not submerged, for 45 minutes. After sonication each tube was placed into a centrifuge (IEC Centra CL3R, Thermo Fisher Scientific, Waltham, MA) at 4000 rpm for 5 min. Finally, 1 mL of each extract was filtered using a 0.2 micron syringe filter (Polyvinylidene Fluoride PVDF Syringe Filters, 0.22 µm, 13 mm, Luer-Lok/Luer Slip, Tisch Scientific, OH, USA) and placed in autosampler vials (amber glass vials with screw tops, Shimadzu, MD, USA) for analysis by HPLC-DAD. The HPLC separations were carried out using a Hewlett Packard pump system (HP 1100 Series) connected to a diode-array detector (Hewlett Packard, HP 1050 Series). The data were collected and integrated by HP Chem Station for LC, A.06 Revision (Hewlett Packard). An Aquasil C18 column (250 × 4.6 mm I.D., 5 µm particles, Western, Analytical Products, Inc., Murrieta, CA) was employed for separation. The injection volume was 50 µL, the flowrate was 0.7 mL/min and the detection wavelength was 254 nm. The mobile phase consisted of acetonitrile/H₂O (70:30 %v/v). Concentrations in each amber glass vial were calculated using a 6-point linear calibration curve at 1:10, 1:50, 1:100, 1:150, 1:250, and 1:500 dilutions of a 10 µg/mL 16-PAH calibration mix analytical standard (47940-U Supelco, Millipore Sigma, MA, USA) with duplicate injections of the 1:10 and 1:500 dilutions ($R^2 > 0.999$). Due to co-elution, acenaphthene and flourene, benzo(b) and benzo(k)fluoranthene, and 1,2-benzathracene and chrysene were not resolved separately. Acenaphthylene and benzo[ghi]perylene were not well resolved or quantified in this study.

Particle-bound PAHs were evaluated in a standard reference material (Standard Reference Material 1648a, Urban Particulate Matter) and recoveries were generally good for PAHs with 3 or more rings (see supplementary information Table S3). As a quality control check, two sets of quartz filters were also spiked in triplicate with 1 mL of a prepared 50 ng/mL PAH Standard Mix (47940-U Supelco, Millipore Sigma, MA, USA), and after 15 minutes or 24-hours sealed in the dark at room temperature were extracted and analyzed by HPLC-DAD following NIOSH Method 5506 to evaluate recovery. Because PAH recoveries were acceptably high (>60% and less than <110%, see supplementary information Table S4) after 15 minutes, no adjustments were made to the measured PAH concentration values presented here. After 24-hours at room temperature lower MW PAHs were mostly absent from the spiked filters (see supplementary information Table S4). Fourteen PAHs are reported here: naphthalene (NAP), acenaphthene/fluorene (ACE/FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), 1,2-benzanthracene/chrysene (BaA/CHY), benzo(b)fluoranthene/benzo(k)fluoranthene (BbK/BkF), benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DahA), and indeno(1,2,3-c,d)pyrene (IcdP). For comparisons between stove/fuel combinations, one-way analyses of variance (1-way ANOVA) followed by Tukey's Honest Significance Tests were used.

5.2.3 Sample detection

Particulate PAHs were detected above the limit of detection (LOD) in 45 out of 46 tests. In the 45 tests where PAHs were detected, 93% of individual PAH species were detected above the limit of detection (Table S2). For tests where a PAH emission was below LOD, the minimum ratio of that PAH to CO₂ above the LOD was substituted for analyses. PAH emissions by compound for each stove/fuel combination are presented as summed on a milligram per kilogram of dry fuel basis (Figure 5.1) and per PAH species (Table 5.2). The cross in Figure 5.1 indicates a borderline significant difference from the Angithi (p-value = 0.06), the double asterisks indicates a significant difference with the Angithi, but not with the other fuels in the Chulha, and triple asterisks indicating significant differences from both the Angithi and dung

in the Chulha. All measured PAH emissions were summed using toxic equivalency factors to produce one measure, benzo[a]pyrene equivalency units (BaPe), that can be used to quantify inhalation risks for chronic exposures to air containing PAHs¹⁰⁷. PAH emissions by compound for each stove/fuel combination are also presented on a BaPe basis, calculated by multiplying the emission factor by its toxic equivalency factor (Table S5) for all PAHs that contribute more than 1% to the total BaPe (Table S1).

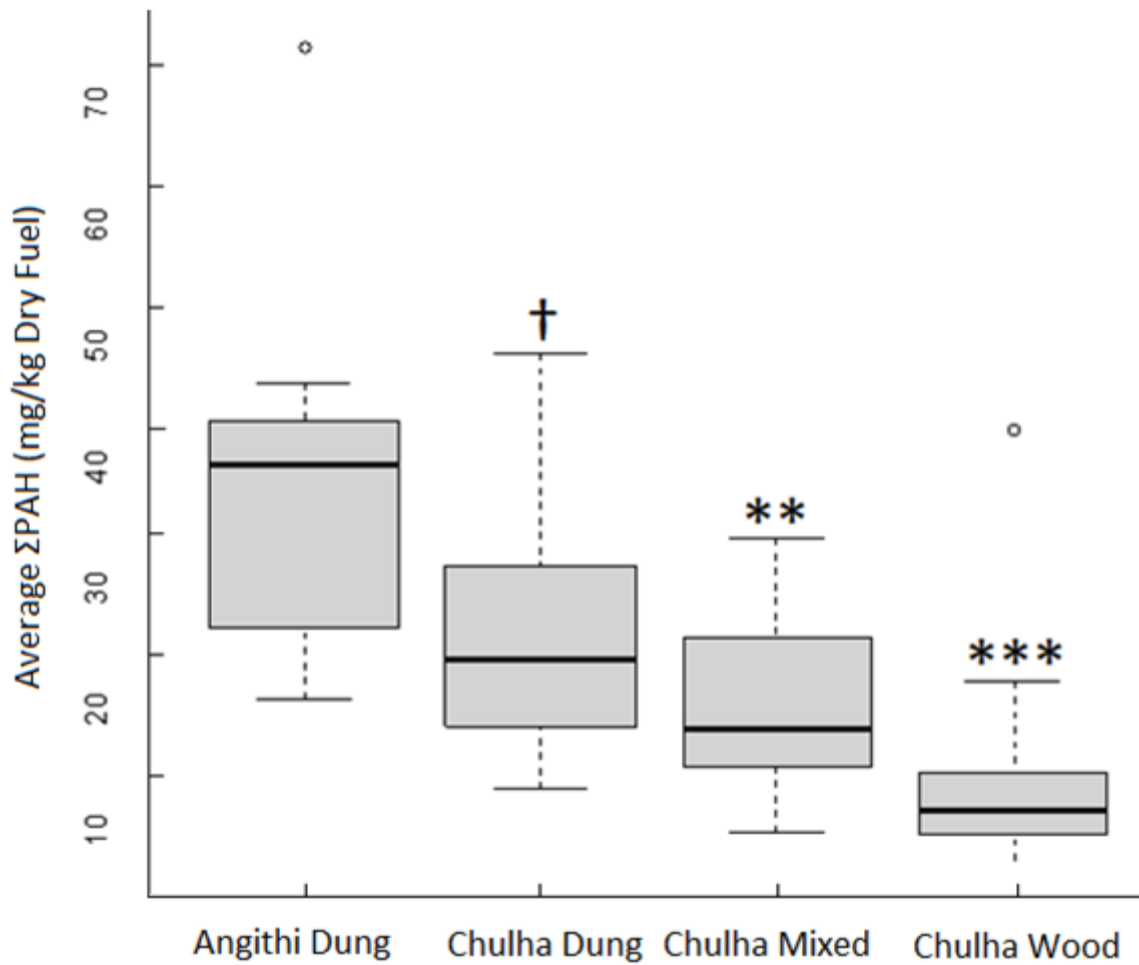


Figure 5.1: Average Σ 16-PAH particulate emissions on a milligram per kilogram of dry fuel basis for each stove and fuel combination.

5.2.4 Modified box model inputs

This research extends previous work on modeling indoor and outdoor cooking emissions and provides a quantitative estimate of cancer risk from field measurements of particulate PAH from mixed-fuel (dung and wood) use on a traditional Indian cookstove – the Chulha. By utilizing a previously existing steady-state box model designed to predict concentrations of pollutants in Indian homes cooking with solid fuels¹³², this section seeks to present an estimate of the total inhalation unit risks for cancer in adults who reside in homes where cooking is taking place with solid fuels.

In order to estimate the total risk for exposed adults and children separately, one needs to parameterize inhalation and air quality. Table 5.1 lists the necessary independent variables utilized in this thesis.

Table 5.1: A list of the inputs utilized in the chapter.

Parameter	Abbreviation	Unit	Mean	Min	Max	SD
Air Exchange Rate ¹³³	AER	min ⁻¹	25/60	3/60	60/60	15/60
Kitchen Volume ¹³³	KV	m ³	30	3	100	15
Stove Burn Time ¹³³	SBT	min/day	240	45	480	120
B[a]P Equivalency	BaPe	g/min	1.1E-04	4.1E-05	1.9E-04	4.2E-05
Inhalation Unit Risk ¹³⁴	IUR	m ³ /g	600			

Equation 5.1 shows how the independent variables in Table 5.1 were used to generate EBaPe – the total emissions per day of BaPe in grams per minute. Equation 5.2 shows the steady-state box model equation used to calculate the average concentration for the indoor environment (labeled as “C”). Equation 5.3 shows how the average indoor environmental concentration is multiplied by the inhalation unit risk to produce the total excess lifetime risk of developing cancer (TELRDC).

Equation 5.1: Total emissions per day of BaPe in grams per minute.

$$EBaPe \text{ (g/min)} = BaPe * SBT / (60 \text{ minutes} * 24 \text{ hours})$$

Equation 5.2: Average concentrations of BaPe for the indoor environment.

$$C \text{ (g/m}^3\text{)} = E_{BaPe} / (AER * KV)$$

Equation 5.3: Total excess lifetime risk of developing cancer.

$$TELRDC \text{ (no units)} = C * IUR$$

Equation 5.4: Excel code utilized to produce estimated emissions per day of BaPe in grams per minute.

$$EBaPe \text{ cell: } =\text{GENLIMITNORMAL}(1.1*10^{-4}, 4.2*10^{-5}, 4.1*10^{-5}, 1.9*10^{-4}) * \text{GENLIMITNORMAL}(240, 120, 45, 480) / (60 * 24)$$

In order to produce distributions for the calculated values for the steady-state box model approach, a Monte Carlo-based approach was utilized. “YasaiW” is a simulation add-in for Excel that easily enables Monte Carlo-based simulations. The “GenLimitNormal” function is an Excel command within YasaiW designed to parse a mean, standard deviation, minimum, and maximum value that generates a value by sampling a normal distribution with the given inputs until it falls within the minimum-maximum values. YasaiW allows one to control the number of sampling events and specify outputs in order to generate probability distributions of outputs. Equation 5.4 shows the Excel code utilized to produce E_{BaPe} as an example of how “YasaiW” was utilized. One-thousand iterations were ran to produce the results with a single fixed seed.

5.3. Results and Discussion

In this chapter quantitative assessments from 45 in-field measurements of 16 particle-phase PAHs from wood, bovine dung, and these two fuels mixed together in a traditional Indian cookstove, the Chulha, and dung in another traditional Indian stove, the Angithi are presented. These Σ PAHs were highest in dung-based fires and mixed fuels were between dung and wood. In-field measurements of PAHs were

generally similar, or slightly lower, than those reported in laboratory studies for dung and much lower than wood. Elemental carbon was closely linked to the higher MW 5 and 6-ring PAHs, but this relationship varied widely across fuel types used in this study. PAH isomer ratios, used in source apportionment, were shown to vary between dung and wood burns and the ratio of fluoranthene to the sum of fluoranthene and pyrene (FLU/FLU+PYR) typically used for fossil-fuel burning overlapped with the mean value for our wood-only testing.

5.3.1 Particle-phase PAH emissions

Table 5.2 shows a summary of PAH emissions from Chulha cookstoves using dung, wood and mixed fuels, and Angithi cookstoves using dung fuels. Figure 5.1 shows boxplots of PAH emissions for different stove and fuel types, which demonstrate borderline significant Σ PAH emissions decreases going from dung burned in the Angithi to dung burned in the Chulha ($p=0.06$), mixed fuels in the Chulha ($p<0.01$), and wood in the Chulha ($p<0.001$). Σ 16PAHs emissions ranged from 75 mg/kg dry fuel in the Angithi to 5 mg/kg dry fuel in the Chulha burning wood. While Σ 16PAHs emissions from mixed fuels in the Chulha were not significantly different to burning wood or dung in the Chulha independently, significant differences in Σ 16PAHs emission were seen between wood and dung burned in the Chulha ($p=0.045$). Average particulate PAH emissions are presented by species on a mass basis (figure S3) and on a BaPe basis (figure S4). $PM_{2.5}$ emissions were higher during smoldering combustion in the Angithi compared to more flaming combustion in the Chulha¹⁸ generally in agreement with increased emissions suggested in theoretical and measured papers on soot formation^{135, 136}. One possible explanation for the increase in Σ 16PAHs for the Angithi is that the placement of the cooking pot on top of the flat dung patty arrangement in the stove limits the availability of fresh oxygen¹³⁵.

Table 5.2: PAH emissions by compound for each stove/fuel combination presented on a milligram per kilogram of dry fuel basis.

	Angithi Dung	Chulha Dung	Chulha Mixed	Chulha Wood
PAH Species	(n=9)	(n=10)	(n=12)	(n=14)
NAP	9.2 ± 1.2	3.1 ± 0.6	1.4 ± 0.3	1.7 ± 0.7
ACE/FLO	7.2 ± 1.6	1.0 ± 0.5	0.1 ± 0.1	0.7 ± 0.2
PHE	3.1 ± 0.5	0.9 ± 0.2	0.4 ± 0.1	0.4 ± 0.2
ANT	0.7 ± 0.1	0.2 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
FLA	6.6 ± 1.5	4.9 ± 1.1	3.5 ± 0.4	1.5 ± 0.5
PYR	5.4 ± 1.2	4.5 ± 0.7	3.3 ± 0.5	1.7 ± 0.5
BaA/CHY	2.1 ± 0.4	2.9 ± 0.5	2.7 ± 0.3	1.4 ± 0.2
BbF/ BkF	1.7 ± 0.4	3.8 ± 0.6	4.0 ± 0.4	2.9 ± 0.4
BaP	1.4 ± 0.3	2.5 ± 0.3	2.5 ± 0.3	1.9 ± 0.2
DahA	1.2 ± 0.2	1.1 ± 0.2	0.9 ± 0.1	0.7 ± 0.3
IcdP	0.7 ± 0.2	1.3 ± 0.2	1.6 ± 0.2	1.0 ± 0.1
BaPe	3.2 ± 0.6	4.3 ± 0.6	4.1 ± 0.4	3.0 ± 0.4
∑16-PAH EF	39.3 ± 5.4	26.3 ± 3.4	20.6 ± 2.2	14.0 ± 2.5

In this chapter greater differences were observed between fuel types in ∑16PAHs emissions from cooking than observed in controlled laboratory testing of fuels using electric heaters⁵⁶, likely due to the differences in starting and maintaining the fire during cooking. ∑16PAHs emissions during in field testing in Chulha were a factor of 2-3 higher than reported emissions from wood fuels using laboratory burning and approximately a factor of 1.5 times higher than reported emissions from dung⁵⁶. Similarly, ∑16PAHs emission from another in-home study on a variety of Indian cookstoves were 4-6 fold higher compared to test chambers⁵⁶. It is not clear what differences in combustion lead to the increase in total summed particulate PAHs in their in-home measurements compared to ours and large differences between

homes, however the authors suggest that these differences may be due to the “presence of high moisture content, design of cook stoves and burning phase of fuels”⁵⁶. One possible explanation for the large difference in emissions compared to our current study is the use of different traditional stoves, as the authors of the other study only used traditional U-shaped Chulha stoves for one home for each fuel⁵⁶. Field measurements have also shown that BaP can adsorb onto the surface of elemental and organic carbon, and is protected from oxidation by complexes formed with co-emitted primary organic aerosol and secondary organic aerosol (SOA)⁴⁹, which are not well represented by controlled laboratory burns.

A better understanding of the influence of factors, such as fuel handling, burn rate, etc., on particulate PAHs emissions is necessary to better constrain $\sum 16$ PAHs emission from cookstoves.

The ratio of $\sum 16$ PAHs to $PM_{2.5}$ were similar across fuels/stoves at 1.8 ± 0.8 (average \pm standard deviation) mg PAHs per gram of $PM_{2.5}$, implying that all these stoves put out $\sim 0.2\%$ of their total fine PM mass as $\sum 16$ PAHs in primary emissions. $PM_{2.5}$ and our individual PAHs and EC are not well correlated in these cookstoves ($R^2 < 0.7$), therefore health effects associated with PM and associated with PAHs and EC are likely to have different mechanisms. Particulate BaP emissions were correlated with total particulate PAH carcinogenicity measured on a BaPe basis ($BaPe = [1.42 \pm 0.14 * BaP + 0.00, R^2 = 0.70, p < 0.001]$) for all stove and fuel combinations, and these relationships were especially robust for dung fuels ($R^2 = 0.88$).

While $\sum 16$ PAHs emissions tended to decrease in the Chulha from dung to mixed to wood fuels, toxicity determined on a BaPe basis were not statistically significantly different between stoves and fuels (all p-values > 0.2). The vast majority of PAH emissions on a BaPe basis were from larger MW PAHs with more fused rings, and BaP was the most abundant PAH on a BaPe basis. On a BaPe basis PAH emissions summed to between 1 and 7 mg per kilogram of dry fuel, with average emissions of 4 mg/kg and a standard deviation of 2 mg/kg. These emissions, on a BaPe basis, are slightly higher than those seen in

in-home measurements of heating and cooking with yak dung in traditional stoves (0.5 and 2 mg/kg)¹³⁷.

While Σ 16PAHs on a BaPe are similar across stoves and fuels, large increases in total PM_{2.5} seen with the Angithi are mostly reflected in increases in lower MW, less toxic, PAHs.

Measuring only the 16 PAHs undercounts total PAH-associated carcinogenicity as it ignores other PAH species¹³⁸, such as nitrated and oxygenated PAHs (so-called nitro-PAHs and oxy-PAHs)¹³⁹ that may be formed during combustion or through atmospheric reactions with parent PAH (pPAH) emissions¹⁴⁰.

Nitro- and oxy-PAHs are known to be directly mutagenic reactive species inside the body and are generally more toxic than their parent PAHs¹⁴¹⁻¹⁴³. While nitro- and oxy-PAHs contributed just 8% to the total PAH mass measured in Beijing ambient air measurements taken in the summer, nPAHs and oPAHs contributed 2 times the mutagenicity of pPAHs¹⁴⁴. One study of urban soil in Nepal found that nitro-PAHs exceeded parent PAHs in terms of overall toxic equivalency¹⁴⁵, and ambient air has been shown to have nPAHs exceeding primary combustion as nPAHs are thought to form through photochemical reactions of primary biomass emissions in ambient air⁵². However, strong correlations (such as $r=0.465 - 0.916$ for Chinese wood burning in residential stoves)⁵² between nitro- and oxy-PAHs and parent PAHs have been observed^{52, 146, 147}, implying that measuring parent PAHs, or simply BaP, can act as a surrogate measure for nitro- and oxy-PAHs and overall PAH-associated health effects. We have previously reported a greater abundance of compounds that are likely to be PAHs that may contain heterocyclic nitrogen atoms for dung-burning compared to wood-burning stoves¹⁸, and the relationship between parent PAHs and nitro- and oxy-PAHs is likely to also vary across different biomass burns.

5.3.2 Modified box model

Estimated excess cancers expressed per 100,000 population as a result of their percentile of exposure to benzo[a]pyrene-equivalency from indoor cooking with mixed dung and wood fuels from particulate PAHs is displayed in figure 5.2 on a logarithmic scale. The mean exposure resulted in 2.7 cancers per

100,000, almost double the median exposure's resulting excess cancer of 1.4 per 100,000. The ratio of the top 5% to the bottom 5% was 25, and this grew to 96 for the top 1% and bottom 1%. At the 99th percentile of exposure the excess cancers is 20.8 per 100,000 – representing just over ¼ of the crude cancer incidence rate of 81.2 in India in 2016¹⁴⁸.

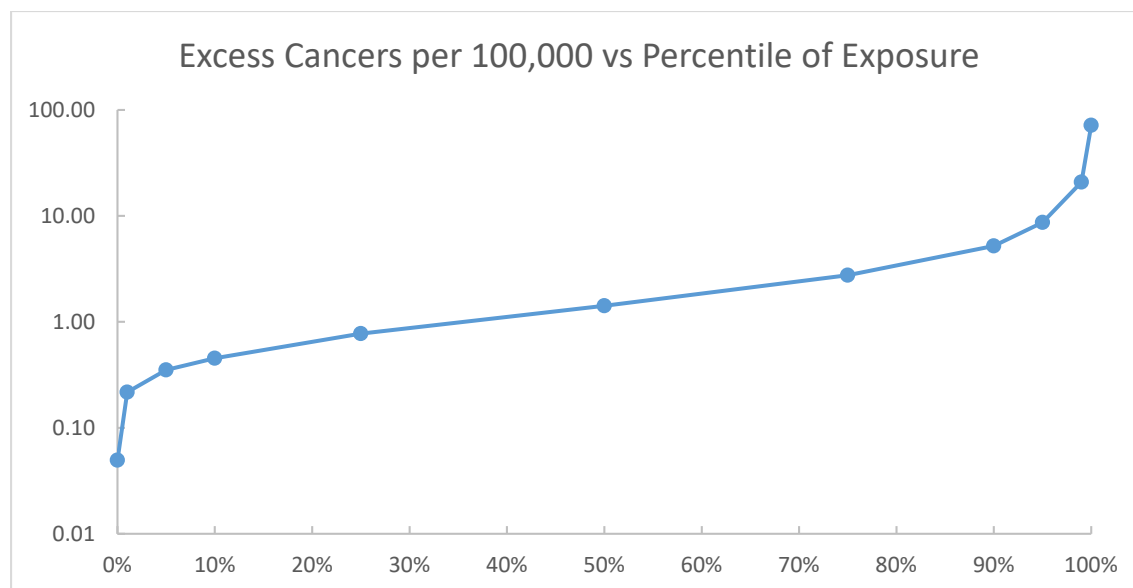


Figure 5.2: Excess Cancers per 100,000 vs percentile of exposure to B[a]Pe PAHs from mixed fuels in indoor cooking with a Chulha in India.

5.3.3 PAH ring size

Figure 5.3 shows $\Sigma 16$ PAHs emissions separated by ring size alongside EC emissions. PAH emissions are presented as a stacked bar chart with 2 or 3 rings, 4 or 5 rings, and 6 rings on a linear scale. EC is presented on a logarithmic scale. While 4- to 6-ring 16PAHs were similar across the different stove and fuel combinations, the 2- and 3-ring 16PAHs were notably larger in dung burning and in the Angithi compared to Chulha stoves. The proportion of total 16PAHs as lower MW 16PAHs (composed of 2-3 rings) were higher in the Angithi than in the Chulha, and in the Chulha with dung compared to mixed fuels (Figure 5.3). The ratio of $\Sigma(5\text{- to }6\text{-ring } 16\text{PAHs})/\text{EC}$ were similar for the Chulha (14 ± 1) and Angithi (15 ± 2) when both burned dung, but were a factor of 2-3 greater for dung (15 ± 2) and mixed fuels (9 ± 1) compared to wood fuels (4 ± 1) (Figure 5.4, $p < 0.05$ for all comparisons other than Chulha dung to Angithi). For figure 5.4, all fuels were burned in the Chulha unless noted otherwise, and the asterisks above "Mixed" indicates significant differences with both the Angithi and dung in Chulha (p-values of 0.01 and 0.02 respectively) and the triple asterisks above "Wood" indicate significant differences with all

other bins with p-values <0.01. No significant differences were observed between the Angithi and the Chulha stoves when both burned dung.

Similar to previous studies, 4- and 5-ring particulate PAHs were the predominate 16PAHs in all Chulha burns⁵³, however the Angithi burns were dominated by 2- and 3-ring 16PAHs and generated small amounts of EC per kilogram of fuel burned.

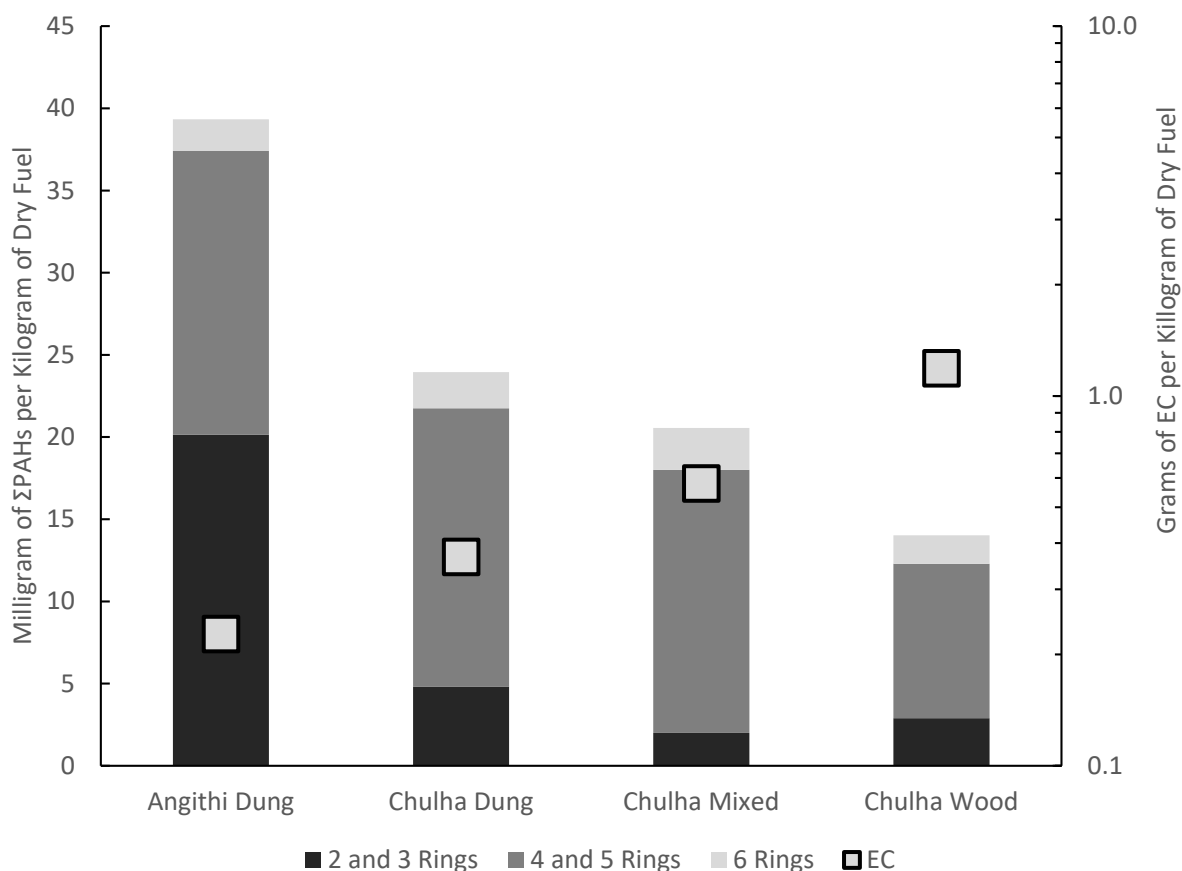


Figure 5.3: Average PAH emissions on a milligram of $\Sigma 16$ -PAH per kilogram of dry fuel separated by ring sizes as well as EC emissions on a gram per kilogram of dry fuel basis.

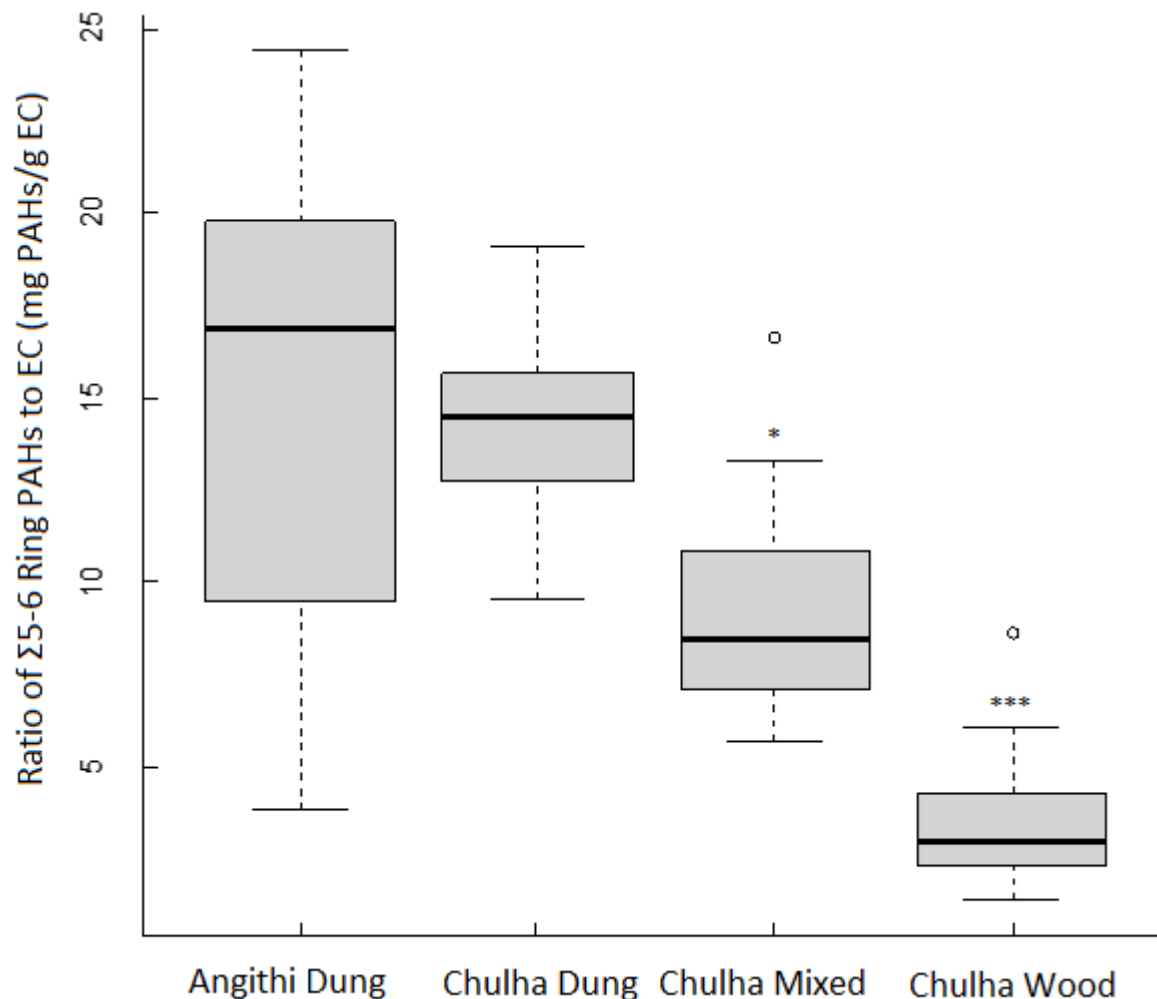


Figure 5.4: Ratio of $\Sigma 5$ and 6-ring PAHs to gram of EC for each stove and fuel combination.

The predicted ratio of high- to low-temperature VOCs¹¹⁸ is given in equation 5.5:

Equation 5.5: Predicted ratios of high- to low-temperature VOCs.

$$\frac{\text{Total Predicted VOC, High Temperature (ppbv)}}{\text{Total Predicted VOC, Low Temperature (ppbv)}} = \frac{\text{Ethyne (ppbv)}/0.0393}{\text{Furan (ppbv)}/0.0159}$$

Particulate PAH ring size, as a function of the lower MW 2- and 3-ring PAHs as a % of total PAHs, were correlated to the natural log of the ratio of high- to low-temperature VOC ($R^2=0.71$, see figure 5.5). As the ratio of high- to low-temperature VOC increases, indicating a transition to flaming rather than

smouldering combustion, the 2- and 3-ring PAHs make-up less of the overall % of total summed particulate PAHs. The trend was fit to a logarithmic curve as the ratio of 2- and 3-ring PAHs as a % of total summed PAHs could not ever fall below 0%. This trend suggests that the ratio of high- to low-temperature VOC is predictive of transition from less to more fused rings in particulate PAHs. This transition from lower to higher-MW PAHs is likely a function of changes in temperature during fire-starting and smoldering combustion when temperatures are lower and a large amount of low-MW PAH are synthesized through the HACA pathway due to oxygen restriction¹¹⁵. As temperatures increase in later burning stages the proportion of higher-MW PAHs increase indicating PAH synthesis as the dominant process¹¹⁶.

Linear models were fit to evaluate the influence of PM_{2.5}, modified combustion efficiency, and elemental carbon, which can act as an end step in the transition from low MW PAHs to high MW PAHs and then finally to soot particles, on the ratio of 2- and 3-ring PAHs to total summed PAHs. These models were compared to a linear model of the 2-3 ring PAHs as a percent of the total summed PAHs to the natural log of the ratio of high to low temperature VOC to examine which variables were significant predictors and associated with increases in overall adjusted R² values. While modified combustion efficiency was not predictive (Pr>t =0.79), increased PM_{2.5} was significantly associated with an increase in the 2- and 3-ring PAHs as a % of total PAHs (Pr>t = 0.03) and EC was borderline significantly associated (Pr>t =0.06). These models were also examined for parsimony using Akaike's Information Criterion (AIC), and a new model was fit including both PM_{2.5} and EC to assess whether either or both PM_{2.5} and EC should be used to predict the ratio of 2- and 3-ring PAHs as a % of total PAHs. While including PM_{2.5} or EC decreased overall AIC, increasing model parsimony, and increased overall adjusted R² when compared to a model with only the ratio of high to low temperature VOC the model with the lowest AIC included only the variables of PM_{2.5} and the calculated ratio of high to low temperature VOC and did not include EC. The best fit model is given in equation 5.6:

Equation 5.6: 2- and 3-ring PAHs as a percent of total summed $\Sigma 16$ -PAH particulate emissions.

$$\text{2- and 3-ring PAHs as \% of total PAHs} = 0.33 - 0.09 \cdot \ln(\text{Ratio of High to Low Temp VOC}) + 0.004 \cdot \text{PM}_{2.5}$$

This model suggests that as $\text{PM}_{2.5}$ increases that the percent of lower MW PAHs increases and as the ratio of high to low temperature VOC increases the PAHs transition more towards higher MW PAHs.

Similarly to the ratio of high to low temperature VOC, elemental carbon had an inverse relationship with the proportion of PAHs as 2- and 3-ring (not presented here as it was not included in the final equation chosen). Taken together these models indicate that MCE is a poor measure of the transition of PAHs from low- to high-MW in this dataset but that $\text{PM}_{2.5}$ and EC are significantly associated with the transition.

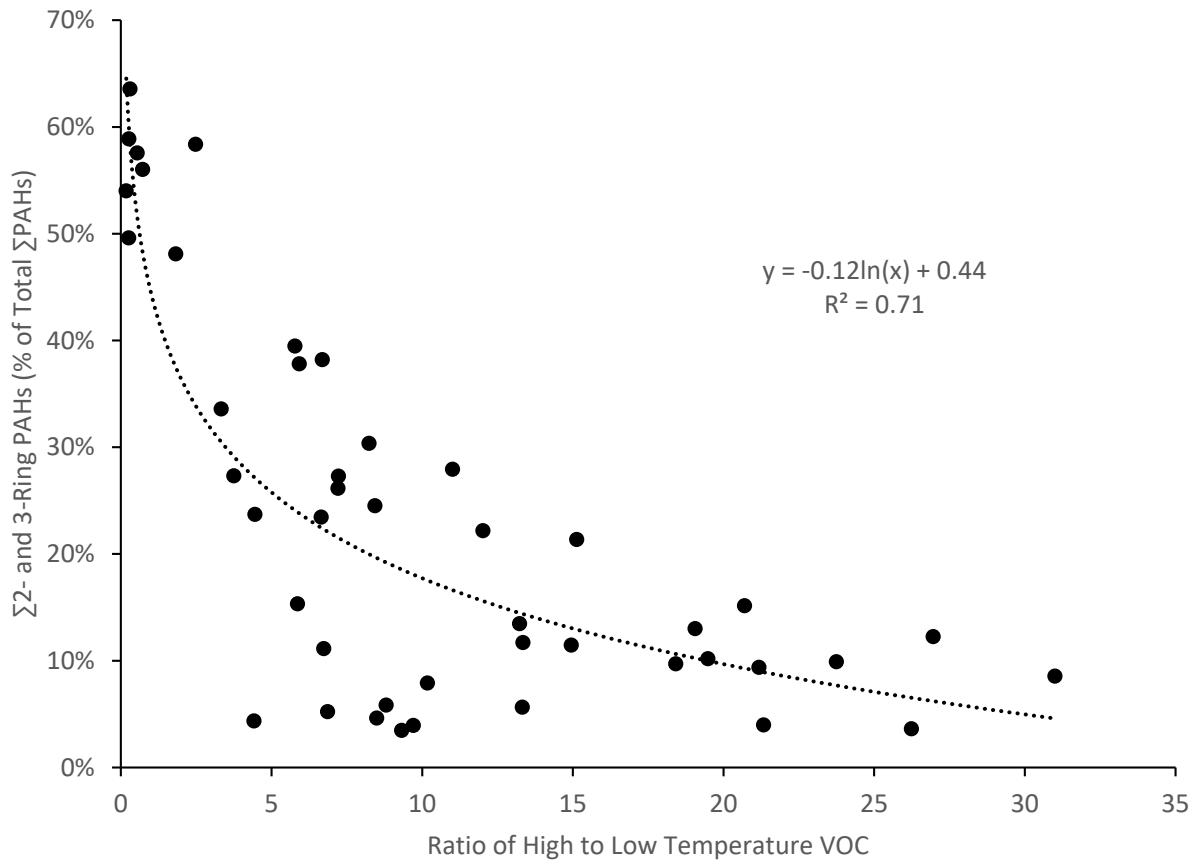


Figure 5.5: Relationship between 2- and 3-ring PAHs and high- to low-temperature VOCs.

5.3.4 Use of PAH isomer ratios in source apportionment

Ratios of anthracene to the sum of anthracene and phenanthrene ($ANT/(ANT+PHE)$) are also used in combustion source characterization, with much lower values (usually a ratio <0.1) for petroleum than those seen with biomass (approximately 0.1 to 0.4)¹²⁸. In the field tests presented in this study all ratios of $ANT/(ANT+PHE)$ were in the range typically seen in biomass (averages 0.18 to 0.35), with higher values for wood-only tests compared to any of the other field testing (p -values <0.01) (see Table 5.3). No significant differences were observed between any of the dung-burning fires (p -values >0.7). The ratios of $ANT/(ANT+PHE)$ in the field tests presented in this chapter generally overlap with those in the literature for wood burning and those seen with controlled burn measurements from Chinese and Nepalese clay pots^{127, 128}.

Ratios of fluoranthene to the sum of fluoranthene and pyrene ($FLT/(FLT+PYR)$) below 0.5 are often ascribed to petroleum combustion, whereas values above 0.5 are taken to indicate coal or biomass burning^{127, 128}. The ratio of FLT to FLT+PYR found for biomass burning in these in field tests overlapped these ranges typically designated for fossil-fuel burning and biomass burning between 0.45 and 0.55,¹²² indicating that these emissions could be incorrectly ascribed to fossil-fuels rather than domestic household biomass combustion. Further, the $FLT/(FLT+PYR)$ ratio decreased in this study going from dung to wood fuels (borderline significant; $Pr>F = 0.15$), indicating that the degree to which cooking sources are misrepresented as fossil fuel burning can be dependent on the specific cooking source and ratio of fuels used. While all dung-burning tests, including mixed fuels, had average ratios over 0.5 the wood-only tests in the Chulha had an average of 0.46 (arithmetic mean; geomean = 0.45). The ratios presented in this chapter are lower than those seen in particulate-matter in controlled burn measurements from Chinese and Nepalese clay pots burning 8 kilogram samples of wood, possibly due to differences in fire tending¹²⁸. Ratios between 0.43 and 0.74 have previously been reported for wood

combustion in traditional stoves, indicating that a cut-off of 0.5 may be inappropriate for ascribing combustion to petroleum sources when wood-burning stoves are also present¹²⁷.

Data from the Chinese and Nepalese clay pots with wood are particulate matter only measurements from using a hood and single measurements of individual four-hour burns of ~8 kilograms of wood¹⁴⁹. All other tests are from this study and are shown as arithmetic means plus or minus standard deviation.

Table 5.3: PAH isomer ratios from solid fuel combustion.

PAH Isomer Ratio	Wood	in	Mixed Fuel	Dung in	Wood in	Wood in	
	Chulha	in	in Chulha	Dung in Chulha	Chinese	Nepalese	
				Angithi	Clay ¹⁴⁹	Clay ¹⁴⁹	
ANT/(ANT+PHE)	0.35 ± 0.13		0.22 ± 0.06	0.18 ± 0.06	0.18 ± 0.01	0.25	0.07
FLT/(FLT+PYR)	0.46 ± 0.08		0.53 ± 0.06	0.51 ± 0.16	0.55 ± 0.02	0.71	0.67
FLT/PYR	0.90 ± 0.26		1.14 ± 0.29	1.15 ± 0.46	1.22 ± 0.08	2.4	2

5.3.5 Limitations and conclusions

PAHs are volatile compounds that exist in both gaseous and condensed states in cooking smoke, but they were only measured here in the particle phase. Because the partitioning coefficient of PAHs between gas and particle phases is strongly temperature dependent, this data set can only be taken to be measurement of these 16 particulate PAHs when emissions are diluted and collected in the filter holder at room temperature and not their initial emissions. Similarly, the correlation between an increased proportion of lower MW PAHs as a % of total PAHs for burns with a lower ratio of high- to low-temperature may differ for gas-phase PAHs. The PAHs measured in this study are not all health-relevant PAHs but only a subset thereof, additional measurements on other PAH species such as oxy- and nitro-PAHs would allow more accurate and precise measures of health effects. While our filters were sealed after collection and immediately kept at -20°C prior to longer-term storage at -80°C before

analyses, PAH volatilization loss has potential to bias results towards lower emissions¹⁵⁰. Measurements of PAH emissions during cooking were done during August and should not be considered to be accurate for heating during Winter or other tasks. Emissions from the food or animal fodder being cooked cannot be ruled out and may contribute to the measured PAHs presented here¹⁵¹.

Emissions of PAHs from wood, dung and mixed fuels in the Chulha highlight the need for better accounting of fuel mixtures in household surveys in this region, and drawbacks of solely using PAH isomer ratios for source apportionment, such as by diagnostic ratio or principal component analyses^{56, 104}. This study highlights more recent understanding of the processes that influence the formation of PAH from residential fuel combustion, such as the relative contribution of high- and low-temperature biomass combustion byproducts¹¹⁸.

6.0 Health implications of emissions measurements

6.1 Introduction

Household air pollution (HAP) is a major contributor to air pollution and resultant health impacts. HAP impact exposures of primary cooks, other household members, and ambient and regional air pollution. Furthermore, carbon credits from the installation of cleaner cookstoves have grown into a global industry worth tens of millions of dollars per year¹⁵². This chapter provides a literature-based evaluation of the effectiveness of semi-gasifier stove types in field based trials globally, and compares them to interventions to vent the smoke out of homes using chimneys.

6.2 Methods

6.2.1 Literature review

Google Scholar was searched with the term combinations: "semi-gasifier" cookstove cook stove and the terms "tlud" cookstove cook stove, returning 148 and 255 results respectively. The abstract of each result was read and any papers that had mentions of particulate emissions, climate-forcing, stove stacking, 'improved' cookstove programs, or other relevant details were scanned and, if relevant, selected for inclusion – yielding 97 papers. The papers were then further divided into non-exclusive categories of laboratory comparisons, field testing, stove intervention assessments, meta analyses, and other papers. Papers that did not report primary measurements of effectiveness of stove interventions (e.g., meta analyses of existing studies) were excluded from pooled analyses. All papers that measured exposure or indoor concentrations from stove interventions using non-pelletized biomass were selected to include in a pooled metanalysis if they contained mean/median measurements and the standard deviation and number of samples were either listed or calculable. A total of 17 stove interventions including 5 chimney stoves and 9 semi-gasifier stoves were selected covering a diverse range of

countries. Stoves were separated into semi-gasifier stoves similar to those available for DRC and chimney stoves for the purpose of analyses.

6.2.2 Interventions analysed

Stove interventions analyzed fell into two broad groups – 1) semi-gasification stoves, herein defined as natural or forced draft stoves using a rocket design designed to reduce particle emissions, and 2) multiple-stage/vented chimney stoves, which can reduce exposure by removing pollutants from the kitchen through a chimney.

6.2.3 Estimation of health impacts

Estimation of household level health impacts were performed using the online household air pollution intervention tool, HAPIT 3.1.1¹⁵³ (<https://householdenergy.shinyapps.io/hapit3/>) model for averted ill-health due to proposed interventions. Key features of this model include the use of multiple relationships between exposure and PM_{2.5}, relating exposure to acute lower respiratory infection (ALRI), chronic obstructive pulmonary disease (COPD), ischemic heart disease (IHD), lung cancer, and stroke; population-specific parameters, such as the number of people per household, child-to-adult ratio, and % of individuals who continue to use the intervention; scaling factors to translate the exposure of a cook to other adults or children in the home; and deaths, years lost to disability (YLDs), and disability-adjusted life years (DALYs).

HAPIT model version 3.1.1 uses the most updated integrated exposure response function to estimate disease impacts for diseases causally related to air pollution (Stroke, IHD, COPD, lung cancer, and ALRI). Air pollution impacts also include a wider range of disease outcomes for which there is growing evidence – such as low birth weight and other adverse pregnancy outcomes, which are not included in these estimates of disease burdens, and thus the results likely underestimate overall impacts of air pollution. Disease incidence rates are taken from countrywide Global Burden of Disease (GBD) estimates for the

Democratic Republic of Congo (DRC). Depending of the disease and intervention area, these incidence rates may over or underestimate the true rates. For example, in urban areas, GBD estimates for ALRI are likely overestimates, while IHD is likely underestimated, based on differences in the proximity to health care services and location in more affluent metropolitan areas. For lung cancer and COPD the impacts are harder to determine since the exposure period before health impacts are seen spans several decades during which there can be significant population migration and urbanization. Given the difficulties in obtaining robust estimates of disease incidence, use of Global Burden of Disease estimates were a pragmatic choice for modelling the impacts of stove interventions on these disease outcomes.

Several assumptions were necessary in order to carry-out these analyses. Because HAPIT requires exposure concentrations, exposures were estimated from kitchen concentrations. Exposures are estimated by multiplying kitchen concentration by a ratio of personal exposures to kitchen concentrations. The ratio used in these analyses was the same as that used for the 2010 Global Burden of Disease at 0.742 for women. Due to the proportion of DRC households that cook outdoors and a paucity of studies that analysed outdoor concentrations/exposures, outdoor concentrations were predicted for all interventions. In order to generate realistic outdoor cooking exposure concentrations from indoor kitchen measurements, a scaling factor of ~ 0.145 was applied based on our outdoor cooking models³⁵. HAPIT also contains values for the ratio of exposure between the mother-child and the cook to other adults which were kept at their default values. In order to generate results in terms of incidence rates per 100,000 individuals, the model was run for a total of 20,000 households with an average of 5 people (0.9 kids, 4.1 adults based on the HAPIT defaults for the DRC), a 5-year intervention, and 100% adoption of the intervention. All modelled benefits are presented per 100,000 individuals / 20,000 households and assume 100% adoption and a 5-year lifetime for the intervention stove. Comparisons for % of child and adult DALYs prevented is in comparison to a reduction to household

concentrations of 7 $\mu\text{g}/\text{m}^3$ rather than a total absence of $\text{PM}_{2.5}$, as the total absence of $\text{PM}_{2.5}$ is neither a realistic counter-factual for comparison nor an achievable public health goal.

6.3 Results

Table 6.1 (below) shows reductions associated with semi gasifier stoves. Table 2 shows reductions associated with chimney stoves.

Table 6.1: Studies included in pooled analysis for semi gasifier stoves.

Improved Stove Name	Country	Natural/ forced draft	Stove	In home $\text{PM}_{2.5}$ concentrations ($\mu\text{g}/\text{m}^3$)		Reduction	Ref.
				Pre	Post		
				Gyapa	Ethiopia		
Envirofit B1200	India	Natural	Semi- gasifier	501	345	31%	155
Envirofit G3300	India	Natural	Semi- gasifier	686	481	30%	155
Prakti Leo	India	Natural	Semi- gasifier	173	153	12%	155
Philips Natural	India	Natural	Semi- gasifier	585	279	52%	155

Philips Forced	India	Forced	Semi-gasifier	1436	750	48%	155
Oorja Forced Draft	India	Forced	Semi-gasifier	292	207	29%	155
Grouped analysis of Prakti, Philips, Envirofit, Ecozoom, and EcoChulha	Kenya	Both	Semi-gasifier	1280	640	50%	156
Ecozoom Dura - Pooled	Rwanda	Natural	Semi-gasifier	905	485	46%	157
Ecozoom Dura - Indoor	Rwanda	Natural	Semi-gasifier	910	558	39%	157
Ecozoom Dura- Outdoor	Rwanda	Natural	Semi-gasifier	910	243	73%	157
Pooled analysis				757	406	46%	

6.3.1 Semi-gasification stoves

Semi-gasification stoves decreased kitchen concentrations of particulate matter relative to traditional cookstoves in all analyzed interventions (n=11). The mean time-weighted kitchen concentrations of fine particulate matter went from 757 ± 384 (1 standard deviation) to $406 \pm 191 \mu/m^3$, representing a 46% reduction. A paired t-test indicates a significant decrease ($p < 0.001$).

In households currently cooking indoors, an intervention to switch from traditional stoves to semi-gasification stoves would result in approximately 29 ± 23 child deaths averted, 2452 ± 1969 child DALYs averted, 35 ± 22 adult deaths averted, and 928 ± 570 adult DALYs averted per 100,000 population. The intervention would result in a reduction of approximately $12\% \pm 11\%$ of child DALYs and deaths attributable to air pollution and $14\text{-}15\% \pm 10\%$ of adult DALYs and deaths attributable to air pollution.

In households currently cooking outdoors an intervention to switch from traditional stoves to semi-gasification stoves would result in approximately 45 ± 31 child deaths averted, 3875 ± 2667 child DALYs averted, 58 ± 35 adult deaths averted, and 1449 ± 852 adult DALYs averted per 100,000 population. The intervention would result in a reduction of approximately $39\% \pm 22\%$ of child DALYs and deaths attributable to air pollution and $45\text{-}46\% \pm 30\%$ of adult DALYs and deaths attributable to air pollution.

Table 6.2: Studies included for chimney based interventions.

Improved Stove	Country	Heating/ cooking	Natural/ forced draft	Stove	In home PM _{2.5} concentrations ($\mu\text{g}/\text{m}^3$)		Reduction	Ref.
					Pre	Post		
Chimney with grate	China	Both	Natural	Chimney Stoves	268	152	43%	158
Patsari	Mexico	Cooking	Natural	Chimney Stoves	257	101	61%	159
Sukhad Stove	India	Cooking	Natural	Chimney Stoves	520	330	37%	160

Patsari	Mexico	Cooking	Natural	Chimney Stoves	1020	340	67%	161
Mud ICS	Nepal	Cooking	Natural	Chimney Stoves	2130	730	66%	162
2-stage mud	India	Cooking	Natural	Chimney Stoves	1250	940	25%	163
Pooled analysis					908 ± 722	432 ± 333	52%	

6.3.2 Chimney stoves

Chimney stoves decreased exposures relative to traditional cookstoves in all analyzed interventions (n=6). The mean time-weighted kitchen concentrations of fine particulate matter went from 908 ± 722 (1 standard deviation) to 432 ± 333 µg/m³, representing a 52% reduction. A paired t-test indicates borderline significance (p = 0.066).

In households cooking indoors an intervention to switch from traditional stoves to chimney stoves would result in approximately 21 ± 25 child deaths averted, 1745 ± 2121 child DALYs averted, 25 ± 26 adult deaths averted, and 668 ± 685 adult DALYs averted per 100,000 population. The intervention would result in a reduction of approximately 9% ± 11% of child DALYs and deaths would be averted and 10-11% ± 11% of adult DALYs attributable to air pollution.

In households currently cooking outdoors an intervention to switch from traditional stoves to chimney stoves would result in approximately 32 ± 36 child deaths averted, 2758 ± 3121 child DALYs averted, 41 ± 46 adult deaths averted, and 1019 ± 1092 adult DALYs averted per 100,000 population. The

intervention would result in a reduction of approximately $28\% \pm 28\%$ of child DALYs and deaths attributable to air pollution and $33\% \pm 35\%$ of adult DALYs and deaths attributable to air pollution.

6.4 Discussion

6.4.1 Indoor and outdoor cooking

In field trials of semi-gasifier stoves or chimney stoves did not lead to reductions in exposures to less than $50 \mu\text{g}/\text{m}^3$. Exposures remained high after 46-52% reductions due to the high concentrations with traditional stoves – with an average pre-intervention exposure of 558 and post-intervention exposure of $338 \mu\text{g}/\text{m}^3$. Cooking outdoors resulted in an estimated drop in exposure from 123 to $53 \mu\text{g}/\text{m}^3$, with 9 of 16 interventions leading to exposures below $50 \mu\text{g}/\text{m}^3$ demonstrating that where possible removing the stove from the home to cook outdoors can provide significant benefits. Simply removing a traditional cookstove and relocating it outdoors would avert approximately 8416 DALYs and 98 deaths for children (39% averted) and 2640 DALYs and 99 deaths for adults (28-29% averted). An intervention to switch from traditional cookstoves used indoors to semi-gasifier stoves used outdoors would avert 11798 DALYs and 138 deaths for children (64% averted) and 3588 DALYs and 136 deaths for adults (39%-41% averted). While stove interventions that relied on indoor cooking lead to 10-15% of DALYs and deaths from air pollution being averted, moving cooking outdoors alone reduces DALYs and deaths by 30-40%, and semi-gasifier stove interventions that move cooking from indoors to outdoors would avert over 60% of child DALYs and deaths from air pollution and 40% of adult DALYs and deaths. Where removal outside is not possible cooking in kitchens separate from the living areas combined with use of chimneys would provide significant exposure reductions especially if combustion could be improved at the same time. Reductions in exposure from changing cooking locations was evaluated in only one intervention¹⁵⁷, in which time-weighted PM concentrations decreased from $910 \mu\text{g}/\text{m}^3$ to 558 for indoor cooking and to 243 for outdoor cooking. Daily exposure to particulate matter from cooking, however, is largely driven

by cooking location¹⁶⁴. Similarly, HAPIT 3.1.1 relies on assumptions regarding the ratio of exposure for mothers and children and for cooks and other adults in the home to predict health benefits. Reducing exposures to multiple members of the household through modifying cooking locations was not examined in this work due to a paucity of research in this area. While these ratios could be modified and possibly reduced in some homes through outdoor cooking or cooking in a different room, there is also the potential for enhancement of exposures due to neighborhood effects, infiltration, and other localized conditions. Assuming that this ratio doesn't change for outdoor cooking is likely a conservative estimate of the benefits of switching from indoor to outdoor cooking for most households.

6.4.2 Limitations

Particulate matter exposure reductions associated with cookstove interventions have non-linear integrated exposure responses¹⁶⁵⁻¹⁶⁷. Predicted intervention benefits from fuel savings, modified emission factors, and other linear predictors of pollutant emissions cannot be effectively used as a surrogate for predicted health effects. Estimated health impacts for chimney stoves are slightly lower than those for semi gasifier stoves, even though percentage reductions are greater in chimney stoves. This apparent contradiction is the result of baseline concentrations with traditional stoves being higher with chimney stoves (mean $908 \pm 722 \mu\text{g}/\text{m}^3$ for chimney stoves vs mean $757 \pm 384 \mu\text{g}/\text{m}^3$ for semi gasifier stoves). Post-intervention concentrations were similar for both chimney and semi-gasification cookstoves (432 vs $406 \mu\text{g}/\text{m}^3$ respectively). Since the reductions with the semi-gasification stoves are on a lower part of the dose response curve, the supralinear nature of the dose response curves leads to slightly larger estimates of health benefits. Incidence of adverse health impacts in burden of disease studies frequently relies on modelling of incidence rates due to incomplete health information and lack of detailed studies for Sub-Saharan Africa. True incidence rates may vary from those estimated both on a country basis and for particular geographic regions. Current fuel use surveys do not capture the full extent of solid fuel use in homes and thus estimates of health impacts are limited by the lack of data on

fuel mixing and secondary fuel types. In addition, it is well-established that stove interventions have the potential to increase exposures through “stove stacking” when households continue to utilize traditional biomass stoves in addition to intervention stoves. Finally, these estimate assume a useful stove life of 5 years, which for many intervention stoves does not reflect actual experiences in rural communities. Stove stacking, stove breakage or abandonment of the intervention have the potential to reduce intervention benefits.

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Supplementary Information

Table S1: PAH emissions by compound for each stove/fuel combination are presented on a milligram BaPe per kilogram of dry fuel basis. The final column lists the % of total BaPe that the Σ PAHs add up to. Only the measured compounds that contribute over 1% of total calculated BaPe are listed here.

Stove / Fuel Type	FLA	BbK / BkF	BaP	DahA	IcdP	% of Total BaPe
Angithi Dung	0.3 ± 0.1	0.1 ± 0.0	1.4 ± 0.3	1.3 ± 0.2	0.1 ± 0.0	99.0%
Chulha Dung	0.2 ± 0.0	0.2 ± 0.0	2.5 ± 0.3	1.2 ± 0.2	0.1 ± 0.0	99.5%
Chulha Mixed	0.2 ± 0.0	0.2 ± 0.0	2.5 ± 0.3	1.0 ± 0.2	0.2 ± 0.0	99.6%
Chulha Wood	0.1 ± 0.0	0.1 ± 0.0	1.9 ± 0.2	0.8 ± 0.3	0.1 ± 0.0	99.6%

Table S2: Percent of samples that were above limit of detection (LoD) for each PAH species or species.

Species	% of Samples above LoD
Napthalene (NAP)	87% (39/45)
Acenaphthene/Fluorene (ACE/FLO)	76% (34/45)
Phenanthrene (PHE)	96% (43/45)
Anthracene (ANT)	80% (36/45)
Fluoranthene (FLT)	96% (43/45)
Pyrene (PYR)	98% (44/45)
1,2-Benzanthracene/Chrysene (BaA / CHY)	100% (45/45)
Benzo(b)fluoranthene/Benzo(k)fluoranthene (BbF / BkF)	98% (44/45)
Benzo(a)pyrene (BaP)	98% (44/45)
Dibenz(a,h)anthracene (DahA)	100% (45/45)
Indeno(1,2,3-c,d)pyrene (IcdP)	100% (45/45)



Figure S1: Pictures of both the Angithi stove during use. A picture of the Angithi used during this project during cooking (on the left) and while the fuel is being lit (on right). The Angithi is loaded with smoldering cow dung patties and used to simmer animal feed.



Figure S2: Pictures of the Chulha stove during use. A picture of the Chulha used during this project during cooking with both wood and dung fuels (on the left) and while the Chulha is empty (on the right). The Chulha is loaded with wood, dung, or both fuels mixed together and used to cook a variety of meals.

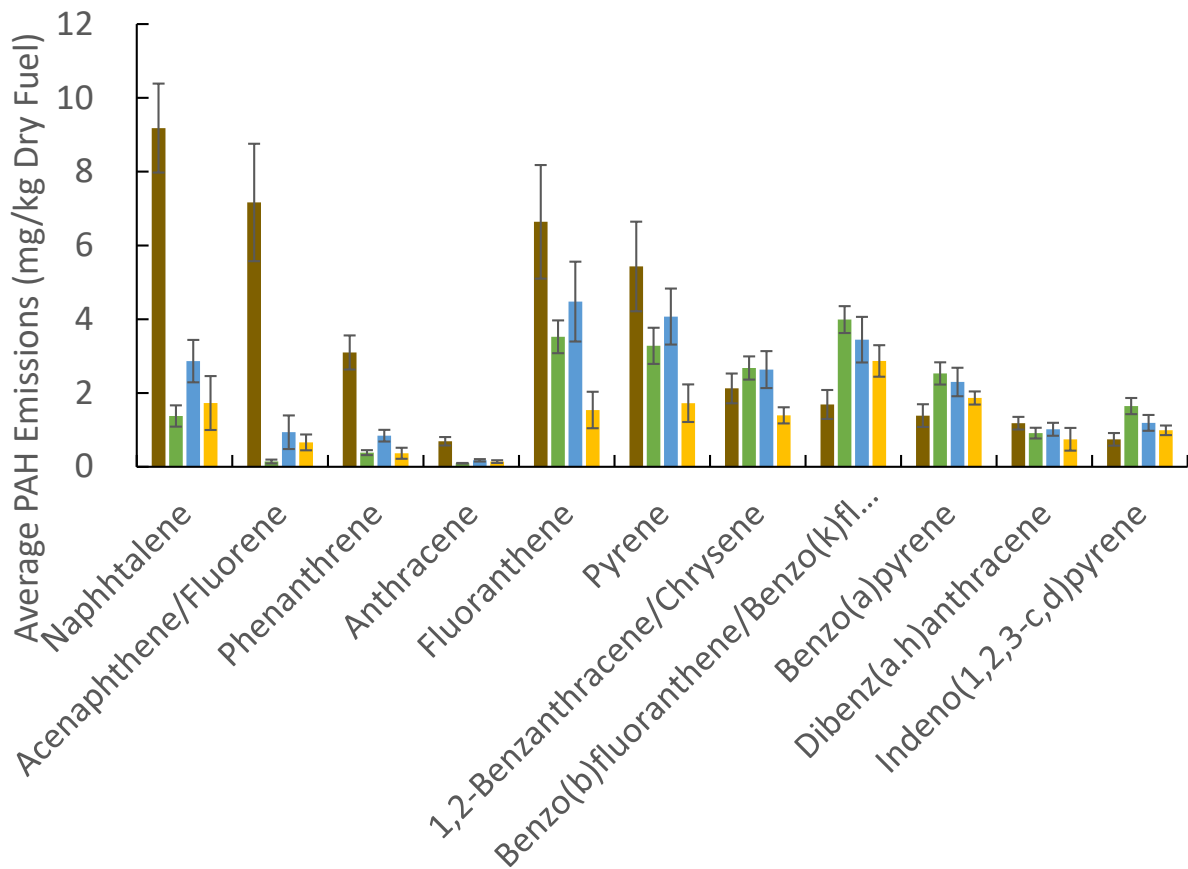


Figure S3: Particulate PAH emissions by species as a bar chart. Average particulate PAH emissions by species on a mass of PAH per kg of dry fuel burned for the Angithi stove (brown), Chulha with mixed fuels (green), Chulha with dung (blue), and Chulha with wood (yellow). Error bars show one standard error of the mean. Benzo(b) and Benzo(k)fluoranthene are summed together.

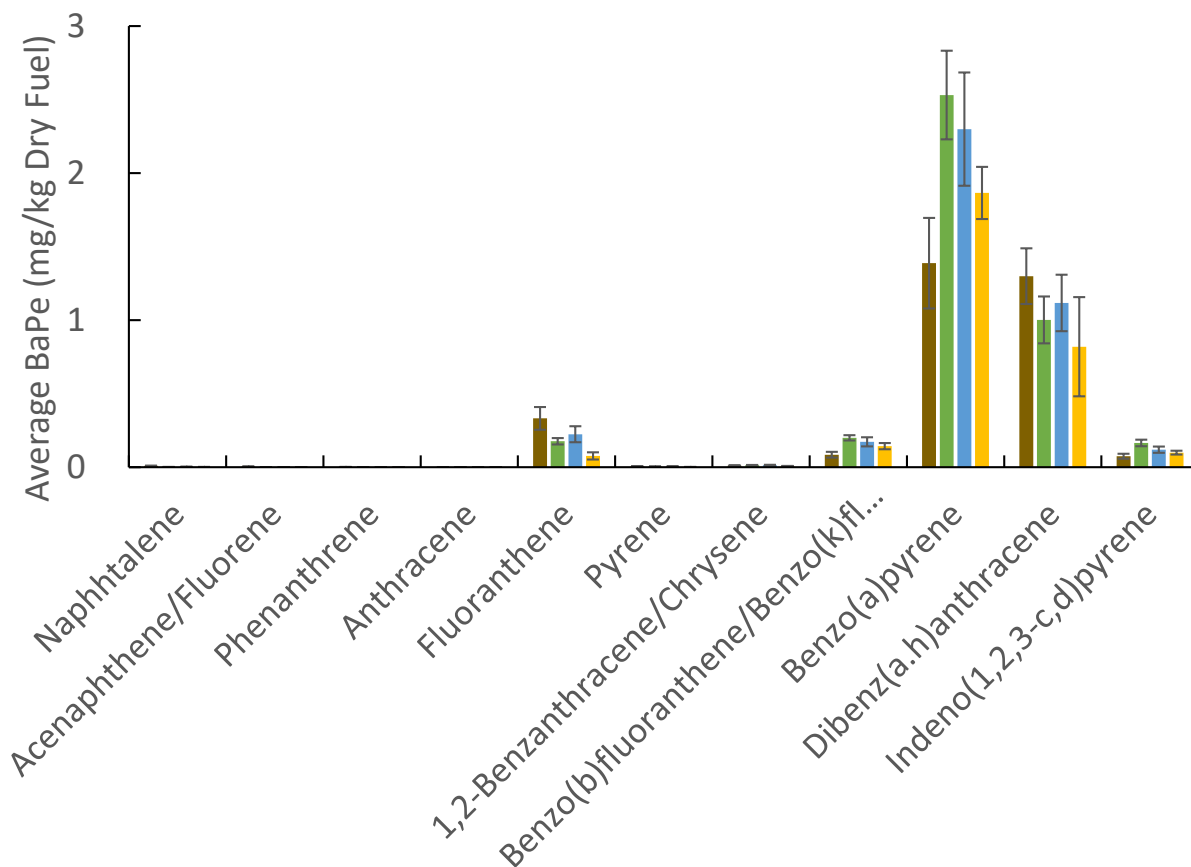


Figure S4: Particulate PAH emissions on a BaPe basis. Average particulate PAH emissions by BaPe per kilogram of dry fuel burned for each species for the Angithi stove (brown), Chulha with mixed fuels (green), Chulha with dung (blue), and Chulha with wood (yellow). Error bars show one standard error of the mean. Benzo(b) and Benzo(k)fluoranthene are summed together.

Table S3: Comparison of HPLC method to GC data from the literature. Concentrations of individual PAH species in standard reference material 1648a, urban particulate matter, measured in this study by HPLC-DAD and values reported by the manufacturer via GC-Soxhlet/pressurized fluid extraction.

PAH Species	HPLC-DAD/Sonication*	GC-Soxhlet/PFE**
Phenanthrene	3.72 ± 0.35	4.86 ± 0.17
Anthracene	0.52 ± 0.02	0.46 ± 0.01
Fluoranthene	6.74 ± 0.24	8.07 ± 0.14
Pyrene	5.81 ± 0.19	5.88 ± 0.07
1,2-Benzanthracene/Chrysene	3.01 ± 0.15	2.71 ± 0.15 / 6.12 ± 0.06
Benzo(b)fluoranthene	7.81 ± 0.19	8.89 ± 0.05
Benzo(k)fluoranthene	2.96 ± 0.14	3.03 ± 0.24
Benzo(a)pyrene	5.18 ± 0.45	2.57 ± 0.10
Indeno(1,2,3-cd)pyrene	5.70 ± 0.35	4.17 ± 0.17

* Means of mass fractions from triplicate samples (extractions in triplicate) with STDV. **Weighted means of the mass fractions from multiple analytical methods with STDV.

Table S4: Recovery after 15-min or 24-h for the PAH species measured in this study on quartz filters. Recovery (in %) of individual PAH species on quartz filters (47 mm) spiked with 1 mL of a 50 ng/mL PAH standard mix, extracted and analyzed by HPLC-DAD following NIOSH Method 5506.

PAH Species	15 Minutes	24 Hours
Naphtalene	63.0 ± 4.0	< 10%
Phenanthrene	84.4 ± 5.3	< 10%
Anthracene	83.1 ± 3.0	< 10%
Fluoranthene	91.1 ± 2.4	< 10%
Pyrene	97.9 ± 1.7	< 10%
1,2-Benzanthracene/Chrysene	104.3 ± 1.1	64.5 ± 1.9
Benzo(b)fluoranthene	102.6 ± 1.5	94.9 ± 2.8

Benzo(k)fluoranthene	104.1 ± 1.4	99.7 ± 2.8
Benzo(a)pyrene	69.6 ± 9.8	91.4 ± 5.5
Dibenz(a,h)anthracene	99.6 ± 1.5	99.8 ± 3.0
Indeno(1,2,3-c,d)pyrene	91.4 ± 6.4	101.5 ± 4.1

Table S5: Toxic equivalency factors as benzo-a-pyrene equivalency (BaPe) for individual PAH species presented in this paper.

PAH Species	Toxic Equivalency Factor (BaPe)
Naphthalene	0.001
Acenaphthene/Fluorene	0.0005
Phenanthrene	0.0005
Anthracene	0.0005
Fluoranthene	0.05
Pyrene	0.001
1,2-Benzanthracene/Chrysene	0.005
Benzo(b/k)fluoranthene	0.05
Benzo(a)pyrene	1
Dibenz(a,h)anthracene	1.1
Indeno(1,2,3-c,d)pyrene	0.1