Exposures to and origins of carbonaceous PM_{2.5} in a cookstove intervention in Northern Ghana

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Abstract

REACTING (Research on Emissions Air Quality, Climate, and Cooking Technologies in Northern Ghana) was a 200-home cookstove intervention study from 2013-2015. Study households were divided into four groups: a control group, a group given two locally made rocket stoves, a group given two Philips forced draft stoves, and a group given a locally made rocket stove and a Philips stove. In a subset of study households, 48-hour PM_{2.5} exposure samples were collected for adults and children, as well as in the primary cooking area. Further, weekly ambient background PM_{2.5} samples were collected for the first nine months of the study. All PM_{2.5} samples were analyzed for elemental and organic carbon (EC/OC), and a subset was also analyzed for organics. Mixed effects modeling was applied to quantify differences in PM exposures between the groups and to assess relationships between exposures and cooking area measurements. Results showed that personal OC exposure for the intervention groups was 56.6% lower than the control group ($p \leq 0.01$). Both intervention groups given Philips stoves had significantly lower EC exposure than the control group (60.6% reduction, $p \leq 0.02$). Only weak relationships were found between personal and cooking area EC or OC. Source apportionment modeling was performed on both the personal/microenvironment and the ambient organics PM_{2.5} data sets to assess the sources of the observed PM. We identified six PM sources. The identified source factors were similar among the data sets, as well as with previous work in Navrongo. Two sources, one characterized by the presence of methoxyphenols, and one by the presence of polycyclic aromatic hydrocarbons and EC, were associated with biomass burning, and accounted for a median of 9.2% of OC and 15.3% of EC personal exposure. Here, we demonstrate the utility of using the cooking-related source apportionment factors

within a mixed effects model for more precise estimation of exposures due to cooking, rather than other combustion sources unrelated to the intervention.

1. Background and motivation

Globally, 2.8 billion people cook with biomass (Bonjour et al., 2013), and pollution from household biomass combustion is the third highest risk factor for the global burden of disease (Lim et al., 2012). Particulate matter with aerodynamic diameter under $2.5\mu\text{m}$ ($\text{PM}_{2.5}$) is a major component of biomass combustion emissions, and various components, including elemental carbon (EC) and organic carbon (OC) have been linked with negative health outcomes (Naeher et al., 2007; Smith et al., 2009; Jansenn et al., 2011). The steep $\text{PM}_{2.5}$ exposure-response curve at low exposures (Burnett et al., 2013) requires drastically cutting $\text{PM}_{2.5}$ exposure in order to achieve health benefits. In contexts where cooking-related emissions are a dominant source of $\text{PM}_{2.5}$ exposures, reducing use of traditional biomass stoves in favor of cleaner alternative technologies may be an effective solution for reducing exposures but likely requires substantial reductions in use (Johnson and Chang, 2015). Quantifying the extent to which exposures are actually reduced when cooking practices change, and understanding how much cooking-related sources drive $\text{PM}_{2.5}$ exposure in different contexts, are key challenges for this field.

Here, we add to the cookstove pollution exposure literature by assessing various effects of a cookstove intervention during the REACTING study (Research on Emissions Air Quality, Climate, and Cooking Technologies in Northern Ghana; Dickinson et al., 2015). Specifically, we present the results from carbonaceous $\text{PM}_{2.5}$ observations from personal, microenvironment, and ambient samples. We focus on EC and OC because they tend to constitute a substantial portion of total $\text{PM}_{2.5}$, are strongly linked to negative health effects, and also because of their importance for regional and global climate (Bond et al., 2013; Ramanathan and Carmichael, 2008). We quantified the carbonaceous components of $\text{PM}_{2.5}$ from 194 personal exposure samples, 127 samples taken in the cooking area and 50 ambient samples. The personal exposure

measurements were then compared to cooking area microenvironment $PM_{2.5}$ and ambient $PM_{2.5}$ levels in an attempt to determine the relationship between personal and microenvironment $PM_{2.5}$. This relationship is an important component of modeling population level exposures with microenvironment sampling, and previous studies that have explored this relationship have produced mixed results depending greatly on specific environmental and behavioral factors (Dionisio et al., 2012; McCracken et al., 2013; Van Vliet et al., 2013).

We then use source apportionment to determine the contribution of various $PM_{2.5}$ sources to the observations at each scale, using a procedure similar to Larson et al. (2004). Many studies investigating exposures to PM from residential biomass combustion assume that differences in observed exposures are purely from cooking activities, after controlling for covariates. We take advantage of our multi-scale measurements to quantify the specific sources of the observed $PM_{2.5}$ concentrations and determine the accuracy of this assumption for our study area. Specifically, we calculate contributions of specific pollution sources to exposures, and use mixed effects modeling to determine whether changes in stove technologies impact those exposures. In-field observations are essential for this purpose, as various works have shown that improved cookstoves perform much differently, and typically better, in controlled laboratory settings than in the field (Johnson et al., 2008; Roden et al., 2009), due to factors such as stove maintenance, training on stove use, and fuel type and preparation. Our methods allow us to directly identify the fraction of carbonaceous $PM_{2.5}$ exposure contributed by cooking-related sources in the field, and can therefore directly assess the intervention effect.

1.1. Relevant previous works

Our approach draws on prior studies examining personal PM exposure in cookstove intervention studies, personal exposure to PM in the region, cooking studies in the region, ambient PM measurements in the region, and source apportionment studies in the region.

Personal PM_{2.5} exposure measurements in previous studies investigating impacts from cookstove interventions have been limited, with varied associated exposure and health outcomes based on differences in a multitude of factors such as fuel choices, regional cooking traditions, behaviors, home design, and climate. Clark et al. (2013) highlights this variability within and between personal exposure and kitchen microenvironment measurements and associated health outcomes from selected cookstove studies around the world. Cynthia et al. (2008) found significantly lower PM_{2.5} exposure for women with homes supplied with a Patsari stove than women in the control group, in Michoacán, Mexico. In Guatemala, McCracken et al. (2007) found significantly lower PM_{2.5} personal exposures and blood pressure for women participants with a plancha stove compared to participants with traditional stoves.

Limited personal PM_{2.5} sampling has been conducted in Ghana. Van Vliet et al. (2013) measured personal, kitchen area, and ambient PM_{2.5} mass and black carbon (BC) in central Ghana, where study participants cook with traditional methods similar to those seen in our study. That study reported average personal exposure to BC of 8.8 µg/m³ and average cooking area BC of 14.5 µg/m³. Arku et al. (2015) performed personal PM_{2.5} sampling in Accra, the national capital located along the coast in Southern Ghana, as part of a 'spatial sources and exposure' study. That work found average PM_{2.5} mass concentrations of 56 µg/m³, with differences in exposure based on gender and neighborhood affluence. Pennise et al. (2009) reported a large and significant reduction in cooking area PM_{2.5} concentrations in a cookstove intervention study using a Gyapa stove similar to the one used in this study.

Of the limited ambient PM studies in Ghana, four have published PMF source apportionment results, all using metals analyzed with x-ray fluorescence and EC/OC. Three of those four studies were undertaken in or around Accra. Ofosu et al. (2012) performed a 6-month sampling campaign in 2008, in an Accra suburb. Eight PMF factors were identified; industrial activities, aged sea salt, fresh sea salt, biomass combustion, diesel combustion, two-stroke engine combustion, gasoline combustion, and soil dust. Zhou et al. (2013; 2014) performed ambient and cooking area PM_{2.5} sampling in four

neighborhoods throughout Accra, identifying six PMF sources; solid waste burning, road dust and vehicle, aged biomass, fresh biomass, sea salt, and crustal. Aboh et al. (2009) performed ambient $PM_{2.5}$ and $PM_{10-2.5}$ sampling in a suburb of Accra, from February 2006 to February 2007, also identifying six factors. The fourth work collected $PM_{2.5}$ and $PM_{10-2.5}$ from 2009-2010 in Navrongo, Northern Ghana (Ofosu et al. 2013), at a site 2 km from the Navrongo Health Research Center, where our ambient sampler was located (Figure 1). That work identified six factors, and provides useful comparisons with our work.

In this study, positive matrix factorization (PMF, Paatero et al., 1997) was used to determine sources of organics $PM_{2.5}$ measured in both the personal-level and ambient samples, aiding the assessment of the intervention effects. Additionally, to our knowledge, organics speciation of $PM_{2.5}$ has not been performed in West Africa, although Bortey-Sam et al. (2015) did collect and speciate a few organic compounds in PM_{10} samples in two sites near Kumasi, the largest city in Ghana, about 500km South of Navrongo. The use of organics here allows us to provide better context to past results by using different tracer species specific to sources (Cass, 1998; Schauer et al., 1996). This work also provides unique data for the less populated and developed rural region of Northern Ghana, and specifically, in relation to a cookstove intervention.

Figure 1. Map of the study region at various scales. At right, the town of Navrongo, with markers on the Navrongo Health Research Center, the base of operations and where ambient $PM_{2.5}$ was sampled (Google Maps, 2016).

2. Methods

This work was conducted in the context of the REACCTING study, a 200-household, cookstove intervention that took place from November 2013 through January 2016 in the Kassena-Nankana districts of Northern Ghana (Figure 1) (Dickinson et al. 2015). The study was conducted in the Kassena-Nankana districts of the upper east region of northern Ghana, with fieldwork based out of the Navrongo Health Research Centre (NHRC). Details on the study region are presented by Oduro et al. (2012) and Dickinson et al. (2015).

The REACCTING study households were divided into four groups of 50 households each, with one arm given two locally-made Gyapa rocket stoves (Gyapa/Gyapa), one arm given two Philips HD4012 LS stoves (Philips/Philips), one arm given one of each (Gyapa/Philips), and the last arm serving as control until the end of

the study, when they were given their choices of two stoves. The improved cookstoves tested in this study were aimed at reducing household fuel use and cooking-related hazardous air pollutant emissions.

2.1. Personal PM_{2.5} sample collection

Study participants at four households each week were asked to wear a PM_{2.5} filter sampler over a 48-hour period. Typically, the participants included the mother and a child over age four. They were asked to wear the sampling pack at all possible times, except during sleep, or when they were stationary, in which case the pack was to be kept within arms reach. The sampling packs were backpacks for children, and waist packs for the adults, as they found that style more convenient and appropriate. Sampling periods started early in the morning, usually before the first meal.

Personal filter samples were collected using Teflon URG impactors (#URG-2000-25F, Chapel Hill, NC, USA) operating at 2 L min⁻¹ to obtain a 2.5µm size cut. Samples were collected on pre-baked Pall TissuQuartz 2500-QAT 25mm filters. These filters were selected in order to perform carbonaceous PM_{2.5} analysis, due to our interest in chemical analysis of the PM_{2.5}, and because the majority of PM_{2.5} from wood combustion is carbonaceous (Kleeman et al., 1999; Schauer et al., 2001). Total PM_{2.5} mass was not measured, since this would require samples on both quartz and Teflon filters, which would have been logistically challenging and was not the primary objective of this study. All filters were transported in pre-baked sterile glass jars from the University of Colorado to the NHRC, and during sampling operations in Ghana. Used and unused filters were stored in a refrigerator onsite, and effort was made to keep them cool during transport.

Filter packs were cleaned biweekly with isopropyl alcohol. SKC AirLite pumps powered by 6000 mAh lithium ion battery packs provided the vacuum, and flow rates were adjusted weekly using a calibrated rotometer. Field blanks were collected every 48-hour period, and were carried to the sampling sites. EC, OC, and organics were median blank subtracted using each filter storage jar's blanks for the final results.

From November 2013 through August 2015, 260 personal filter samples were collected. 66 filters were discarded due to the sampler battery running out by the time the field team returned to finish the sampling period. A summary of key sample demographics is shown in Table 1. The sampling schedule was designed to balance observations across stove groups. However, logistical constraints resulted in fewer samples from the Gyapa/Philips group. EC and OC were analyzed on all samples that passed quality assurance (191), and speciated organics were analyzed for all samples collected from November 2013 to February 2014 (53).

Table 1. Household and seasonal statistics for the personal exposure (194, 89 unique individuals) and microenvironment samples (127).

		Personal Exposure Samples		Microenvironment Samples		Ambient Samples	
		<u>All samples:</u> 191 samples from 88 unique individuals		<u>All samples:</u> 127 samples from 37 unique households		<u>All samples:</u> 51 samples	
		<u>Organics subset (in parentheses):</u> 37 samples from 37 unique individuals		<u>Organics subset (in parentheses):</u> 16 samples from 16 unique households		<u>Organics subset (in parentheses):</u> 25 samples	
Individual-level covariates	Age						
	Minimum	3	(5)				
	Median	26	(17)				
	Maximum	73	(41)				
	Gender						
	Female	148	(30)				
	Male	46	(7)				
	Gender/Age						
	F<10 years	20	(5)				
	F>10 years	128	(25)				
	M<10 years	31	(5)				
	M>10 years	15	(2)				
Household-level covariates	Stove group						
	Gyapa/Gyapa	48	(6)	42	(5)		
	Philips/Philips	74	(14)	42	(5)		
	Gyapa/Philips	27	(5)	24	(4)		
	Control	45	(12)	19	(2)		
	Language						
	Household size						
	Minimum	3	(4)	3	(4)		
	Median	8	(7)	8	(8)		
	Maximum	18	(17)	18	(17)		
	Socioeconomic status						
	Least poor	32	(3)	19	(0)		
	Less poor	33	(11)	16	(4)		
	Poor	34	(5)	20	(2)		
	Poorer	73	(13)	48	(6)		
	Poorest	19	(5)	24	(4)		
Seasonal covariates	Seasons						
	Harmattan bush burning	58	(35)	33	(14)	12	(10)
	Heavy Rainy	66	(0)	41	(0)	17	(2)
	Hot dry	26	(0)	16	(0)	6	(5)
	Light Rainy	24	(0)	20	(0)	5	(4)
	Transition	20	(2)	17	(2)	11	(4)

2.2. Microenvironment PM_{2.5} sample collection

Cooking area microenvironment sampling followed the same procedures and used the same equipment as the personal exposure sampling. Cooking area measurements were collected concurrently at the same households with the personal sampling, and also for 48-hour periods (Table 1). The filter samplers were housed within custom battery powered U-pod monitors that also collected gas phase measurements (Dickinson et al., 2015). The U-pods were placed approximately 1 meter

away and 1 meter above the most used cookstove in the primary cooking area (**Error! Reference source not found.**). In all, 137 microenvironment samples were collected from November 2013 to August 2015, with 10 of those discarded for sampling time deviating more than 6 hours from a full sampling day of 24 or 48 hours. The U-pod data logger provided an accurate timer for pump operation. All samples were analyzed for EC/OC, and the 16 filters collected from September 2013 to June 2014 were also analyzed for organics.

Figure 2. Ambient sampling inlet shown at the Navrongo Health Research Center. On the right, typical cooking areas, shown with the U-pod air quality samplers.

2.3. Ambient PM_{2.5} sample collection

Ambient PM_{2.5} samples were collected for one-week periods at the NHRC from November 2013-September 2014. Samples were collected on 90mm TissuQuartz filters with a 2.5µm size cut achieved using a 6 Lmin⁻¹ cyclone (URG-2000-30EHB). The sampler was situated on a second story ledge at the NHRC, 6m from the ground and 0.8m from the wall (**Error! Reference source not found.**). Average flow rate was measured with a mechanical timer coupled with a flow totalizer. The desired 5.5 Lmin⁻¹ flow was periodically disrupted due to power failures and pump failures, thus contributing most to the measurement uncertainty. Filter changing times were inconsistent in some cases, with one filter collected for 33 days in December 2013, and two other samples collected over two weeks each. A total of 50 samples were collected and all were analyzed for EC/OC. 25 samples collected between November 2013 and June 2014 were analyzed for organics and included in PMF analysis.

2.4. Chemical analysis

Details of the bulk OC and EC analysis method applied in this study are provided in Dutton et al. (2009a). A 1.5 cm² punch was taken from each quartz fiber filter and analyzed using a Sunset Laboratory ECOC analyzer operated under NIOSH 5040 thermal optical transmission (TOT) method (NIOSH, 2003; Schauer et al., 2003). The methods for quartz fiber filter extraction and organic speciation are provided by Dutton et al. (2009b) and Xie et al. (2014). Briefly, each quartz fiber filter sample was pre-spiked with a mixture of internal standard containing isotopically labeled standards before extraction. Each pre-spiked sample was extracted twice with methylene chloride ultrasonically. After filtration and concentration, the final extracts were analyzed by gas chromatograph-mass spectrometer (GC-MS). The GC-MS analysis for our sample extracts were performed in sequences along with five dilutions of quantification standards. Quadratic calibration curves were generated from all available runs of quantification standards in each batch of sequences. The mass amount of each target compound on quartz fiber filters was obtained by converting peak area ratios to mass ratios using calibration curves and known mass of pre-spiked internal standards. The

analytical uncertainties associated with calibration curves were estimated within each batch empirically (Dutton et al., 2009b).

The final airborne concentrations of OC, EC and each identified organic compound were determined by dividing their total amount on each filter with the sampled air volume. The final uncertainties were calculated using root sum squares (RSS) method, incorporating the analytical uncertainties, standard deviation of the field blanks and the sample volume uncertainties (Dutton et al., 2009a). All measurements were field-blank corrected by subtracting off the median blank value.

2.5. Source apportionment

PMF2, the program developed by Paatero and coworkers to implement PMF (Paatero et al., 1997) was coupled with a stationary block bootstrap technique to perform the source apportionment (Hemann et al., 2009). PMF resolves factor profiles and contributions from a time series of observations. The PMF2-based tool developed by Hemann and coworkers assigns confidence intervals to the factor contributions as well as the factor profiles, providing a better understanding of the solution stability. In this study, solutions with 3 – 7 factors were considered. The final factor number applied in this study was determined based on interpretability of each solution as well as fit diagnostics.

All organics species with greater than 40% of values below detection limits, or signal-to-noise ratios below two, were removed from the PMF analysis data set. For the ambient data set, 19 organics species were retained due to the low number of samples available for analysis, to reduce rotational ambiguity. The personal and microenvironmental (P-M) samples were combined, since we are fundamentally interested in identifying typical pollution sources experienced by people in their homes. For the P-M data set, 41 observed chemical species were retained for analysis. The species were selected based on their representativeness of factor/sources in a previous source apportionment study (Xie et al., 2013). All missing values of individual species were replaced by the geometric mean of the remaining measurements, and their

corresponding uncertainties were set to four times the geometric mean. Similarly, the BDL values were set to half the detection limit, with uncertainties set at five-sixths the detection limit (Polissar et al., 1998; Reff et al., 2007). Six samples with high sample volume uncertainty were retained for the PMF solution, yielding a total of 59 samples, but were removed for subsequent comparisons and modeling.

2.6. Mixed effects model specification

Mixed effects models were applied to quantify relationships between observed $PM_{2.5}$ concentrations and various study covariates, and allow for inference about future measurements and outcomes. These mixed effects models allow us to determine how certain covariates (such as stove group or gender) impact $PM_{2.5}$ measurements. Repeated participant samples produce inherent correlations between the measurements, which can impact the estimated effects of the covariates and possibly skew the results. The mixed effects models allow us to account for this correlation due to the repeated samples, so that we can accurately assess the impact of the different covariates on the exposure levels. Additionally, these models can estimate two different sources of variation within our data: the within person variance (i.e. the variation within an individual, which is related to the correlation) and the between-participant variance components (i.e., how the measurements vary from person to person) (Zeger et al., 1988). Estimating these variance components is helpful in exposure studies to identify exposure reduction strategies (Peretz et al., 2001), and is also used for estimation of health effects in exposure-response studies since effect attenuation (biasing the regression slope towards zero) increases with the within-to-between variance ratio, as noted by Clark et al. (2013).

Separately, mixed effects models were fit for the personal exposure EC and OC data sets, to assess the differences in exposures between study groups. The dependent variables (EC or OC) were log transformed to satisfy the required normality assumption needed for linear modeling. We estimated the effects of the study arm (i.e. stove group) on EC or OC exposure levels, while also adjusting for season, a gender/age

interaction variable, the number of family members, and socioeconomic status. We allowed subjects to have a variable baseline exposure level (i.e. we included a random intercept), but we assumed that the degree of change in exposure levels were the same from person-to-person (i.e., we did not include a random slope), as there was no evidence of this produced by the data. This model is described by Equation 1:

$$\ln(Y_{ij}) \sim \theta_0 + \theta_1(\text{stovegroup})_i + \theta_2(\# \text{ in family})_i + \theta_3(\text{gender*age})_i + \theta_4(\text{season})_i + \theta_5(\text{SES})_i + \theta_6(\text{primary cook})_i + \alpha_j + e_{ij} \quad (1)$$

where Y_{ij} represents EC or OC on sample day i for individual j . ‘Gender*age’ is a categorical variable that is defined as male/female and over/under 10 years old. ‘Season’ is a categorical variable defined as ‘light rainy’ (April-June), ‘heavy rainy’ (June-October), ‘transition’ (only October), ‘Harmattan’ (November to mid-February), and ‘hot dry’ (mid-February to April). Socioeconomic status (SES) is a categorical variable categorized as ‘poorest’, ‘very poor’, ‘poor’, ‘less poor’, and ‘least poor’ (as by Awini et al, 2010), calculated for each household using data from a district-wide Health and Demographic Surveillance Survey (Oduro et al 2012). ‘Primary cook’ is a categorical variable for whether the person monitored was listed as the household’s primary cook, as determined at baseline (Dickinson et al., 2015). α_j represents the random intercept that accounts for the correlation within subjects, and e_{ij} represents the variation from subject to subject. In addition to the covariates mentioned above, we also investigated a ‘cooking area geometry’ variable, but it was not included in the final analysis because it was found to be collinear with the SES variable. This is a logical finding, as the poorest households tended to have no indoor cooking areas, and the wealthier households generally had more indoor and semi-enclosed cooking areas.

Mixed effects models were also applied to the cooking area microenvironment $\text{PM}_{2.5}$ EC and OC, though the model was different, with the random intercept included at the household level (instead of at the personal level), and with the gender and age categories removed (Equation 2). 87 unique household EC and OC samples were available for analysis.

$$\ln(Y_{ij}) \sim \theta_0 + \theta_1(\text{stovegroup})_i + \theta_2(\# \text{ in family})_i + \theta_3(\text{season})_i + \theta_4(\text{SES})_i + \theta_6(\text{primary cook})_i + \alpha_j + e_{ij} \quad (2)$$

Finally, to relate personal and cooking area microenvironment carbonaceous PM_{2.5}, mixed effects models were applied to the EC and OC independently. Here, the model (Equation 3) was the same as Equation 1, but with the addition of the log-transformed microenvironment EC and OC average mass concentrations, and a cross-term between the mass concentrations and stove groups, to account for potential changes in the relationship due to changes in emissions or cooking behaviors by group.

$$\ln(Y_{ij}) \sim \theta_0 + \theta_1 \ln(\text{MicroEnv})_i + \theta_2 (\text{stovegroup})_i + \theta_3 (\# \text{ in family})_i + \theta_4 (\text{season})_i + \theta_5 (\text{SES})_i + \theta_6 (\text{primary cook})_i + \alpha_j + e_{ij} \quad (3)$$

Of the 191 personal exposure PM_{2.5} samples, 141 were available with simultaneous microenvironment PM_{2.5} measurements for use in this model.

3. Results and discussion

3.1. Personal carbonaceous PM_{2.5}

Summary statistics for the EC, OC, and organics are shown separately for personal and microenvironmental samples in Table 2. Mean personal OC concentration was 38.9 µg/m³ (±54.3 µg/m³), while mean EC was 2.8 µg/m³ (±10.5 µg/m³). 0.8% of OC and 5.4% of EC samples were below the instrument detection limit. 63 of the 191 48-hour samples (32%) registered above the WHO 24-hour interim-1 maximum average of 35 µg/m³. As we only assess the carbonaceous components, it is an underestimate of the total PM_{2.5} mass concentration. A distribution of the sum of EC and OC is presented in Figure 3.

The observed average PM_{2.5} personal exposures in REACCTING appear lower than comparable previous works, although it must be emphasized that here the sum of EC and OC is reported, while the following works used total gravimetric PM_{2.5} mass. In Ghana, personal PM_{2.5} sampling was performed on high school students in Accra, Ghana, (Arku et al., 2015), where they found average 24-h concentrations of 56 µg/m³ (±33.5 µg/m³). In regards to cookstove study-related personal exposures, McCracken et al. (2007) reported daily average PM_{2.5} concentrations from rural homes in Guatemala of 264 µg/m³ for the control group, and 102 µg/m³ for homes with plancha stoves.

Baumgartner et al. (2011) measured PM_{2.5} exposures in rural Yunnan China, finding average concentrations for adult women of 117 µg/m³ in winter, and 55 µg/m³ in summer. Dionisio et al. (2012) measured integrated PM_{2.5} on 31 children in The Gambia, reporting average 48-h exposures of 65 µg/m³.

Figure 3. Personal exposure distributions for summed EC and OC, compared with relevant WHO standards.

Table 2. Summary of personal carbonaceous PM_{2.5} EC, OC, and organics samples. Flagged samples were removed from analysis due to various possible issues including sampling duration and contamination.

All personal EC and OC PM _{2.5} statistics (n = 191)						
	Mean	Median	Std dev	Geo mean	%BDL	Flagged samples
OC (µg/m ³)	39.7	23.1	54.5	24.3	0.8	66
EC (µg/m ³)	2.8	1.1	10.6	1.0	5.4	66
EC/OC	0.2.8	0.0.1	10.5	1.0	5.4	66
All cooking area microenvironment PM _{2.5} EC and OC statistics (n = 127)						
	Mean	Median	Std dev	Geo mean	%BDL	Flagged samples
OC (µg/m ³)	108.0	54.2	117.9	61.2	0.0	10
EC (µg/m ³)	8.3	3.4	12.5	3.3	5.1	10

EC/OC 0.2.8 0.1.1 10.5 1.0 5.4 66

PMF personal and microenvironment PM_{2.5} dataset statistics (n = 59)

<i>*units in ng/m3 unless noted</i>	Mean	Median	Std dev	Geo mean	%BDL	S/N
OC (µg/m3)	59.4	40.9	69.0	43.6	0.0	10.0
EC (µg/m3)	5.4	2.2	10.3	2.9	0.0	10.0
docosane (C22)	4.3	2.2	9.5	2.1	1.7	8.1
tricosane (C23)	10.2	4.2	27.7	4.4	0.0	8.5
tetracosane (C24)	16.8	4.7	64.9	4.8	0.0	4.7
pentacosane (C25)	24.7	8.4	81.1	9.3	0.0	7.5
hexacosane (C26)	21.6	7.9	65.5	8.3	0.0	8.5
heptacosane (C27)	32.7	14.2	72.0	15.1	0.0	9.8
octacosane (C28)	24.3	10.8	59.0	11.4	0.0	9.6
nonacosane (C29)	56.4	25.4	146.6	28.1	0.0	10.0
triacontane (C30)	14.6	9.4	16.9	9.3	0.0	9.2
hentriacontane (C31)	57.2	23.0	149.4	25.5	0.0	9.9
dotriacontane (C32)	11.7	6.7	15.5	7.4	0.0	8.8
tritriacontane (C33)	19.3	15.0	20.3	13.8	0.0	9.7
tetratriacontane (C34)	6.3	4.0	8.4	3.9	0.0	7.8
pentatriacontane (C35)	11.0	5.8	18.6	6.4	0.0	8.5
hexatriacontane (C36)	7.3	4.4	11.1	4.3	0.0	8.0
heptatriacontane (C37)	4.0	2.3	5.9	2.5	6.9	6.8
octatriacontane (C38)	3.6	2.1	5.2	2.4	19.0	4.9
nonatriacontane (C39)	3.7	2.2	5.0	2.5	24.1	4.4
tetracontane (C40)	3.3	2.0	4.5	2.2	36.2	3.5
fluoranthene	8.9	1.8	27.2	2.0	0.0	9.5
pyrene	11.5	2.0	36.6	2.2	0.0	9.2
benzo[ghi]fluoranthene	12.6	1.5	39.6	1.6	0.0	9.4
cyclopenta[cd]pyrene	16.2	1.3	56.7	1.6	0.0	7.8
benz[a]anthracene	16.9	1.7	49.9	2.0	0.0	8.0
chrysene/triphenylene	17.6	2.1	46.0	2.8	0.0	8.3
benzo[b&k]fluoranthene	31.2	7.5	73.2	7.6	0.0	9.8
benz[a&e]pyrene	26.8	5.7	67.7	6.2	0.0	9.5
indeno[1,2,3-cd]pyrene	9.0	1.9	24.1	2.1	0.0	7.9
benzo[ghi]perylene	10.3	2.8	26.6	2.9	0.0	7.5
coronene	5.2	1.1	14.6	1.1	0.0	8.3
2-methylfluoranthene	20.6	2.6	65.2	3.1	0.0	8.6
methyl-202-PAHsum	65.6	7.6	207.3	9.7	0.0	8.7
anthracene-9,10-dione	2.5	0.5	6.4	0.5	12.1	5.6
benz[de]anthracene-7-one	8.9	1.9	20.3	2.3	1.7	8.3
ba-30-norhopane	1.5	1.3	0.8	1.3	37.9	3.3
ab-hopane	1.1	0.9	0.7	0.9	37.9	4.1
syringaldehyde	261.0	49.6	679.8	65.1	0.0	9.1
coniferaldehyde	119.8	18.5	321.6	25.0	3.4	5.5
acetosyringone	63.1	13.4	158.3	15.2	0.0	6.7

3.1.1. Personal OC PM_{2.5} modeling results

Results from Equation 1 showed significant reductions in personal PM_{2.5} OC exposure for the three intervention groups over the control group (Table 3. Full model output is presented in the Supplementary Information Section 2). Average OC exposures were 57.3% lower for the Gyapa/Gyapa group (27.8 µg/m³, 95% confidence interval (15.2, 50.9), p = 0.01), 49.4% lower for the Philips/Philips group (32.9 µg/m³, (19.6, 55.2), p = 0.01), and 63.2% lower for the Gyapa/Philips group (23.9 µg/m³, (12.0,

47.7), $p < 0.01$) relative to the control groups ($65.0 \mu\text{g}/\text{m}^3$, (28.6, 147.8)). Women listed as ‘primary cooks’ had a 60.4% higher average exposure than other individuals ($p = 0.09$). Analysis of the SES variable showed that the two poorest groups (‘poorer’ and ‘poorest’) had higher predicted exposures than the ‘poor’ group: 62.8% higher for the ‘poorer’ group ($p = 0.05$), and 116.0% higher for the ‘poorest’ group ($p = 0.04$), possibly indicative of an effect on exposure levels due to fuel choices, behaviors, or home types. There was no significant effect by number of family members. The random effect by individual was significant at 95%, with between-participant variance of 0.86, while the within-participant variance was 0.50.

Table 3. Summary of personal exposure model results

	OC (N = 191)		EC (N = 191)		PAH Factor (N = 37)		Methoxyphenol Factor (N = 37)	
	Coefficient	P value	Coefficient	P value	Coefficient	P value	Coefficient	P value
Intercept	4.17	0.00	0.91	0.04	6.44	0.00	5.88	0.00
Gyapa/Philips	-1.00	0.00	-0.90	0.02	-0.27	0.85	-1.53	0.26
Philips/Philips	-0.68	0.01	-0.97	0.00	-4.00	0.00	-1.57	0.11
Gyapa/Gyapa	-0.85	0.01	-0.58	0.07	0.10	0.94	-0.50	0.70
Family members	-0.02	0.53	0.03	0.33	0.15	0.29	0.05	0.69
Primary cook	0.47	0.09	0.27	0.34	0.34	0.77	1.54	0.16
Over 10y					0.18	0.89	0.46	0.70
Females > 10y	-0.71	0.06	-0.24	0.53				
Males < 10y	-0.30	0.39	-0.23	0.55				
Males > 10y	-0.25	0.59	0.37	0.45				
Heavy rainy season	-0.42	0.01	-0.71	0.00				
Hot dry season	-0.13	0.56	-0.79	0.01				
Light rainy season	-0.42	0.08	-1.96	0.00				
Transition season	-0.37	0.15	-0.68	0.03				
Poorer	0.52	0.05	0.32	0.23				
Less poor	-0.24	0.46	-0.32	0.33				
Least poor	0.38	0.22	0.20	0.54				
Poorest	0.79	0.04	0.70	0.08				

There were seasonal effects, with 34.6% lower average exposure levels during the heavy rainy season ($p = 0.01$) and 34.5% lower during the light rainy season ($p = 0.08$), relative to the Harmattan season when there are high winds that generate large amounts of dust, and there is substantial regional bush burning. Although fuel moisture content and behavior change may be expected to increase exposures during rainy seasons, the results here are consistent with other findings. L’Orange et al. (2012) found inconsistent and insignificant differences in cookstove PM emission dependence from 4-30% fuel moisture content, suggesting that during the rainy periods, a higher PM emission rate should not be assumed. In this work, fuel wood moisture was measured with a General Tools MMD4E moisture meter at a subset of homes that had fuel on

hand during personal exposure measurement periods. Three moisture measurements were recorded for each wood size range, with categories of small (<1cm diameter), medium (1-3cm diameter), and large (>3cm diameter). Fuel moisture content was higher during both rainy seasons (SI Figure 1), but values were generally between 5-20% and 5-30% for the most commonly used small and medium sized pieces, respectively, in line with the range reported by L'Orange et al. Therefore, we can expect no substantial seasonal differences in PM emissions due to fuel moisture.

In terms of behavior change, it is common for cooking location to shift to a covered area if there is rain, likely increasing the cook's exposure if they remain there for the duration of cooking. However, exposure of others members in the household could decrease since rainy-weather cooking takes place in covered or enclosed areas, usually near the perimeters of courtyards, away from where most people spend time. Lower rainy season exposures could also result from PM removal by the rain, seasonal fuel switching, and shifts in the relative importance of regional PM sources.

3.1.2. Personal EC PM_{2.5}

Average exposures for PM_{2.5} EC (Equation 1) were 44.3% lower for the Gyapa/Gyapa group (1.4 $\mu\text{g}/\text{m}^3$ (0.7, 2.6), $p = 0.07$), 62.0% lower for Philips/Philips group (1.0 $\mu\text{g}/\text{m}^3$ (0.6, 1.6), $p < 0.01$), and 59.3% lower for the Philips/Gyapa group (1.0 $\mu\text{g}/\text{m}^3$ (0.5, 2.1), $p = 0.02$), relative to the control group (2.5 $\mu\text{g}/\text{m}^3$ (1.0, 6.0)). Within-participant variance was 0.30 (significant at 95%) and between-participant variance was 1.12.

The results of our model are reasonable, given that substantially lower EC is emitted by Philips stoves than both natural draft rocket stoves and traditional stoves, as previously shown in field measurements reported by Kar et al. (2012). However, the differences are somewhat surprising given the results from Piedrahita et al. (2016), who show that there was a continued high use rate of traditional stoves for all intervention groups (between 48.7% and 73.0% of homes surveyed reported using traditional stoves on the previous day), substantial stove stacking of the intervention stoves and traditional stoves, and reduced usage of the Philips stoves over time. The lower

exposures of either group given Philips stoves over the Gyapa/Gyapa group is also noteworthy because the Gyapa use rate was substantially higher than the Philips use rate. Reductions in $PM_{2.5}$ EC and OC exposures must then be due to the differences in the emissions from the various stove types, changes in behavior that affected exposure, and longer duration cooking on traditional stoves in the control homes than in the intervention homes.

There were no significant differences in the EC exposures among participants of different ages or gender, or by number of family members. EC exposure for the primary cook was 31% higher than non-cooks ($p = 0.34$). All seasons had significantly lower EC than the Harmattan season, likely due to the regional biomass burning during that period. The poorest group had higher expected EC exposures than the poor group ($p = 0.08$), but there were no significant differences by SES category at the 95% level (Table 3, SI section 3).

Van Vliet et al. (2013) reported average personal exposure to black carbon (BC) of $8.8 \mu\text{g}/\text{m}^3$ (7.4, 10.3) in rural Central Ghanaian homes cooking with wood and/or charcoal, much higher than the EC measured here. In that study, all participants were the home's primary cooks and there was more reported kerosene use and indoor cooking than in this study. Additionally, the BC measurement in that work was made using an optical reflectance method, which has previously reported higher values than the traditional thermo-optical EC measurement method in side-by-side comparisons (Yan et al., 2011).

3.2. Microenvironmental carbonaceous $PM_{2.5}$

Average cooking-area microenvironment $PM_{2.5}$ OC using Equation 2 was 68.6% lower for the Gyapa/Gyapa group ($33.3 \mu\text{g}/\text{m}^3$ (16.7, 66.4), $p < 0.01$), 45.5% lower for Philips/Philips group ($49.6 \mu\text{g}/\text{m}^3$ (25.3, 97.4), $p = 0.04$), and 40.4% lower for the Philips/Gyapa group ($57.3 \mu\text{g}/\text{m}^3$ (25.2, 130.7), $p = 0.18$), relative to the control group ($100.2 \mu\text{g}/\text{m}^3$ (41.7, 240.4), $p < 0.01$). There was no effect for OC by number of family members in the home, and the seasonal variable results were not consistent with the personal exposure results. Here, only the 'transition' season was significantly higher

(171.9% higher) relative to the 'Harmattan bush burning' season, while the 'heavy rainy' season had a positive effect ($p = 0.06$), and the 'hot and dry' and 'light rainy' seasons had negative and not significant effects on cooking area OC $PM_{2.5}$.

Cooking area microenvironment $PM_{2.5}$ EC results were generally consistent with the OC results, with negative effects for the Gyapa/Gyapa ($4.2 \mu g/m^3$ (1.9, 9.4), $p = 0.28$), Philips/Philips ($3.3 \mu g/m^3$ (1.5, 7.2), $p = 0.09$), and Gyapa/Philips ($5.1 \mu g/m^3$ (2.0, 13.0), $p = 0.6$) groups relative to the control group ($6.5 \mu g/m^3$ (2.3, 19.0)). The seasonal effects had the same effect directions as for the OC model, with the 'light rainy' and 'hot and dry' seasons lower than the 'Harmattan bush burning' season by 64.2% ($p = 0.03$) and 55.9% ($p = 0.11$), respectively. It is important to note that participants regularly moved their intervention stoves, often finding shaded areas to cook, contributing to the variability. Participants were asked to move the U-pods with the stoves if they moved them, but we had no way of ensuring compliance. Although we found significant reductions in OC and EC in homes given intervention stoves, we have also collected in-field stove emissions measurements to inform these differences (Coffey et al., *in preparation*).

The control group results ($6.5 \mu g/m^3$ (2.3, 19.0)) are lower than the observations of Van Vliet et al. (2013), who reported cooking area BC concentrations of $14.5 \mu g/m^3$ (12.0, 16.9) based on 24-h samples at 29 rural homes in Central Ghana. Total $PM_{2.5}$ was measured in kitchens in Accra as part of a before/after study looking at the effectiveness of a Gyapa wood cookstove with a design similar to the one used in this study (Pennise et al., 2009). Average concentrations, measured there with the UCB-PATS (Edwards et al., 2006) were $650 \mu g/m^3$ before the introduction of the Gyapa, and $320 \mu g/m^3$ after introduction, but the kitchens and use patterns are likely to have very different characteristics in Accra than in the North where our study was conducted. Kitchen geometry and behavior are very important for determining in-home PM concentrations and estimating personal exposure from cooking area measurements, as more open geometries will increase spatial pollution heterogeneity. Also in Accra, Zhou et al. (2013; 2014) performed $PM_{2.5}$ and PM_{10} sampling in four neighborhoods

throughout the city simultaneously outdoors, and inside households' kitchens. Cooking area PM_{2.5} differed substantially by neighborhood, ranging from 58-74 µg/m³ for the 'poorer' neighborhoods, to 25-33 µg/m³ for the 'affluent' neighborhoods, similar to observations presented here.

3.3. Personal vs. microenvironmental carbonaceous PM_{2.5} modeling results

To reduce the complexity of cookstove exposure studies, it is often a hope that cooking-area microenvironment concentrations can be used as a proxy for personal exposures. However, we did not observe strong relationships between personal PM_{2.5} and cooking area microenvironment PM_{2.5} (Equation 3). Microenvironment OC was a significant positive predictor for personal OC ($p < 0.01$), but the complete model explained only 14.6% of the observed variance. Similar seasonal effects were seen as in the personal exposure results presented in Section 3.1.1, with lower expected exposures in the light and heavy rainy seasons ($p = 0.32$, $p = 0.03$) (SI section 6).

Microenvironment EC was not a significant predictor for personal EC ($p = 0.08$), though the EC interaction term with the Gyapa/Gyapa stove group was significantly lower than the control group ($p = 0.05$). Once again, the light and heavy rainy seasons had significantly lower EC exposures ($p < 0.01$, $p = 0.02$). However, the model only explained 7.6% of the observed variance (SI Section 7). Van Vliet et al. (2013) reported a similar lack of relationship between cooking area and personal BC in Central Ghana.

3.4. Ambient carbonaceous PM_{2.5}

Average ambient PM_{2.5} EC and OC concentrations were 0.3 µg/m³ and 4.1 µg/m³, respectively (Figure 4 and Table 4). These were lower than those measured in Navrongo by Ofosu et al. (2013), who reported 0.95 µg/m³, and 11.36 µg/m³. That study reported an average ratio of total particulate carbon to total PM_{2.5} mass of 38.0%. Using these results, the mean total mass concentrations in our work should be in the range of 11.6 µg/m³, substantially lower than that measured by the Ofosu study (mean total PM_{2.5} mass concentration of

32.4 $\mu\text{g}/\text{m}^3$ was reported). The difference could be due in part to the fact that the Ofosu study site was closer to a main road (not reported, but between 10-200m), than here (395m) (**Error! Reference source not found.**).

Our ambient measurements can also be compared with other regional measurements from Ouagadougou, Burkina Faso, and from Accra. In Ouagadougou, Boman et al. (2009) measured total $\text{PM}_{2.5}$ concentrations from 27-164 $\mu\text{g}/\text{m}^3$, and BC concentrations from 1.3-8.2 $\mu\text{g}/\text{m}^3$ during intensive sampling from 29 November to 11 December 2007, in the midst of the Harmattan season. In Accra, Zhou et al. (2013; 2014) measured ambient $\text{PM}_{2.5}$ concentrations ranging from 63-104 $\mu\text{g}/\text{m}^3$ for the 'poorer' neighborhoods, to 21-55 $\mu\text{g}/\text{m}^3$ for the 'affluent' neighborhoods. They also found seasonal increases in regional and locally resuspended $\text{PM}_{2.5}$ and PM_{10} , which were attributed to the Harmattan winds. Van Vliet et al. (2013) reported average ambient $\text{PM}_{2.5}$ BC concentrations of 2.0 $\mu\text{g}/\text{m}^3$ (1.1, 2.9) ($n = 9$), and $\text{PM}_{2.5}$ mass concentrations of 20 $\mu\text{g}/\text{m}^3$ (12.1, 27.9).

Figure 4. Time series of weekly EC and OC $\text{PM}_{2.5}$ samples collected in Navrongo from September 2013-November 2014, presented with pointwise uncertainty estimates.

Table 4. Summary of ambient carbonaceous $\text{PM}_{2.5}$ EC, OC, and organics samples. Flagged samples were removed from analysis due to various possible issues including sampling duration and contamination.

	Mean	Median	Stdev	Geometric Mean	%BDL	
OC ($\mu\text{g}/\text{m}^3$)	4.1	3.2	4.3	2.8	2.0	
EC ($\mu\text{g}/\text{m}^3$)	0.3	0.1	0.3	0.1	47.4	
EC/OC %	6.5%					
PMF Ambient PM_{2.5} Dataset (n = 25)						
<i>*units in ng/m³ unless noted</i>	Mean	Median	Stdev	Geometric Mean	%BDL	S/N
OC ($\mu\text{g}/\text{m}^3$)	3.65	3.80	1.67	3.13	0.0	6.7
EC ($\mu\text{g}/\text{m}^3$)	0.39	0.25	0.33	0.23	0.0	3.0
docosane (C22)	0.34	0.36	0.26	0.26	0.0	5.7
tricosane (C23)	0.53	0.41	0.39	0.40	0.0	5.3
tetracosane (C24)	0.66	0.48	0.67	0.44	0.0	4.5
pentacosane (C25)	1.10	0.77	0.86	0.86	0.0	5.4
heptacosane (C27)	2.07	1.68	1.84	1.66	0.0	6.8
nonacosane (C29)	3.96	2.20	7.02	2.46	0.0	7.7
hentriacontane (C31)	3.16	2.28	3.49	2.27	0.0	8.0
fluoranthene	0.05	0.04	0.04	0.03	0.0	5.2
pyrene	0.03	0.02	0.02	0.02	0.0	5.2
benzo[b&k]fluoranthene	0.18	0.19	0.10	0.15	0.1	7.9
benz[a&e]pyrene	0.14	0.15	0.07	0.12	0.1	7.4
indeno[1,2,3-cd]pyrene	0.06	0.06	0.03	0.05	0.0	5.7
benzo[ghi]perylene	0.09	0.08	0.05	0.07	0.0	5.5
ba-30-norhopane	0.15	0.14	0.07	0.13	0.3	6.0
ab-hopane	0.12	0.11	0.07	0.10	0.3	5.2
coniferaldehyde	2.31	1.66	1.98	1.71	0.3	5.4
acetosyringone	0.34	0.29	0.32	0.23	0.1	3.7

3.5. Personal PM_{2.5} source apportionment results

A 6-factor PMF solution (Figure 5) was identified for the P-M data set. The temporal patterns of factors were of little help in identifying sources in the personal and microenvironmental data set due to the short and overlapping time periods over which samples were collected (SI Figure 3). Organics source fingerprints typically used for source identification and validation have not been generated for Africa to our knowledge, limiting our ability to interpret some of the solutions. We thus rely on

previously collected source fingerprints from around the world, primarily the United States. Future work will aim to fill these gaps.

Figure 5. 6-factor PMF solution factor profiles for the personal and microenvironmental data, reported as percent of species, along with bootstrapped standard deviations. Mean contributions to OC and EC are shown for each factor in units of ng/m³, and by the percentage of total apportioned.

The ‘Light n-alkane’ factor is characterized by a high proportion of the low molecular weight n-alkanes, and hopanes. This factor contributes 20.5% and 11.7% (Table 1, SI) of the median apportioned OC and EC, respectively, where medians are used due to skewed distributions. This pattern is indicative of combustion of fossil fuels such as diesel (Rogge et al., 1993a). The ‘light SVOC’ factor has a high proportion of light n-alkanes and semi-volatile PAHs, including flouranthene and pyrene. This factor could be attributed to early stage (smoldering) biomass combustion when light volatiles first escape, or to charcoal burning, which has been shown to have high flouranthene and

pyrene concentrations (Hou et al., 2009). The 'light SVOC' factor has a median of 7.9% of OC and 0.0% of EC apportioned to it. The 'odd n-alkane' factor shows the well-known pattern of odd n-alkanes enrichment characteristic of biogenic detritus like plant waxes (Rogge et al., 1993c), and has a median of 38.4% of the OC and 59.2% of EC apportioned to it. A source with a biogenic component would be expected, as the study participants all live on small rural agriculture farms, but the high EC and OC apportionment point to contributions from additional sources. These samples were primarily collected during the windy Harmattan season, so increased biogenic material due to resuspension would be expected, but this also occurs contemporaneously with seasonal crop burning. Bin Abas et al. (1995) showed that smoke samples of forest litter from the Amazon show a clear odd n-alkane preference, suggesting that local and regional biomass residue and crop burning may contribute to this source. The 'hopane and n-alkane' factor contributes the largest fraction of the heavier alkanes. This profile is indicative of gasoline engine combustion, road dust, motor oil, and tire wear (Rogge et al., 1993a and b; Schauer et al., 1999) and has 24.0% of OC, and 13.7% of EC apportioned to it. This factor could also be related to trash burning, as various plastic types have been shown to have similar fingerprints to motor vehicle combustion (Mohr et al., 2009), though the data on organic PM from combustion of such materials is limited (Estrellan and Lino, 2010). The 'PAH' factor has a median of 3.2% of the apportioned OC and 14.2% of the apportioned EC, suggesting that this may be from combustion sources such as the flaming phase of biomass combustion, or from non-catalyst equipped gasoline vehicles. The 'methoxyphenol' factor has the greatest proportion of the syringone, acetosyringone, and coniferaldehyde, which are all biomass combustion markers (Schauer et al., 2001), along with small amounts of the light alkanes and hopanes. This factor contributes a median of 6.0% of the OC and 1.1% of the EC.

3.5.1. Factor enrichment in personal vs. cooking area samples

Comparing the factor contributions of the personal and cooking area microenvironment samples (Figure 6), we find enrichment of some factors, providing

additional evidence on the sources of PMF factor profiles. The data were aggregated into the personal and cooking area samples because there were not enough samples ($n=10$) to directly model the individual vs. cooking area PMF factor contributions. The 'methoxyphenol' factor, associated with wood smoke, had a median contribution in the cooking area microenvironment 90% higher than in the personal measurements, and comes from a different distribution according to the Kruskal-Wallis test, at $p = 0.06$. We would expect enrichment in this factor since the assumed main source of $PM_{2.5}$ in the cooking area is generally from biofuel combustion. Similarly, the 'PAH' factor was enriched in the cooking area samples, with a median contribution 249% higher than the personal samples ($p = 0.12$), consistent with flaming household combustion emissions. The lack of cooking area enrichment for the 'light SVOC' factor suggests that this factor may have other sources apart from smoldering biomass combustion. We would also expect personal samples to be enriched in motor vehicle-related sources, and we find that the 'hopane and n-alkane' factor is 61% higher in the personal samples, though not significant. There was no significant enrichment of the 'odd n-alkane' factor, suggesting that this biomass detritus and vegetation combustion-related source is more regional in nature. Overall, the two factors most strongly linked to home biomass combustion, the 'methoxyphenols' and 'PAHs', contribute a median of 2.5% of the OC, 11.5% of the EC, and 3.0% of the carbonaceous $PM_{2.5}$ to the personal samples, and 8.0% of the OC, 30.5% of the EC, and 9.5% of the carbonaceous $PM_{2.5}$ for the cooking area samples (SI Table 1). The largest contributors to total carbonaceous $PM_{2.5}$ in the personal samples are the 'odd n-alkane' source (41.1%), associated with resuspended biomass detritus and local and regional biomass burning, along with the 'light n-alkane' (15.9%) and 'hopane and n-alkane' (26.4%), which are both related to vehicular combustion emissions.

Figure 6 Comparisons of factor contributions to personal vs. cooking area microenvironment samples. P-values shown are from the Kruskal-Wallis non-parametric ANOVA test to compare distributions of two or more groups.

3.5.2. Personal exposure mixed effects modeling using PMF factors

The two identified PMF factors related to home biomass combustion, the 'PAH' and 'methoxyphenol' factors, enable an analysis of the differences in exposure to those factors based on stove intervention group. A mixed effects model (Eq. 2) is again used, with fewer covariates than that specified in Equation 1 due to the smaller subset of samples available with speciated organics data (Table 2).

$$\ln(Y_{ij}) \sim \beta_0 + \beta_1(\text{stovegroup})_i + \beta_2(\text{family})_i + \beta_3(\text{gender*age})_i + \beta_4(\text{primary cook})_i + \alpha_j + e_{ij} \quad (4)$$

First, Equation 4 was applied with OC as the dependent variable, and we found consistent results with the larger data set: participants in the intervention groups have lower OC PM_{2.5} exposures than the control groups ($p < 0.05$ for the groups with Philips stoves, $p = 0.12$ for the Gyapa/Gyapa group), and there are no effects by number of family members, age category, or primary cook status (SI Section 8). This process was repeated with EC as the dependent variable, and we found that the results were not in exact agreement with the main data set results. While in the main model (Section 3.1) all intervention groups had lower EC exposure, here only the Philips/Philips and

Philips/Gyapa group had lower EC exposures relative to the control group ($p = 0.10$ and $p = 0.19$, respectively), and there were no significant differences by the other covariates.

Equation 4 was then applied to the source contributions from PMF factors as the dependent variables, in effect isolating the source-specific portions of the measured $PM_{2.5}$ (Table 3). Personal exposure to the 'methoxyphenols' factor was 79.2% lower for the Philips/Philips group ($p = 0.11$), 78.3% lower for the Gyapa/Philips ($p = 0.26$), and 39.6% lower for the Gyapa/Gyapa group ($p = 0.70$). Exposure for the primary cook was 364% higher ($p = 0.16$) for this factor than non-cooks, and there were no significant differences by age-by-gender, or number of family members. The 'PAH' factor had 98.2% lower exposure for the Philips/Philips group ($p < 0.01$), and no other significant differences or large effects. This is a reasonable result given that our 'PAH' source appears to be associated with flaming combustion. This approach would have benefitted from a larger sample size.

3.5.3. Ambient PM_{2.5} source apportionment

Six PMF factors were identified for the ambient PM_{2.5} data set (Table 4). These ambient factor profiles (Figure 7) were similar to those of the P-M profiles (

Figure 5), with all Spearman's correlation coefficients significant (0.53 to 0.88, $p < 0.05$), except 0.35 for the 'PAH' factor, which may reflect a merging with the biogenic factor seen in the P-M profiles. The 'hopane and n-alkane' factor from the P-M profiles were separated into a 'hopane' factor in the ambient analysis, with more of the alkanes instead apportioned to the 'odd n-alkane' factor here.

To better understand source origins of these ambient factors, the PMF factor contributions over time were plotted against ambient temperature and wind speed data. Meteorological data were collected from a Weather Underground station (Station 65401). Data from this site are reported sparsely, with 1-4 measurements made per day. Only maximum temperature and wind speeds observed during the sample collection periods were used for analysis (results were consistent with other extracted features

like the median and mean). Comparisons of factor profiles with temperature were made with the Spearman correlation coefficient due to non-normality. Since PM relationships with wind speed are often more complex due to dilution and resuspension, trends were analyzed after spline smoothing the data (SI Figure 2).

The 'odd n-alkane' factor shows an increase during the Harmattan season from November to mid-January, consistent with long-range transport and generation and resuspension of biogenic detritus. The smoothed relationship with wind shows decreasing PM concentrations associated with wind speeds up to 10m/s, and then increasing concentrations above 10m/s, though the data are sparse at higher wind speeds. This 'U' shape has previously been identified in various works, and was attributed to dilution of PM_{10-2.5} at lower wind speeds and resuspension at higher speeds (Harrison et al., 2001; Clements et al., 2012).

The 'PAH' factor peaks between November and mid-January, consistent with seasonal crop burning. To further confirm the source origin, we compared PAH factor contributions with ambient carbon monoxide (CO) concentrations and emissions. Ambient CO concentrations were measured continuously at the same site as the ambient PM_{2.5} filter sampler (Dickinson et al., 2015) (Figure 8) and were averaged over time to match the PM_{2.5} samples. We also considered CO emissions (kg/day) from open biomass burning, calculated by the Fire INventory from NCAR (Wiedinmyer et al., 2011). The 'PAH' factor contributions has a Spearman correlation of 0.76 with the locally measured average CO, and a correlation of 0.72 with the FINN emission estimates, providing compelling evidence that this source is associated with regional and seasonal biomass burning. This factor is the biggest contributor of median OC, EC, and total carbonaceous mass (54.5%, 80.9%, and 57.0%, respectively (Figure 7)). Additionally, a distinct 'U' shaped relationship with wind speed suggests that this factor also has local sources, like residential biomass waste burning, while the increase with wind speed is likely due to correlation with seasonal crop burning, and the seasonal wind pattern.

The 'light SVOC' factor is more pronounced in the hot-dry season from mid-January to April, and accounts for 7.4% of the apportioned EC. This factor could be

associated with vehicle combustion and evaporative fuel emissions (Schauer et al., 1999). There is a diesel storage tank on the NHRC grounds, and as with most others in the region, it is poorly sealed, potentially contributing evaporative emissions. This factor had a modest increase in contributions with higher wind speeds, suggesting that it may be due to regional transport of combustion emissions as well, especially since evaporative emissions would not account for the EC present in this factor. Although there were no statistically significant correlations with temperature and any of the identified factors, the largest correlation was for this factor, which had a correlation coefficient of 0.29, lending some support to the possibility that evaporative fuel emissions contribute to this source.

The 'light n-alkane' factor and the 'hopane and n-alkane' factor both display a lack of seasonality, consistent with their expected motor vehicle or fossil fuel combustion sources. The 'light n-alkane' factor exhibited a dilution effect as wind increased above 7m/s, though a few data points could be driving that trend. The 'hopane and n-alkane' factor showed a slight increase with wind speed.

The 'methoxyphenol' factor has several peaks in its factor contributions (Figure 8), including the most pronounced in November. Wood biomass combustion is known to occur throughout Navrongo all year round, so we do not expect distinct seasonality here. This factor showed correlations of 0.53 and 0.56 with ambient CO concentrations and open biomass burning CO emissions. It also decreased with increasing wind speed, indicative of dilution. As such, this factor is likely from local wood burning, which we assume is primarily from residential and commercial activities. This factor contributes a median of 15.3% of apportioned OC and 0.2% of EC. The two biomass combustion-related factors, the 'methoxyphenol' factor and the 'PAH' factor, together are responsible for a median of 67.8% of the OC and 81.1% of the EC. These are substantially higher contributions than for the P-M results, for which those sources contributed 15.3% and 9.2% of the EC and OC, likely because in the ambient results the 'PAH' factor has also been associated with regional crop burning.

Figure 7. Ambient PMF 6-factor solution profiles for the weekly samples collected at the NHRC.

Figure 8. Ambient PMF factor contributions for weekly-integrated samples collected at the Navrongo Health Research Center. Red bands shown are standard deviations from the bootstrapped solution. Factor correlations with locally measured CO averages and satellite-measured CO emissions are shown, as well as the respective CO time series.

4. Comparison with past works and regional implications

The identified ambient source profiles can be somewhat reconciled with the pollution sources in Navrongo described by Ofori et al. (2012). For example, two-stroke engine combustion emissions in that work were associated with observed OC and the motor oil additives Ca and Zn. This factor contributed an average of 9.9% of the observed $PM_{2.5}$ mass. This source could be related with our ‘hopane and n-alkane’ factor that includes high levels of ab-hopane and ba-30-norhopane, both of which are associated with lubricating oil combustion, and contributes a median of 11.8% of the carbonaceous $PM_{2.5}$. Their gasoline combustion source, identified by OC, K, Ca, and Fe (contributing a mean of 10.9% of mass), is somewhat similar to the ‘light n-alkane’ factor here (contributing a median of 11.3% of carbonaceous $PM_{2.5}$), which also has some of the hopanes and OC. As with all of the vehicle combustion-related factors in

that work, they did not observe significant weekend/weekday differences, consistent with known behavioral trends in the region. Their soil factor, containing crustal elements like Na and Mg, and contributing a mean of 35.9% of mass, can be most closely connected with our 'odd n-alkane' factor, which shares a Harmattan increase, and appears to be of biogenic origin, but contributes a median of only 4.4% of carbonaceous mass. The discrepancy in contribution may be due to how the PMF solution split our co-temporal 'PAH' and 'odd n-alkane' sources, as can be noted by the wide confidence bounds on their source contributions (Figure 8). Our 'PAH' factor may thus be associated with local and regional sources of combustion like wood, bush, and crop burning, as well as regional biogenic sources, whereas the longer time series in Ofosu et al. would aid in separating those sources. Similarly, the diesel combustion profile reported by Ofosu et al. that includes a large fraction of their observed EC and OC (contributing a mean of 11.5% of mass), could also be associated with our 'PAH' factor (contributing a median of 57.0% of carbonaceous mass). Ofosu et al. describe a biomass-burning factor, identified by OC, EC and K, which they associate with bush burning, and contributes a mean of 15.8% of mass. This can be matched with our 'methoxyphenol' factor, and again, the 'PAH' factor. Finally, their road dust factor, which looks similar to their soil factor but with more OC (contributing a mean of 16.0% of mass), may be most closely matched with our 'light SVOC' factor (contributing a median of 1.6% of carbonaceous mass) and the other vehicle-related factors. We find that it is difficult to separate road dust with vehicle emissions due to co-emission. We hypothesize that the 'light SVOC' factor here is associated with smoldering biomass combustion and evaporative fuel emissions.

The overall results of these two source apportionment studies are consistent and highlight the relatively balanced contribution of biomass combustion, biogenic, and vehicular sources to ambient PM_{2.5} in northern Ghana in terms of both organics and metals in PM_{2.5}. The significant Harmattan-influenced dust source is clearly difficult to control regionally. A portion of the 'PAH' source, however, could be lowered by reducing flaming biomass burning in homes and regionally, with a combination of

improved household combustion practices, and changes in farming and land use behaviors. Other sources also have potential for reduction, such as through vehicle emissions controls. Waste is currently disposed of via open burning, and although we did not directly identify a waste-burning specific source, this local and regional source could contribute to several of the PMF factors we observed. Improvements to waste management would be expected to yield air quality benefits.

5. Conclusions

Personal, cooking area, and ambient carbonaceous PM_{2.5} were quantified as part of the REACCTING cookstove intervention study in Navrongo, Northern Ghana. We used these measurements to determine personal exposures, and to determine the differences in exposures for different groups of a cookstove intervention study. Further, we used analysis of the PM_{2.5} organic species to identify sources of PM at different scales, and to determine the impact of local biomass burning on particulate exposure. The results from this study can be used to inform air quality studies and provide robust information towards the impact of improved stoves in Northern Ghana.

The cookstove intervention produced reductions in OC exposure in all of the groups given improved stoves, compared to the control group, despite continued use of traditional cooking methods across all study groups (Piedrahita et al., 2016). We also found significant reductions in EC exposure in both of the groups receiving a Philips stove, which is expected to have benefits for both health and climate.

The average concentrations of carbonaceous PM_{2.5} in the personal samples (42.5µg/m³) were much higher than the concentrations of the ambient samples (4.4µg/m³). Source apportionment performed on both personal exposure and regional ambient data showed strong similarities in the factor profiles, suggesting that the processes that drive regional air quality are the same as those that drive exposures. In both the exposure/cooking area and ambient samples, we identified two biomass combustion-related sources, likely indicative of different phases of combustion, or stove types. These factors contributed a median of 9.2% of OC and 15.3% of EC to personal

and cooking area samples. This result suggests that although cooking is an important source of PM_{2.5}, personal PM_{2.5} exposures in this region are also heavily impacted by other sources.

Finally, we directly modeled the effect of stove group on cooking-specific exposures using the PMF factors. We found lower but not significant differences in ‘methoxyphenol’ factor exposure in the intervention groups relative to controls, and significantly lower ‘PAH’ factor exposure in the Philips/Philips group relative to controls. This type of approach informs the relative importance of PM sources and would be a valuable addition to studies around the world, given the regional variability cooking behaviors, cookstoves, fuels, and prevalence of non-cooking PM sources.

Ethical Considerations

This work was reviewed and approved by the Institutional Review Boards at the University of Colorado Boulder, the National Center for Atmospheric Research, and the Navrongo Health Research Center, a part of the Ghana Health Service.

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References

- Aboh, I. J. K., Henriksson, D., Laursen, J., Lundin, M., Ofosu, F. G., Pind, N., ... Wahnström, T. (2009). Identification of aerosol particle sources in semi-rural area of Kwabenya, near Accra, Ghana, by EDXRF techniques. *X-Ray Spectrometry*, 38(4), 348–353. <http://doi.org/10.1002/xrs.1172>
- Arku, R. E., Dionisio, K. L., Hughes, A. F., Vallarino, J., Spengler, J. D., Castro, M. C., ... Ezzati, M. (2015). Personal particulate matter exposures and locations of students in four neighborhoods in Accra, Ghana. *Journal of Exposure Science and Environmental Epidemiology*, 25(6), 557–566. <http://doi.org/10.1038/jes.2014.56>
- Awini, E., Mattah, P., Sankoh, O., & Gyapong, M. (2010). Spatial variations in childhood mortalities at the Dodowa Health and Demographic Surveillance System site of the INDEPTH Network in Ghana. *Tropical Medicine & International Health*, 15(5), 520–528. <http://doi.org/10.1111/j.1365-3156.2010.02492.x>
- Baumgartner, J., Schauer, J. J., Ezzati, M., Lu, L., Cheng, C., Patz, J., & Bautista, L. E. (2011). Patterns and predictors of personal exposure to indoor air pollution from biomass combustion among women and children in rural China. *Indoor Air*, 21(6), 479–488. <http://doi.org/10.1111/j.1600-0668.2011.00730.x>
- bin Abas, M.R., Simoneit, B.R.T., Elias, V., Cabral, J.A., Cardoso, J.N., 1995. Composition of higher molecular weight organic matter in smoke aerosol from biomass combustion in Amazonia. *Chemosphere* 30, 995–1015. doi:10.1016/0045-6535(94)00442-W
- Boman, J., Lindén, J., Thorsson, S., Holmer, B., & Eliasson, I. (2009). A tentative study of urban and suburban fine particles (PM_{2.5}) collected in Ouagadougou, Burkina Faso. *X-Ray Spectrometry*, 38(4), 354–362. <http://doi.org/10.1002/xrs.1173>
- Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., DeAngelo, B.J., Flanner, M.G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim, M.C., Schultz, M.G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P.K., Jacobson, M.Z., Kaiser, J.W., Klimont, Z., Lohmann, U., Schwarz, J.P., Shindell, D., Storelvmo, T., Warren, S.G., Zender, C.S., 2013. Bounding the role of black carbon in the climate system: A scientific assessment: BLACK CARBON IN THE CLIMATE SYSTEM. *Journal of Geophysical Research: Atmospheres* 118, 5380–5552. doi:10.1002/jgrd.50171
- Bonjour, S., Adair-Rohani, H., Wolf, J., Bruce, N. G., Mehta, S., Prüss-Ustün, A., ... Smith, K. R. (2013). Solid Fuel Use for Household Cooking: Country and Regional Estimates for 1980–2010. *Environmental Health Perspectives*, 121(7), 784–790. <http://doi.org/10.1289/ehp.1205987>
- Bortey-Sam, N., Ikenaka, Y., Akoto, O., Nakayama, S. M. M., Yohannes, Y. B., Baidoo, E., ... Ishizuka, M. (2015). Levels, potential sources and human health risk of polycyclic aromatic hydrocarbons (PAHs) in particulate matter (PM₁₀) in Kumasi, Ghana. *Environmental Science and Pollution Research*, 22(13), 9658–9667. <http://doi.org/10.1007/s11356-014-4022-1>

- Burnett, R. T., Pope, C. A., III, Ezzati, M., Olives, C., Lim, S. S., Mehta, S., ... Cohen, A. (2014). An Integrated Risk Function for Estimating the Global Burden of Disease Attributable to Ambient Fine Particulate Matter Exposure. *Environmental Health Perspectives*. <http://doi.org/10.1289/ehp.1307049>
- Cass, G. R. (1998). Organic molecular tracers for particulate air pollution sources. *TrAC Trends in Analytical Chemistry*, 17(6), 356–366. [http://doi.org/10.1016/S0165-9936\(98\)00040-5](http://doi.org/10.1016/S0165-9936(98)00040-5)
- Clark, M. L., Peel, J. L., Balakrishnan, K., Breysse, P. N., Chillrud, S. N., Naeher, L. P., ... Balbus, J. M. (2013). Health and Household Air Pollution from Solid Fuel Use: The Need for Improved Exposure Assessment. *Environmental Health Perspectives*. <http://doi.org/10.1289/ehp.1206429>
- Clements, N., Piedrahita, R., Ortega, J., Peel, J. L., Hannigan, M., Miller, S. L., & Milford, J. B. (2012). Characterization and Nonparametric Regression of Rural and Urban Coarse Particulate Matter Mass Concentrations in Northeastern Colorado. *Aerosol Science and Technology*, 46(1), 108–123. <http://doi.org/10.1080/02786826.2011.607478>
- Cynthia, A. A., Edwards, R. D., Johnson, M., Zuk, M., Rojas, L., Jiménez, R. D., ... Masera, O. (2008). Reduction in personal exposures to particulate matter and carbon monoxide as a result of the installation of a Patsari improved cook stove in Michoacan Mexico. *Indoor Air*, 18(2), 93–105. <http://doi.org/10.1111/j.1600-0668.2007.00509.x>
- Dickinson, K. L., Kanyomse, E., Piedrahita, R., Coffey, E., Rivera, I. J., Adoctor, J., ... Wiedinmyer, C. (2015). Research on emissions, air quality, climate, and cooking technologies in Northern Ghana (Reaccting): study rationale and protocol. *BMC Public Health*, 15(1), 126. <http://doi.org/10.1186/s12889-015-1414-1>
- Dionisio, K. L., Howie, S. R. C., Dominici, F., Fornace, K. M., Spengler, J. D., Adegbola, R. A., & Ezzati, M. (2012). Household Concentrations and Exposure of Children to Particulate Matter from Biomass Fuels in The Gambia. *Environmental Science & Technology*, 46(6), 3519–3527. <http://doi.org/10.1021/es203047e>
- Dutton, S.J., Schauer, J.J., Vedal, S., Hannigan, M.P., 2009a. PM2.5 characterization for time series studies: Pointwise uncertainty estimation and bulk speciation methods applied in Denver. *Atmospheric Environment* 43, 1136-1146.
- Dutton, S.J., Williams, D.E., Garcia, J.K., Vedal, S., Hannigan, M.P., 2009b. PM2.5 characterization for time series studies: Organic molecular marker speciation methods and observations from daily measurements in Denver. *Atmospheric Environment* 43, 2018-2030.
- Edwards, R., Smith, K. R., Kirby, B., Allen, T., Litton, C. D., & Hering, S. (2006). An inexpensive dual-chamber particle monitor: laboratory characterization. *Journal of the Air & Waste Management Association*, 56(6), 789–799.
- Estrellan, C. R., & Iino, F. (2010). Toxic emissions from open burning. *Chemosphere*, 80(3), 193–207. <http://doi.org/10.1016/j.chemosphere.2010.03.057>

- Harrison, R. M., Yin, J., Mark, D., Stedman, J., Appleby, R. S., Booker, J., & Moorcroft, S. (2001). Studies of the coarse particle (2.5–10 μm) component in UK urban atmospheres. *Atmospheric Environment*, 35(21), 3667–3679. [http://doi.org/10.1016/S1352-2310\(00\)00526-4](http://doi.org/10.1016/S1352-2310(00)00526-4)
- Hemann, J. G. (2009). Assessing PMF fit. Retrieved from <http://www.atmos-chem-phys.net/9/497/2009/acp-9-497-2009.pdf>
- Hou, X., Zhuang, G., Lin, Y., Li, J., Jiang, Y., & Fu, J. S. (2009). Emission of fine organic aerosol from traditional charcoal broiling in China. *Journal of Atmospheric Chemistry*, 61(2), 119–131. <http://doi.org/10.1007/s10874-009-9128-3>
- Janssen, N.A.H., Hoek, G., Simic-Lawson, M., Fischer, P., van Bree, L., ten Brink, H., Keuken, M., Atkinson, R.W., Anderson, H.R., Brunekreef, B., Cassee, F.R., 2011. Black Carbon as an Additional Indicator of the Adverse Health Effects of Airborne Particles Compared with PM_{10} and $\text{PM}_{2.5}$. *Environmental Health Perspectives* 119, 1691–9.
- Johnson, M., Edwards, R., Alatorre Frenk, C., Masera, O., 2008. In-field greenhouse gas emissions from cookstoves in rural Mexican households. *Atmospheric Environment* 42, 1206–1222. doi:10.1016/j.atmosenv.2007.10.034
- Johnson, M. A., & Chiang, R. A. (2015). Quantitative Stove Use and Ventilation Guidance for Behavior Change Strategies. *Journal of Health Communication*, 20(sup1), 6–9. <http://doi.org/10.1080/10810730.2014.994246>
- Kar, A., Rehman, I. H., Burney, J., Puppala, S. P., Suresh, R., Singh, L., ... Ramanathan, V. (2012). Real-Time Assessment of Black Carbon Pollution in Indian Households Due to Traditional and Improved Biomass Cookstoves. *Environmental Science & Technology*, 46(5), 2993–3000. <http://doi.org/10.1021/es203388g>
- Kleeman, M.J., Schauer, J.J., Cass, G.R., 1999. Size and Composition Distribution of Fine Particulate Matter Emitted from Wood Burning, Meat Charbroiling, and Cigarettes. *Environ. Sci. Technol.* 33, 3516–3523. doi:10.1021/es981277q
- Larson, T., Gould, T., Simpson, C., Liu, L.-J.S., Claiborn, C., Lewtas, J., 2004. Source Apportionment of Indoor, Outdoor, and Personal $\text{PM}_{2.5}$ in Seattle, Washington, Using Positive Matrix Factorization. *Journal of the Air & Waste Management Association* 54, 1175–1187. doi:10.1080/10473289.2004.10470976
- L'Orange, C., DeFoort, M., & Willson, B. (2012). Influence of testing parameters on biomass stove performance and development of an improved testing protocol. *Energy for Sustainable Development*, 16(1), 3–12. <http://doi.org/10.1016/j.esd.2011.10.008>
- Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., ... Ezzati, M. (2012). A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk

- factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. *The Lancet*, 380(9859), 2224–2260. [http://doi.org/10.1016/S0140-6736\(12\)61766-8](http://doi.org/10.1016/S0140-6736(12)61766-8)
- McCracken, J. P., Schwartz, J., Diaz, A., Bruce, N., & Smith, K. R. (2013). Longitudinal Relationship between Personal CO and Personal PM_{2.5} among Women Cooking with Woodfired Cookstoves in Guatemala. *PLoS ONE*, 8(2), e55670. <http://doi.org/10.1371/journal.pone.0055670>
- McCracken, J. P., Smith, K. R., Díaz, A., Mittleman, M. A., & Schwartz, J. (2007). Chimney Stove Intervention to Reduce Long-term Wood Smoke Exposure Lowers Blood Pressure among Guatemalan Women. *Environmental Health Perspectives*, 115(7), 996–1001. <http://doi.org/10.1289/ehp.9888>
- Mohr, C., Huffman, J. A., Cubison, M. J., Aiken, A. C., Docherty, K. S., Kimmel, J. R., ... Jimenez, J. L. (2009). Characterization of Primary Organic Aerosol Emissions from Meat Cooking, Trash Burning, and Motor Vehicles with High-Resolution Aerosol Mass Spectrometry and Comparison with Ambient and Chamber Observations. *Environmental Science & Technology*, 43(7), 2443–2449. <http://doi.org/10.1021/es8011518>
- Naeher, L.P., Brauer, M., Lipsett, M., Zelikoff, J.T., Simpson, C.D., Koenig, J.Q., Smith, K.R., 2007. Woodsmoke Health Effects: A Review. *Inhalation Toxicology* 19, 67–106. doi:10.1080/08958370600985875
- Oduro, A. R., Wak, G., Azongo, D., Debpuur, C., Wontuo, P., Kondayire, F., ... Binka, F. (2012). Profile of the Navrongo Health and Demographic Surveillance System. *International Journal of Epidemiology*, 41(4), 968–976. <http://doi.org/10.1093/ije/dys111>
- Ofosu, F. G., Hopke, P. K., Aboh, I. J. K., & Bamford, S. A. (2012). Characterization of fine particulate sources at Ashaiman in Greater Accra, Ghana. *Atmospheric Pollution Research*, 3(3), 301–310. <http://doi.org/10.5094/APR.2012.033>
- Ofosu, F. G., Hopke, P. K., Aboh, I. J. K., & Bamford, S. A. (2013). Biomass burning contribution to ambient air particulate levels at Navrongo in the Savannah zone of Ghana. *Journal of the Air & Waste Management Association*, 63(9), 1036–1045. <http://doi.org/10.1080/10962247.2013.783888>
- Paatero, P. (1997). Least squares formulation of robust non-negative factor analysis. *Chemometrics and Intelligent Laboratory Systems*, 37(1), 23–35. [http://doi.org/10.1016/S0169-7439\(96\)00044-5](http://doi.org/10.1016/S0169-7439(96)00044-5)
- Pennise, D., Brant, S., Agbeve, S. M., Quaye, W., Mengesha, F., Tadele, W., & Wofchuck, T. (2009). Indoor air quality impacts of an improved wood stove in Ghana and an ethanol stove in Ethiopia. *Energy for Sustainable Development*, 13(2), 71–76. <http://doi.org/10.1016/j.esd.2009.04.003>
- Peretz, C., Goren, A., Smid, T., & Kromhout, H. (2002). Application of Mixed-effects Models for Exposure Assessment. *Annals of Occupational Hygiene*, 46(1), 69–77. <http://doi.org/10.1093/annhyg/mef009>
- Piedrahita, R., Dickinson, K.L., Kanyomse, E., Coffey, E., Alirigia, R., Hagar, Y., Rivera, I., Oduro, A., Dukic, V., Wiedinmyer, C., Hannigan, M., 2016. Assessment of cookstove stacking in Northern Ghana using

- surveys and stove use monitors. *Energy for Sustainable Development* 34, 67–76.
doi:10.1016/j.esd.2016.07.007
- Polissar, A.V., Hopke, P.K., Paatero, P., 1998. Atmospheric aerosol over Alaska - 2. Elemental composition and sources. *Journal of Geophysical Research-Atmospheres* 103, 19045-19057.
- Ramanathan, V., Carmichael, G., 2008. Global and regional climate changes due to black carbon. *Nature Geosci* 1, 221–227. doi:10.1038/ngeo156
- Reff, A., Eberly, S. I., & Bhawe, P. V. (2007). Receptor Modeling of Ambient Particulate Matter Data Using Positive Matrix Factorization: Review of Existing Methods. *Journal of the Air & Waste Management Association*, 57(2), 146–154. <http://doi.org/10.1080/10473289.2007.10465319>
- Roden, C.A., Bond, T.C., Conway, S., Osorio Pinel, A.B., MacCarty, N., Still, D., 2009. Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmospheric Environment* 43, 1170–1181. doi:10.1016/j.atmosenv.2008.05.041
- Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., & Simoneit, B. R. T. (1993a). Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks. *Environ. Sci. Technol.*, 27(4), 636–651. <http://doi.org/10.1021/es00041a007>
- Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., & Simoneit, B. R. T. (1993b). Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: roads as sources and sinks. *Environ. Sci. Technol.*, 27(9), 1892–1904. <http://doi.org/10.1021/es00046a019>
- Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., & Simoneit, B. R. T. (1993c). Sources of fine organic aerosol. 4. Particulate abrasion products from leaf surfaces of urban plants. *Environ. Sci. Technol.*, 27(13), 2700–2711. <http://doi.org/10.1021/es00049a008>
- Schauer, J. J., Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., & Simoneit, B. R. T. (1996). Source apportionment of airborne particulate matter using organic compounds as tracers. *Atmospheric Environment*, 30(22), 3837–3855. [http://doi.org/10.1016/1352-2310\(96\)00085-4](http://doi.org/10.1016/1352-2310(96)00085-4)
- Schauer, J. J., Kleeman, M. J., Cass, G. R., & Simoneit, B. R. T. (1999). Measurement of Emissions from Air Pollution Sources. 2. C1 through C30 Organic Compounds from Medium Duty Diesel Trucks. *Environmental Science & Technology*, 33(10), 1578–1587. <http://doi.org/10.1021/es980081n>
- Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 2001. Measurement of Emissions from Air Pollution Sources. 3. C1–C29 Organic Compounds from Fireplace Combustion of Wood. *Environ. Sci. Technol.* 35, 1716–1728. doi:10.1021/es001331e
- Schauer, J.J., Mader, B.T., Deminter, J.T., Heidemann, G., Bae, M.S., Seinfeld, J.H., Flagan, R.C., Cary, R.A., Smith, D., Huebert, B.J., Bertram, T., Howell, S., Kline, J.T., Quinn, P., Bates, T., Turpin, B., Lim, H.J., Yu, J.Z., Yang, H., Keywood, M.D., 2003. ACE-Asia intercomparison of a thermal-optical method for the determination of particle-phase organic and elemental carbon. *Environmental Science & Technology* 37, 993-1001.

- Smith, K.R., Jerrett, M., Anderson, H.R., Burnett, R.T., Stone, V., Derwent, R., Atkinson, R.W., Cohen, A., Shonkoff, S.B., Krewski, D., Pope, C.A., Thun, M.J., Thurston, G., 2009. Public health benefits of strategies to reduce greenhouse-gas emissions: health implications of short-lived greenhouse pollutants. *Lancet* 374, 2091–2103. doi:10.1016/S0140-6736(09)61716-5
- US Department of Commerce, N. (1997). NIST Technical Note 1297. Retrieved February 9, 2016, from <http://www.nist.gov/pml/pubs/tn1297/index.cfm>
- Van Vliet, E.D.S., Asante, K., Jack, D.W., Kinney, P.L., Whyatt, R.M., Chillrud, S.N., Abokyi, L., Zandoh, C., Owusu-Agyei, S., 2013. Personal exposures to fine particulate matter and black carbon in households cooking with biomass fuels in rural Ghana. *Environmental Research* 127, 40–48. doi:10.1016/j.envres.2013.08.009
- Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., & Soja, A. J. (2011). The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. *Geosci. Model Dev.*, 4(3), 625–641. <http://doi.org/10.5194/gmd-4-625-2011>
- Xie, M., Piedrahita, R., Dutton, S.J., Milford, J.B., Hemann, J.G., Peel, J.L., Miller, S.L., Kim, S.-Y., Vedal, S., Sheppard, L., Hannigan, M.P., 2013. Positive matrix factorization of a 32-month series of daily PM_{2.5} speciation data with incorporation of temperature stratification. *Atmospheric Environment* 65, 11-20.
- Xie, M., Hannigan, M.P., Barsanti, K.C., 2014. Gas/particle partitioning of n-alkanes, PAHs and oxygenated PAHs in urban Denver. *Atmospheric Environment* 95, 355-362.
- Yan, B., Kennedy, D., Miller, R.L., Cowin, J.P., Jung, K., Perzanowski, M., Balletta, M., Perera, F.P., Kinney, P.L., Chillrud, S.N., 2011. Validating a nondestructive optical method for apportioning colored particulate matter into black carbon and additional components. *Atmospheric Environment* 45, 7478–7486. doi:10.1016/j.atmosenv.2011.01.044
- Zeger, S. L., Liang, K.-Y., & Albert, P. S. (1988). Models for Longitudinal Data: A Generalized Estimating Equation Approach. *Biometrics*, 44(4), 1049–1060. <http://doi.org/10.2307/2531734>
- Zhou, Z., Dionisio, K. L., Verissimo, T. G., Kerr, A. S., Coull, B., Arku, R. E., ... Ezzati, M. (2013). Chemical composition and sources of particle pollution in affluent and poor neighborhoods of Accra, Ghana. *Environmental Research Letters*, 8(4), 044025. <http://doi.org/10.1088/1748-9326/8/4/044025>
- Zhou, Z., Dionisio, K. L., Verissimo, T. G., Kerr, A. S., Coull, B., Howie, S., ... Ezzati, M. (2014). Chemical Characterization and Source Apportionment of Household Fine Particulate Matter in Rural, Peri-urban, and Urban West Africa. *Environmental Science & Technology*, 48(2), 1343–1351. <http://doi.org/10.1021/es404185m>