On the assessment of air pollution and behavior within a cookstove intervention study in Northern Ghana and development of improved measurement techniques

by

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ABSTRACT

Piedrahita, Ricardo Antonio (Ph.D., Mechanical Engineering)

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Thesis directed by Associate Professor Michael P. Hannigan

2.8 billion people burn solid fuels for cooking (Bonjour et al., 2013) and the resulting air pollution is the third leading risk factor for the global burden of disease, contributing to 4 million premature deaths per year (Lim et al., 2012). There are also impacts on global and regional climate systems, and ecological health. This dissertation investigates aspects of these issues as part of REACCTING (Research on Emissions, Air quality, Climate, and Cooking Technologies in Northern Ghana), a 200-home cookstove intervention study in the Kassena-Nankana (K-N) Districts of Northern Ghana that took place from 2013-2016. It contributes quantitative results from the REACCTING study, and novel exposure estimation techniques to improve intervention assessment, in five chapters. First, traditional and intervention stove use results are presented from both electronic stove usage monitors and surveys. Stove use patterns are explained, and improvements to stoves and measurement techniques are proposed. Second, origins and exposure of $PM_{2.5}$ are assessed. Personal, kitchen area microenvironment, and ambient organic $PM_{2.5}$ data were analyzed using positive matrix factorization, to better understand source types and relative importance at the different scales. Third, personal carbon monoxide exposure results are presented for intervention participants. Fourth, methods and results are presented for a proximity assessment system used to enrich personal exposure measurement data. Fifth, a laboratory assessment is presented for a widely used electrochemical carbon monoxide (CO) exposure monitor to better understand its strengths and limitations as relevant to this study.

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ETHICAL CONSIDERATIONS

The work presented in this thesis was part of the REACCTING study, which 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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CHAPTER 1

1.1. 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). Cookstoves present a compelling avenue towards providing benefits on three fronts: 1) improving human health, 2) preserving local ecosystems, and 3) reducing greenhouse gas emissions. Introduction of 'improved' cookstoves (ICS) targeting these goals also addresses multiple focus areas of the millennium development goals and the sustainable development goals (UN, 2016). Here, the term 'improved' cookstoves is written in the sense suggested by Smith and Dutta (2011), noting that cookstoves are more likely to improve health and quality of life the more similar they are to the gold standard of cooking with natural gas. Among many regions that have been heavily impacted by inefficient household energy use practices with high costs on human and environmental health, we chose to undertake a cookstove intervention research project in the Kassena-Nankana (K-N) Districts in Upper East Ghana.

Ghana as a whole is experiencing alarming deforestation rates, with 33.7% of forest area (2.5E6 hectares) lost since 1990, and a 2.19% annual deforestation rate from 2005-2010 (FAO, 2010), due in part to the 84% of the rural population that uses wood for cooking (Ghana Millenium Development Goals Report, 2015). The K-N Districts, with a population of 156,000 (80% of households are considered rural) (Oduro et al., 2012), is almost entirely categorized as a high-risk region for desertification (Adanu et al., 2013). The Sahel, which encompasses the K-N Districts, was devastated by a severe drought in the 1970's-1980's (Dai et al., 2004). Future climatic conditions show mixed results, with some projecting increased rainfall as the oceans warm, and others projecting reductions (Dai et al., 2014). Multi-model aggregations by Aloysius et al. (2016) report average precipitation increases in Equatorial West Africa in the range of 1.7-4.3% in the near term (2021-2050), and 2.6-8.4% in the long term (2070-2099), while average surface temperature increases are from 1.4-1.5 ^{o}C in the short term, and 2.1-4.3 ^{o}C in the long term. Temperature increases alone will be challenging for the population to adapt to, and deforestation of the region will render the area increasingly inhospitable and may increase the rate of migration (Warner et al., 2009; Antwi-Agyei, et al., 2012).

The Ghanaian government recognized the health and environmental risk early on, and implemented various programs to reduce biofuel consumption, though most have emphasized fossil fuel use as a means to reduce consumption. One effort to reduce biofuel consumption came in 1990, when the Ahibenso charcoal stove program was introduced to develop and deploy a fuel-efficient charcoal stove. 40,000 stoves were sold by 1993, but then government funding ran out and the venture ceased (Kemausuor et al., 2012). Despite some success with improved cookstoves, the focus of government energy programs for home energy use was liquefied petroleum gas (LPG), which was better aligned with economic aspirations of reaching middle-income status as a country by 2015, and seemed likely due to the completion of the West African Pipeline, providing Nigerian gas to West African countries. Despite a lack of government emphasis, the ICS market grew substantially in the 2000's, due to a combination of economic conditions described in the insightful Ghanaian cookstove market analysis by Agbemabiese et al. (2012). In 2002, Enterprise Works introduced the Gyapa coalpot, an improved coal burning stove like the Ahibenso, and sold 200,000 in the next four years. In 2006, a company called Toyola brought a similar charcoal stove to market, and have continued growing, but their experience is indicative of the difficulties faced by the sector, with numerous near-collapses due to issues like the inability to secure financing, and power shortages slowing production (Agbemabiese et al., 2012). Both Toyola, and Enterprise Works, with whom we partnered to produce the Gyapa woodstove used in our intervention, continue to operate in Ghana. The majority of current Ghanaian government efforts to reduce biofuel from cooking, however, are centered on LPG once again (Ghana Country Action Plan for Clean Cooking, N.D.) with 23.1% of Ghanaian households currently using that fuel as their main cooking fuel (Karimu et al., 2016).

Ghana has also attempted to improve environmental conditions with forest management plans, including efforts to reforest the North of the country (Makain et al., 2005). In 2007, a coalition of 20 countries led by the African Union began the 'great green wall project' with the intention of slowing desertification by planting millions of trees to improve land health. Ghana is expected to formally join the program in 2016, possibly due to the success observed in Niger and Burkina Faso (Reij and Winterbottom, 2015). To succeed, there must be

buy-in from local communities to nurse the trees into adulthood and to reduce pressure from biomass fuel harvesting, and ICS could be a major part of the solution.

Technologically, the problem of open fire cooking using solid fuels seems relatively straightforward to address: a wide variety of improved cookstoves and cleaner fuel sources exist that are more efficient and can reduce air pollutant emissions. Yet efforts to make these technologies available in areas of need throughout the world have often failed to achieve their intended results (Smith et al., 2014; Hanna et al., 2012). Human behaviors – specifically, acceptance and use of improved stoves – are key to the success of any cookstove intervention (Smith et al., 2014; Hanna et al., 2012; Gupta et al., 2006; Taylor et al., 2012; Lewis and Pattanyak et al., 2012; Mensah and Adu, 2015).

Two key and related challenges are locally appropriate stove selection and promotion by those introducing new technologies, and sustained stove adoption and use among target populations. By stove selection, we are referring to the processes of selecting the "right" technology (or mix of technologies) that is most likely to meet the needs of the target population while achieving meaningful reductions in negative health and environmental impacts. Some argue that only the cleanest, most advanced, and usually imported cooking technologies should be promoted, since these have the highest probability of having meaningful impacts on health and environmental outcomes. Others contend that introducing affordable, feasible, locally-produced cookstoves that are more efficient than open fires and more aligned with the unique cooking practices and needs of a given context can be an effective first step toward moving households up the "technology ladder" in the long run (Hiemstra-van der Horst and Hovorka, 2008; Simon et al., 2014). Conceptually, the stove or energy "ladder" model is rooted in a neo-classical understanding of energy use that implies cleaner fuel usage with rising socioeconomic status (Van der Kroon et al., 2013). Typically, this model also implicitly assumes that households rely on a single source of cooking energy at any given time.

Empirically, however, studies have found that rather than moving linearly up this energy ladder in a step-by-step fashion, households often rely simultaneously on multiple types of fuel

and cooking technologies to meet their cooking needs (Davis, 1998; Taylor et al., 2011; Van der Kroon et al., 2013; Campbell et al., 2003; Ruiz-Mercado et al., 2011). This energy or technology "stacking" allows households greater flexibility: they can use different types of stoves for different purposes, or alternate among different fuels (essentially moving both up and down the ladder) depending on availability and cost (Masera et al., 2000; Van der Kroon et al., 2013; Campbell et al., 2003, Simon et al., 2014; Elias and Victor, 2005; Ruiz-Mercado et al., 2011). Of course, these two models may both be correct in some respects; while households may continue to use a mix of technologies, it is possible that the technologies that comprise the cooking "stack" may become cleaner over time.

The extent to which new stoves are folded into the technology stack and can ultimately displace traditional cooking methods (leading to cleaner kitchens overall) depends heavily on how well suited these new technologies are to local culture and cooking practices. For example, a study of cooking practices in Guatemala showed that more affluent households (receiving remittances from migrant family members) had liquid petroleum gas (LPG) stoves but continued to rely on wood-burning stoves for most of their cooking needs because these stoves were better suited to the preparation of staple food items (beans, corn, and tortillas) (Taylor et al., 2011). Ultimately, without incorporating traditional cooking practices into the design process, even low-cost stoves are unlikely to be used (Heltberg, 2005).

Considering such evidence of stove and fuel stacking throughout the world, and practical questions of market access, successful introduction of gold standard LPG systems to new populations will require substantial time and effort, even on the scale of generations. Some argue that the majority of effort put into the field should be put towards transitioning directly to LPG (Smith and Dutta, 2011; Simon et al., 2014), because indeed, LPG has a much better chance than any biomass cookstove of achieving the reductions in HAP exposure that can yield life-changing health benefits (Burnett et al., 2013). This work, among a growing body of literature, provides further understanding of how Northern Ghana is impacted by traditional and 'improved' cooking practices, and how it may transition in the future. Will the population consistently and exclusively use an improved cooking technology if given the opportunity? Will

real-world use of improved cookstoves result in exposure reductions of components of woodsmoke? Is woodsmoke from cooking in fact the most significant source of pollution impacting personal exposure, or are there other important sources and behaviors associated with exposure? And are personal exposure measurements reliable and unbiased?

1.2. Our Approach to filling the knowledge gaps

In this study, we considered the causal chain between cooking behaviors and health/environmental outcomes, as visualized in Figure 1-1.

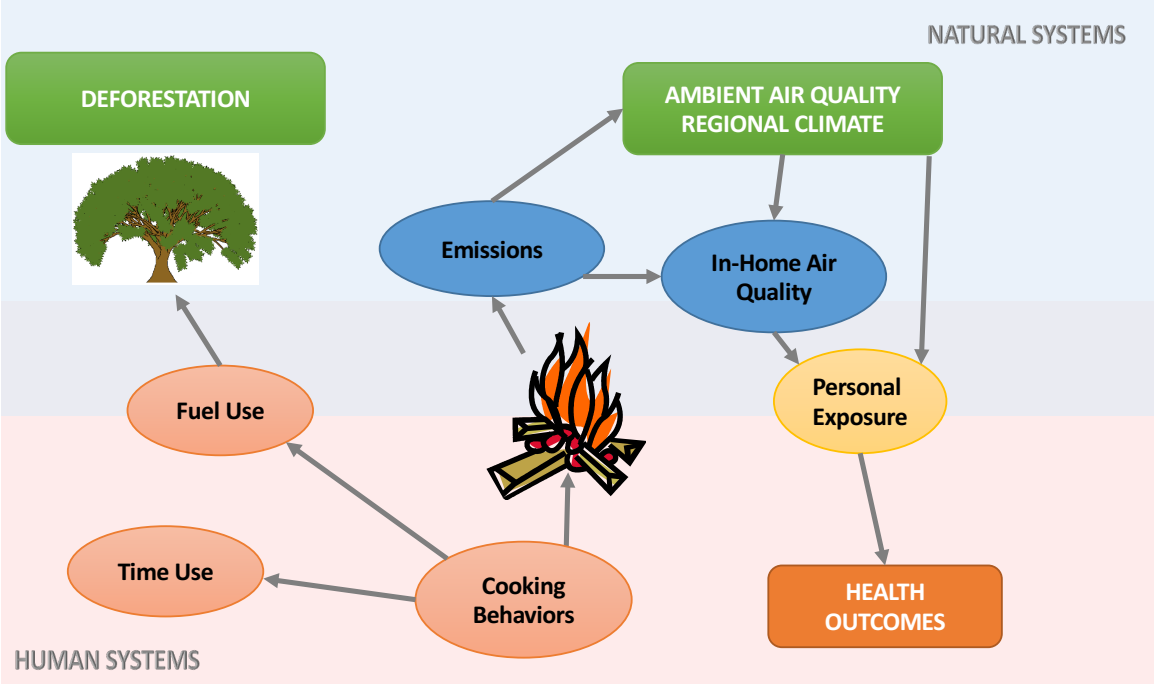

Figure 1-1. Causal chain of the REACCTING study. Our cooking technology intervention affects cooking behavior, which both affect emissions, which affect regional, household, and personal air quality, which affect health and the environment.

The first step in the causal chain involves cooking behavior, specifically stove adoption and use among households. Chapter one presents our current understanding of cookstove use and adoption from the REACCTING intervention. The first year of cookstove use data derived from stove usage monitors (SUMs) is analyzed in conjunction with quantitative survey results. Prior works have developed methods to assess stove use of both traditional and intervention stoves using stove use monitors (SUMs) (Ruiz-Mercado et al., 2009, 2011), and surveying

(Simons et al., 2014; Thomas et al., 2013). Each of these measurement approaches has its strengths and weaknesses. Surveys are subject to recall and social desirability bias (i.e., respondents may be reluctant to admit that they have not used new stoves provided by researchers), but allow collection of detailed, qualitative information on why stoves may or may not be used (Mobarak et al., 2012). Meanwhile, SUMs allow cooking events to be estimated from a time series of stove temperature measurements, but require substantial investment in temperature monitors and their maintenance, and effort in characterization of the temperature monitor/stove system. A combination of methods may thus be optimal to fully characterize stove use. Mixed methods in general have gained attention (Stanistreet et al., 2015) as researchers and projects come to understand the interconnectedness of home energy use practices seen through practices like stove stacking, the use of multiple cooking technologies to meet their household energy needs.

Prior study of cookstove adoption in the region was limited to a study by Burwen and Levine (2012) in the Sissala district of Upper West Ghana, that assessed the use of a locally designed built-in brick and mortar stove. They found substantial continued use of traditional stoves (stove stacking), especially as stoves fell into disrepair over time. It is in the interest of the region to further explore cooking technologies and behaviors there, with the goal of a thriving market for clean household energy systems.

The next step in the chain involves the quantification of cooking emissions from the improved and traditional cooking methods. Many studies have measured real-time biofuel cooking emissions in laboratory settings using standardized testing procedures (e.g., Carter et al., 2014; Jetter et al., 2012), but fewer have done field-based measurements (Johnson et al., 2008, 2011; Roden et al., 2006, 2009). Emission measurements in the field are essential since many key factors may vary between the lab and field setting. These results are presented in forthcoming work by other members of the REACCTING team.

Chapter two fills gaps in the next step of the chain, the impact of changes in cooking emissions on personal and household pollutant levels. To measure these impacts, studies have

most commonly monitored household and personal exposures to carbon monoxide (CO) and particulate matter with aerodynamic diameter less than 2.5 micrometers in diameter ($PM_{2.5}$). $PM_{2.5}$ has been strongly linked to negative health outcomes in epidemiologic and biological studies (Pope et al., 2009; Naeher et al., 2007; Smith et al., 2009; Janssen et al., 2011), and can have positive and negative climate forcing effects, depending on optical characteristics (Ramanathan and Carmichael, 2008; Bond et al., 2013). Indeed, residential solid fuel combustion is the largest single contributor of global anthropogenic black carbon $PM_{2.5}$ emissions, at 45.6% (Klimont et al., 2016).

In contexts where cooking-related emissions are a dominant source of $PM_{2.5}$ exposures, reducing use of traditional biomass stoves in favor of ICS may be an effective solution for reducing exposures. Of course, understanding the sources of $PM_{2.5}$ helps determine if that is the case. Chapter two investigates the effect of the intervention on carbonaceous $PM_{2.5}$ at the ambient, home, and personal scale using chemical and statistical analyses.

At the regional scale, there has been limited study of the sources of $PM_{2.5}$ in Africa, though it is increasing with population and economic growth, which has lead countries to pursue air quality management plans (Ndamitso et al., 2016). One of those few studies took place in the city of our study site, Navrongo, Ghana, where $PM_{2.5}$ filter samples were collected from 2009–2010 (Ofosu et al., 2013). Using source apportionment techniques, observed particulate elemental carbon (EC) and organic carbon (OC) and speciated elements were used to identify six sources of $PM_{2.5}$, namely two-stroke engine combustion, diesel combustion, gasoline combustion, soil, biomass combustion, and road dust. After dust, biomass combustion was found to be the second largest contributor to ambient PM concentrations in Navrongo. Regional $PM_{2.5}$ monitoring has also been performed in the capital of Ghana, Accra (Ofosu et al., 2012; Zhou et al., 2013; Aboh et al., 2009), Nigeria (Obioh et al., 2013), Ouagadougou, Burkina-Faso (Boman et al., 2009), Kenya (Gatari and Boman, 2003), and Cairo (Abu-Allaban et al., 2007).

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. This study is one of only a handful to have separated the contributions from sources related to biomass combustion from other sources unrelated to the intervention. Although the chemical analysis is challenging relative to other measurement approaches, the results can provide very strong conclusions relative to other methods. By using the chemical analysis results, 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.

Substantial reductions in traditional stove use are required to achieve significant health improvements (Burnett et al., 2013; Johnson and Chang, 2015), and quantifying the extent to which exposures are reduced when cooking practices change, and understanding how much cooking-related sources drive $PM_{2.5}$ exposure in different contexts, are key challenges for this field.

Chapter three presents the intervention effects on personal exposure to carbon monoxide (CO), another component of biomass woodsmoke associated with negative health and climate effects. Carbon monoxide (CO) is commonly measured in cookstove and air pollution exposure studies due to its association with adverse health effects (Smith et al., 2000; Longo et al., 1977; Astrup, 1972), the low cost of real-time wearable monitors, logistical challenges of measuring other pollutants, and correlation with other co-emitted pollutants. Short-term CO exposure is associated with respiratory and cardio-vascular morbidity, as well as mortality, while long-term CO exposure has been associated with negative birth outcomes, developmental effects, and central nervous system effects, among others (Saldiva et al., 2004; Naeher et al., 2007; Smith et al., 2009; Longo et al., 1977).

Simplicity and cost of measurement, along with the well-established health effects associated with $PM_{2.5}$ (e.g., Naeher et al., 2007), have led to the study and use of CO as a

surrogate for $PM_{2.5}$. Previous works have found varied results for this relationship, with differences ascribed to variability in source types like fuels and stoves, behaviors, and home designs, among other characteristics (Naeher et al., 2001; Northcross et al., 2010; McCracken et al., 2013; Dionisio et al., 2012a; Carter et al., 2016 *in press*). In a meta-study on the 61 studies in 27 countries that have collected personal or cooking area measurements of both CO and $PM_{2.5}$, Carter et al. (2016 *in press*) found correlation coefficients (*r*) for personal exposure ranging from 0.22 to 0.97 (median=0.53), and from 0.10 to 0.96 (median=0.71) for cooking areas. Here, we add to the literature on this topic to further the understanding of source types for rural communities in Northern Ghana.

Chapter four describes methods and results for a novel Bluetooth Low Energy (BLE) Beacon-based time-activity measurement system designed to improve personal exposure assessment. Time-activity information is valuable in many fields of study, including personal exposure assessment, but traditional time-activity measurement approaches (Freeman and Tejada, 2002) are resource intensive and can result in misclassifications (Clark et al., 2013). Past cookstove studies have used self-reported or manually collected time-activity data (Brauer et al., 1996; Albalak et al., 1999; Zuk et al., 2007; Cynthia et al., 2008; Dionisio et al., 2012a), but an automated system would simplify measurements and improve accuracy. Improved data could then lead to improved modeling of personal exposure from microenvironment measurements. Such a system could also allow for separate analysis of exposure from sources at home vs. away from home, and other such categories. The BLE Beacon proximity monitoring system we have developed and tested fills these gaps in the exposure assessment toolbox.

Previously, there was no viable system available for our application. Wireless technologies such as Wi-Fi allow precise indoor location estimates, but resources are required to train the identification system and the presence of Wi-Fi access points is required (Jiang et al., 2011). Global Positioning System (GPS) devices can be used to assess location (Elgethun et al., 2003; Rooney et al., 2012), but these tools tend to have a relatively high power consumption and accuracy can suffer in regions with certain geographic characteristics and, perhaps more crucially, indoors. 'Passive' Radio Frequency Identification (RFID) tags can be

uses as binary room-location indicators, but users must place their small 'passive' type badges close to the RFID receiver, making compliance a concern. Larger 'active' RFID badges that use a battery to increase transmission power have been shown to perform well in indoor location testing (Ni et al., 2004), and could be a viable technology for this application if the additional logging capabilities conferred by the phones are not needed. Costs are also generally higher than Beacon systems. Allen-Piccolo et al. (2009) introduced an ultrasound-based time-activity monitoring platform (UCB-TAMS) for cookstove applications that displayed promising results. Ultrasound has lower attenuation than Bluetooth, improving signal consistency in difficult geometries or crowded spaces, but such systems have not come into widespread use. System cost is similar to our BLE Beacon system, but we were unable to perform a direct comparison between the systems due to UCB-TAMS availability.

In this work, our BLE Beacon proximity detection technology was first demonstrated in a calibration and validation study, along with relevant data processing methods. The system was then used in Ghana, in conjunction with personal CO exposure monitoring and cooking area CO monitoring. This deployment demonstrated the benefits of such a system by analyzing these relationships and interactions in greater resolution than previously possible with a low-cost system.

Chapter five delves into the workings of an electrochemical CO sensor that was used throughout our study (Lascar USB-CO, using the Nemoto NAP-505 sensor). Electrochemical gas sensors have enabled gas sensing reliable and accurate enough to improve scientific and engineering understanding in a variety of fields, from combustion processes, to ambient pollution monitoring. CO sensors used throughout the REACCTING study were of the electrochemical type, specifically Alphasense CO-B4 and CO-BX units in cooking area microenvironment measurements, and Nemoto NAP-505 sensors (used in Lascar USB-300 and USB-1000 monitors) for personal exposure measurements. Prior works have proposed calibration methods for such sensors for personal exposure monitoring (Smith et al., 2009). Young and Jones (2014) investigated response times of the Lascar CO monitor used in our

study, applying a model developed by Cheng et al. (2010) for a different model of personal CO monitor, to correct for slow response times that can affect exposure estimates.

Electrochemical sensors have been characterized extensively by the manufacturers and independent researchers (Masson et al., 2014; Mead et al., 2013; Borrego et al., 2016; Spinelle et al., 2016; Buck et al. 2013) but important questions remain about properties and behaviors, especially as related to long-term use. Specifically, how sensor aging impacts sensitivity and time-response have not been reported in past works. Additionally, we are not aware of uniform guidelines on calibration for exposure studies. Providing recommendations based on our experience and laboratory observations should be of help to the field.

In our study, a group of ten sensors was tested intensively in a laboratory setting to characterize performance stability over time, rise and decay times, temperature effects, and how the sensor history and changes in physical sensor properties could affect these characteristics. Our work focused on the stability of sensor properties during and throughout intense usage periods, similar to measurements encountered in high-exposure cookstove studies. There is wide variability in the level of expertise and resources available for groups trying to make personal exposure measurements in the developing world. We sought to characterize various performance parameters of a commonly used CO measurement instrument to provide practical guidance on use, and pitfalls to avoid.

The final step in the causal chain from the intervention to health and environmental outcomes remains to be analyzed and are not presented as part of this dissertation. The REACCTING study directly assessed health indicators as a measure of the intervention impacts, namely, self-reported symptoms such as eye irritation and headaches (Diaz et al., 2007), and biomarkers of systemic inflammation linked to smoke exposure from blood samples (Banerjee et al., 2012). Environmental outcomes on biomass fuel consumption will be presented along with cooking emissions results from tests performed in the field.

CHAPTER 2

Assessment of cookstove stacking in Northern Ghana using surveys and stove use monitors

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Abstract

Biomass burning for home energy use is a major health and environmental concern. While transitioning to cleaner cooking technologies has the potential to generate significant health and environmental benefits, prior efforts to introduce improved cookstoves have encountered many hurdles. Here, we focus on the increased stove use hurdle; households tend to use improved stoves alongside their traditional stoves rather than replacing them entirely, a phenomenon called cookstove "stacking." This work provides a systematic, multi-method assessment of households' cooking behaviors and cookstove stacking in the context of a 200-home randomized cookstove intervention study in Northern Ghana. Two stoves were selected for the intervention, a locally made rocket stove (Gyapa) and the Philips HD4012 LS gasifier stove. There were four intervention groups: a control group, a group given two Gyapa stoves, a group given two Philips stoves, and a group given one of each. Two stoves were distributed to each home in an attempt to induce more substitution away from traditional stoves. Adoption and usage patterns were quantified using temperature loggers at a subset of homes, as well as quarterly surveying in all households. We find that using multiple stoves each day is common practice within each intervention group, and that the two groups given at least one Gyapa had the largest reductions in traditional stove use relative to the control group, though use of traditional stoves remained high in all groups.

2.1. Background and motivation

2.8 billion people burn solid fuels for cooking (Bonjour et al., 2013) and the resulting air pollution is the third leading risk factor for the global burden of disease, contributing to 4 million premature deaths per year (Lim et al., 2012). The environmental impacts from this

activity are substantial. In addition to contributing to regional deforestation and forest degradation (Chidumayo et al., 2013), residential combustion (including wood, agricultural waste, animal waste, and coal) contributes an estimated 32% of particulate black carbon, and 64% of particulate organic carbon to global non-open burning emissions (Bond et al., 2013).

To address these issues, cookstove distribution programs and studies to replace traditional cooking methods with cleaner, more efficient ones continue to grow in scope and magnitude. Measurement of cookstove adoption is critical in determining the feasibility and likelihood of success of these programs. There are many factors involved in the decision to adopt a new stove, among them income, education, availability of viable clean cookstoves, fuel availability, financing, location, and cultural norms (Barnes et al., 1993; Pine et al., 2011; Jan et al., 2012; Jeuland et al., 2012; Lewis and Pattanayak, 2012; Malla et al., 2014). Previous studies have found evidence that even when intervention cookstoves are used regularly, households often maintain regular use of their traditional stoves, a practice known as stove stacking (Pillarisetti et al., 2014; Stanistreet et al., 2015).

Research on Emissions, Air quality, Climate, and Cooking Technologies in Northern Ghana (REACCTING) (Dickinson et al., 2015) is a 200-home cookstove intervention study in the Kassena-Nankana (K-N) Districts of Northern Ghana, designed to learn about cooking behaviors and their impacts in this region. Past personal air pollution exposure studies in Ghana have measured worryingly high levels of CO (Burwen and Levine. 2012) and PM (Arku et al., 2008; Rooney et al., 2012; Van Vliet et al., 2013) due to cooking and other combustion sources. The ecological motivation is also strong, as the study is located within a climatically sensitive region at high risk of drought and forced migration (Warner et al., 2009; Antwi-Agyei, et al., 2012). Ghana as a whole is experiencing alarming deforestation rates, with 33.7% of forest area (2.5E6 ha) lost since 1990, and a 2.19% annual deforestation rate from 2005-2010 (FAO, 2010). The Upper East region, encompassing our study area, is almost entirely categorized as a high-risk region for desertification (Adanu et al., 2013). Assessment of adoption and stacking has not been undertaken in this region of Africa, where the mix of remoteness and indoor/outdoor cooking offers new challenges.

2.2. Measuring Stove Use

In REACCTING, we determine the extent of stove use and stove stacking using two methods, stove usage monitoring with temperature data loggers (here referred to as stove usage monitors, or SUMs), and quantitative surveying. Both types of data have strengths and limitations. Stove usage monitoring allows identification of cooking events from extended time series of stove temperature (Ruiz-Mercado et al. 2012; Mukhopadhyay et al., 2012; Graham et al., 2014), with lower potential for the reporting biases encountered with surveying (Thomas et al., 2013; Wilson et al., 2015). SUMs have the advantage of eliminating the biases associated with self-reporting that have been observed in some studies (Thomas et al., 2013; Wilson et al., 2015). However, other sources of bias and measurement error are still possible with SUMs, including reactivity effects (higher use due to the knowledge of being monitored – see Thomas et al. 2016). Considerable uncertainty also remains in detecting cooking events using SUMs data, particularly for the traditional 3-stone fires (TSFs). In addition, SUMs data collection is costly. As a result, we were only able to collect SUMs data for a subset of study households rather than the entire sample. Meanwhile, surveys were conducted in all households at multiple discrete time points (quarterly), and provide us with detailed contextual information along with (potentially mis-reported) stove use information. Survey information such as foods cooked and fuel types used with each stove shed light on how and why certain stoves are being used by different households. Used in combination, survey and SUMs data can more effectively inform future cookstove and fuel improvement efforts in the region.

Our study makes an important contribution to the literature by examining cooking behaviors in a region that has received relatively little attention: Northern Ghana. In addition, this work is among the first to publish results on the use of multiple intervention stoves alongside traditional stoves. Previously, Loo et al. (2016) performed a study in Kenya assessing user perspectives on six different improved combustion stoves (ICSs) rotated through homes for two-week periods.

2.3. Methods

2.3.1. Study population and design

The REACCTING study ran from November 2013 to January 2016. The study population consisted of households in the K-N Districts that 1) were classified as rural, 2) used biofuels as

their main cooking fuel source, 3) had at least one woman of childbearing age (18-55) and one child under five, 4) used a borehole as their primary water source, and 5) did not have electricity in the home. Using data from the district-wide Health and Demographic Surveillance Survey (HDSS) (Oduro et al. 2012), we identified the sample frame of households that met these eligibility criteria, and then used a cluster random sampling method to select 200 households for inclusion in the study. Detailed information on study design and sample selection is presented in Dickinson et al. (2015).

A baseline survey conducted in all 200 households prior to the stove intervention provided detailed information about local cooking practices that confirmed observations the study team made during the two years prior to the start of the study, and which informed the design of the REACCTING intervention. Even before the introduction of any new stoves, households in this area were cooking with multiple stoves, and with a mix of cooking technologies (Figure 2-1 Baseline (pre-intervention) cookstove technology mix among study householdsFigure 2-1). The most common cooking technology in this area is a traditional wood-fired 3-stone stove, but the majority of households (70%) owned at least one charcoal stove, locally known as a "coal pot," as well. Only 10% of households relied on a single stove to meet their cooking needs; 38% of households had two stoves at baseline, and the remaining 53% had three or more stoves.

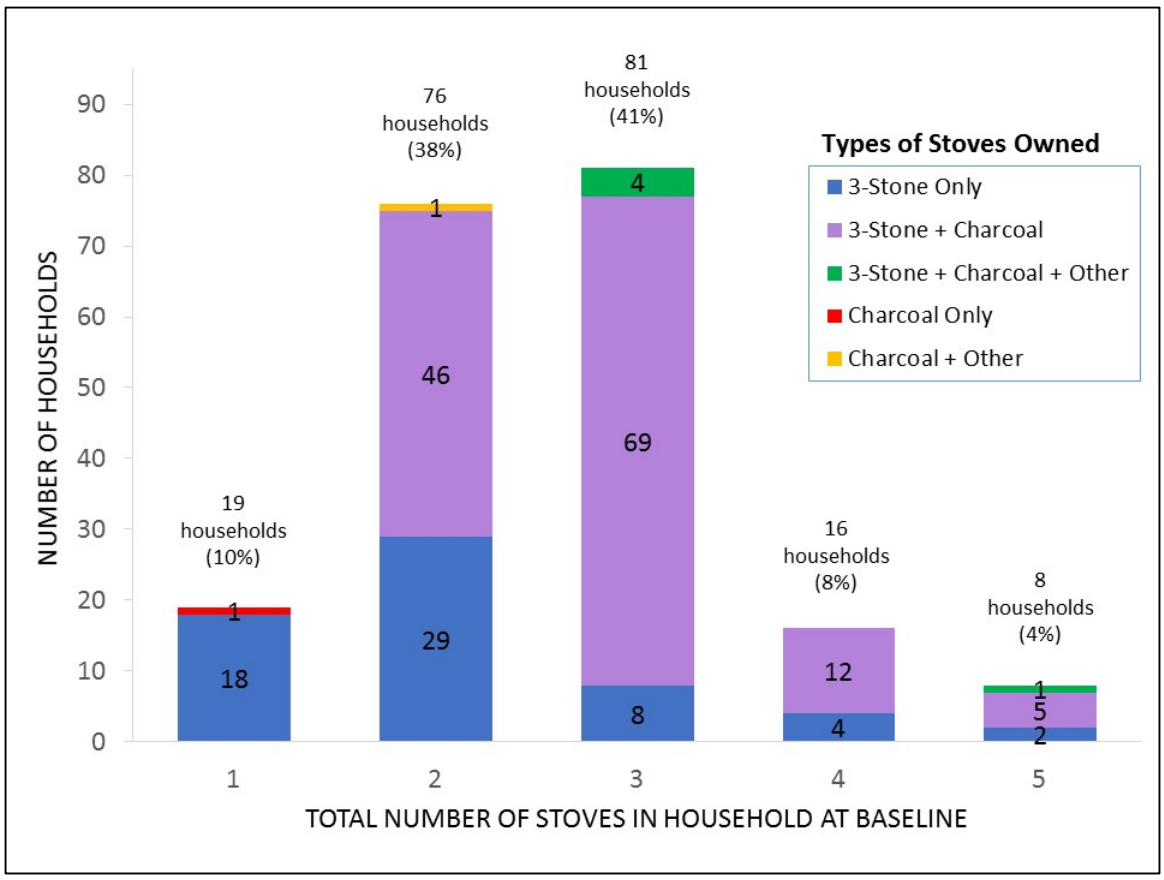

Figure 2-1 Baseline (pre-intervention) cookstove technology mix among study households

Further, we observed that local cooking practices link stove and fuel types to specific foods. The dishes that are commonly prepared and eaten in this area determine households' stove needs; these staple dishes and associated cooking methods are listed in Table 1. The items on this list can be broken into two groups according to their cooking method or requirements. Dishes that require rapid boiling (such as soup or rice) fill one category, while dishes requiring more intensive effort such as vigorous stirring (Tuo Zaafi, or TZ for short) are in a second. The baseline survey, conducted from November-December 2013, indicated that by far the most common dishes in this area are TZ and vegetable soup, which are often consumed together, and the next most common dish is rice. We also observed fairly consistent patterns in the types of stoves used to cook each dish. The TSF is the most commonly used stove for all types of dishes. However, a sizeable portion of vegetable soup meals are cooked over charcoal stoves, and these stoves are also used to cook rice on some occasions. Meanwhile, TZ is cooked almost exclusively on TSFs.

Table 2-1: Dishes cooked in the K-N Districts study homes

Dish	Description	Cooking Method	% Of Households Cooking Dish		
			Total	On 3-Stone Stove (TSF)	On Charcoal Stove
Tuo Zaafi (TZ)	Thick porridge made with millet or maize flour, often served with vegetable soup	A mixture of water and millet flour is added to boiling water. Cold water is then added, and after the mixture is heated, half is moved to another bowl. More millet flour is then added, all the while stirring vigorously. The separated portion is added back in, and it is served hot.	57.0%	55.5%	1.5%
Vegetable soup	Soup made with keneff-kanzaga, vio, alefu, yambola, okro, all local vegetables. Fish or meat is sometimes added depending on availability.	Cut vegetables are boiled 10-15 minutes. Groundnut paste or bean flour is added, sometimes with fish or meat. Cooked until it boils again.	56.5%	43.0%	19.0%
Rice	Multiple varieties are available and are used to make jollof rice, rice balls, or plain rice, often with a sauce.	Cooked in metal or earthenware pots	39.0%	33.0%	6.5%
Beans	Many varieties are commonly eaten in the K-N districts, in various dishes	Varies	6.0%	6.0%	0.0%
Other dishes	Pompuka (3.0%), Bombara beans (1.5%), Tubani (1.5%), Tubers (1.5%), Corn (1.5%), Porridge (1.5%), Banku (0.5%)	Varies	10.5%	7.5%	3.5%

Data on % of households cooking each dish are from REACCTING baseline survey questions that asked about use of each stove in the household on the day prior to the survey. Households may have cooked a dish multiple times on that day using different types of stoves, so that the 3-stone (TSF) and charcoal columns do not necessarily equal the "total" column. Given local cooking practices and previous observations of stove stacking behavior in other contexts, the REACCTING study intervention was designed to distribute two stoves to each household randomized into an intervention group in order to create greater potential for households to substitute away from their traditional stoves while continuing to meet their cooking needs. Furthermore, we selected two different stove technologies for our intervention based on two key considerations. First, as discussed in Dickinson et al. (2015), we hoped to

contribute to an ongoing debate among cookstove researchers and policymakers over whether movements up the stove technology ladder should be made incrementally (i.e., starting with locally made, affordable, low-tech stoves) or transformationally (moving directly to the cleanest technologies available). Thus, we decided to compare adoption and performance of a locally made ceramic and metal rocket stove (Gyapa) alongside the theoretically cleaner Philips stove. The imported Philips is more expensive, and of the forced draft design, requiring battery charging with the provided solar panel every few days. Second, we suspected that households might use these two technologies differently, with each being suited to meeting different cooking needs. In particular, in piloting several types of cookstoves in the study area, households expressed doubts about being able to cook TZ on some of the stove models given the need for vigorous stirring. These concerns informed the design of the Gyapa stove, which was developed specifically for this project (though it bears resemblance to the one used in a study from Accra by Pennise et al. (2009)), as well as the design of a rebar stand to increase the stability of the Philips stove (Figure 2-2).

Figure 2-2. Digit-TL SUM placements on the Gyapa, Philips, and 3-stone fires, from left to right. The Philips stove is shown with the specially designed rebar pot stand. The 3-stone fire at right is shown with wood and millet stalks for fuel.

In November of 2013, the households were randomly placed into one of four intervention groups: one with two Gyapa stoves, one with two Philips stoves, one with each of those stoves, and a control group (no new stoves until the conclusion of the two-year study). During stove distribution assemblies, retired nurses working with the project team educated participants on the health, timesaving, and financial and environmental benefits of using the

improved stoves, and those receiving new stoves were encouraged to stop using their traditional stoves.

2.3.2. SUM methods

SUMs were initially deployed in January/February of 2013, on 103 stoves distributed over 45 households. They were placed on the improved cookstoves and the most-used traditional cookstove at 10-12 households from each study group. More SUMs were added in late 2014 and early 2015 to replace broken ones and to monitor more stoves in each home, as most homes had 3 or more stoves at baseline (Figure 2-1). This paper presents the first year of SUM and survey data, ranging from January 2014 – January 2015. The SUMs deployment time series is shown Figure 2-3, organized by the number of households monitored in each intervention group on the left panels, and the number of each type of stove monitored in each intervention group in the right panels. A one-month ramp-up time is apparent as SUMs were deployed, with a reduction in data coverage in the second half of the year in most groups, due to lost or damaged SUMs, or mistakes in SUM data management.

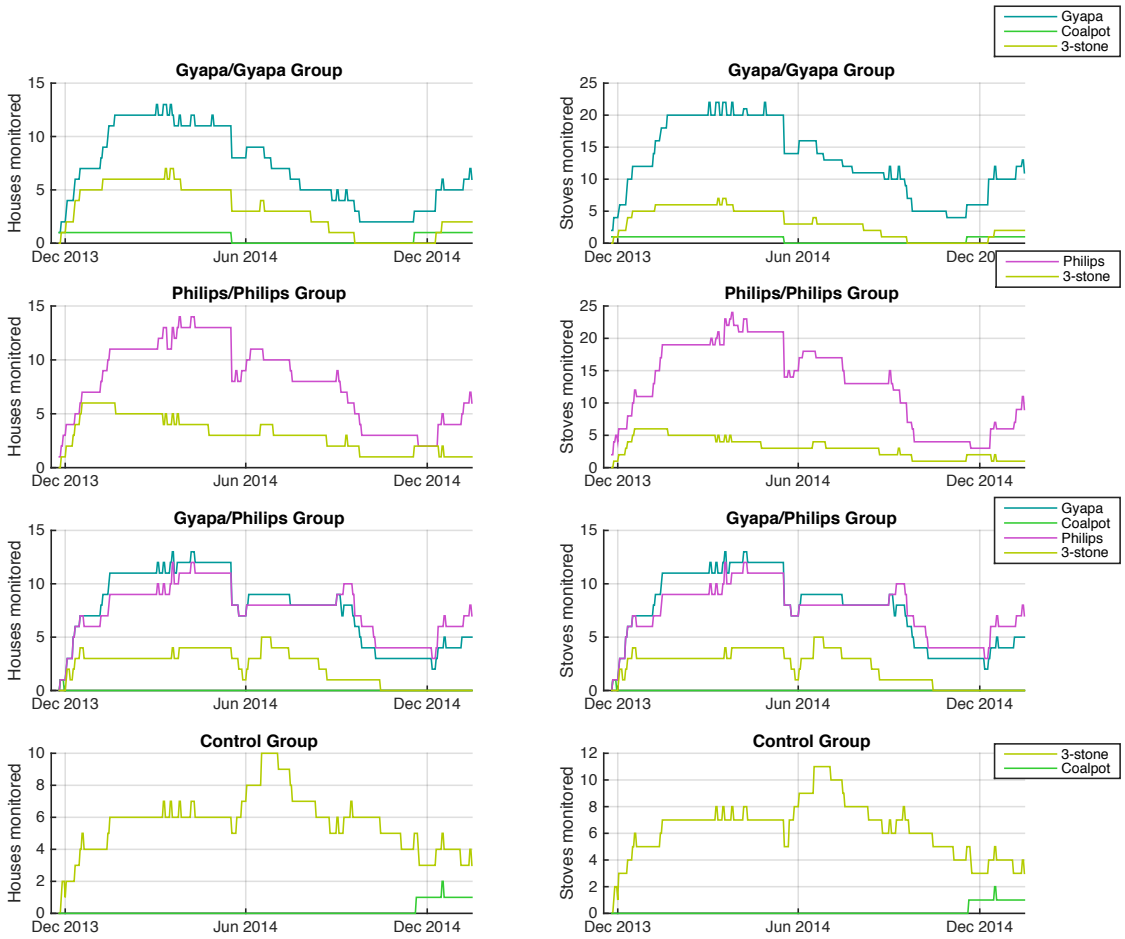

Figure 2-3. Digit-TL SUM deployment time series for each stove intervention group. The left panels show the number of households monitored for each stove type, while the right panels show the number of stoves monitored for each stove type. Note the difference in y-axis scales from left to right panels, as households often have multiple stoves monitored of the same type.

LabJack Digit-TLs temperature, light, and humidity monitors (LabJack, Lakewood CO) were used as SUMs. Humidity logging was disabled in favor of using a waterproof enclosure for the SUMs, while light logging was enabled on a subset of SUMs. The US$35 Digit-TLs measure temperature from -40 to 85 ^{o}C, with 0.5 ^{o}C resolution and reported uncertainty of 1.0 ^{o}C. Battery life is specified at over 3 years, with onboard memory capacity of 500,000 records. This allowed field workers to visit the homes every 4-6 months, at the selected 1-minute sampling rate. However, these SUM features also resulted in lost data because deployment errors or broken SUMs were not identified until the next scheduled visit, often months away.

The SUMs were placed strategically on the stoves to avoid surpassing the SUM temperature limit of 85 ^{o}C. SUMs placement on each stove type was tested prior to

deployment for the Philips and Gyapa stoves, but were placed conservatively to ensure they were not in danger of over-heating. This resulted in more challenging data analysis, as the peaks were less pronounced than in some other studies. On the Gyapa stove, SUMs were affixed with a metal strap near the center of the stove, while on the Philips they were attached with a hose clamp, which was screwed into one of the existing screw holes near the top of the stove. On the TSFs, the SUMs were wedged behind the largest stone in the fire with metal stakes. We were not able to put them underneath a stone as done in other studies, because the residents use plaster finishes in cooking areas that would have been damaged in the process of installing our relatively large SUMs. Typical placements are shown in Figure 2-2.

Cooking events were identified using a modified version of the algorithm described in Ruiz-Mercado et al. (2012), here referred to as the RMM algorithm. The 1-minute data from the SUMs were first smoothed using a 2^{nd} order polynomial with a 10-minute window. High variability remained from sample to sample even after this step, so the algorithm was modified to identify the start and end of a cooking event within 90 minutes of its peak to ensure cooking events were not mistakenly prolonged. This value was selected based on controlled cooking tests (CCTs; Bailis, 2004) performed concurrently during the study, which had a mean cooking time of 92.5 minutes, and maximum cooking time of 160 minutes (60 tests total). The thresholds required in the RMM algorithm were selected based on visual analysis of performance. The slope thresholds selected were based on the 0.1^{st} and 99.9^{th} percentile of the differences in consecutive slopes of the ambient temperature, or $0.09 C^{o}$/min and -0.12 C^{o}/min for rise and decay, respectively. This was higher than the thresholds selected using the 99^{th} percentile of non-cooking days by Ruiz et al. (2012). Our study experienced hotter and more dynamic ambient conditions, so we adjusted to capture events more effectively. Peaks within 1 hour of each other were grouped into single cooking events.

Individual SUMs cooking event identification was somewhat sensitive to parameter selection in the RMM algorithm, but overall usage trends did not change substantially when varying the parameters. Results were most sensitive to the threshold slopes for entering and departing a peak. In a subset of SUM time series, it was also difficult to identify cooking events with a high degree of confidence, mostly due to indoor-outdoor movement of stoves and a

preference for cooking outdoors during the non-rainy season. We may have falsely identified cooking events due to fast radiative heating from sunlight, predominantly during the midday hours. SUMs in direct sunlight were found to reach temperatures of up to 60 ^{0}C, whereas cooking events could reach peaks between 30 ^{0}C and 130 ^{0}C.

We assessed the uncertainty of the SUMs results by validating the SUMs cooking event identification algorithms using calibration data sets, in which thermocouples were collocated with SUMs on multiple stoves in the field. The training sets show that quick cooking events can be missed by the SUM if the peak identification threshold temperature is set too high, as such we set it at 30 ^{0}C for this analysis. On the longest training data set with a Gyapa stove, 39 cooking events were identified with the thermocouple, and the RMM algorithm identified 22 of those, with 11 false positives. This stove was left outdoors most days, making it one of the most challenging cases, so we expect this to be the upper limit of misclassification performance.

The performance for TSFs is likely similar to this in the worst cases, as TSFs are challenging to monitor (Burwen and Levine, 2012). However, even with a bias in the cooking period estimates from the stove types, the biases are expected to be comparable within stove types, so for example, comparisons and usage patterns over time for all Gyapa stoves can be considered valid.

Resource limitations prevented us from monitoring every stove in every home monitored, so cooking events with charcoal stoves and secondary TSFs were rarely measured. Some categories, such as the TSFs in the Philips/Philips group, had limited data due to damage to the SUM, theft of the SUM, or insufficient temperature variability during cooking events from poor SUM placement. The complete picture of cooking in some of these households may thus be obscured. To assess the impact of the low sample size in some of these categories, we randomly removed all cooking events from 1 household (jackknifing), to test the reliability of the use predictions on that particular dataset (Figure 2-4). The error bars on each stove-use category are the 5th and 95th percentiles of the 100 jackknifed data sets. The uncertainties associated with each group do not change our conclusions about use of the intervention stoves,

but low sample size and high uncertainty of the traditional stove use make our stacking and replacement results more tentative, as discussed later.

2.3.3. Survey methods

All 200 households participating in the study were surveyed at multiple time points throughout the study to measure stove use and preferences, among other topics. The first survey round was conducted in November/December of 2013, prior to the stove distribution, called baseline herein. Subsequent survey rounds were conducted in March, May/June, and August of 2014, and December 2014/January 2015. These survey rounds (rounds 2-5) were then used for comparison with SUM data (December 2013-January 2015). As part of each survey visit, interviewers completed a stove use questionnaire for all stoves in the household (Supplementary Information). That is, questionnaires were completed for each "old" stove used by the household prior to the study (charcoal stove and/or TSF), as well as each new stove (Philips and/or Gyapa). For each stove, respondents were asked to estimate a category of how many times the stove was used in the last week (0, 1-3, 4-6, 7 times). If the respondent reported that the stove was not used at all in the past week, the remaining questions for that stove were skipped and the interviewer moved to the next stove in the household. For stoves that were reportedly used at least once, the interviewer recorded whether the stove was in use "now" (i.e., at the time of the survey), and whether the stove had been used "yesterday." For current cooking and the prior day's cooking, additional questions asked what dishes were cooked and what fuels were used, as well as who did the cooking and how many people were fed by the meals cooked on the stove in question. Since cooking is a daily activity for most households, the "yesterday" question was designed to provide a good snapshot of cooking practices across the sample.

Self-reported behavioral data always raise concerns about possible mis-reporting. For example, in a study similar to ours in certain ways, Thomas et al. (2013) measured cookstove and water filter adoption in Rwanda using both surveying and quantitative monitoring, and found that respondents overreported use of both new technologies relative to monitor data. In the case of cookstoves, reported improved stove uses in the last week were 40% higher than measured with SUMs. In our study, we chose short recall periods (i.e., past week, yesterday) to

minimize recall bias and make it easier for respondents to provide specific answers about their cooking practices. In an effort to limit reporting bias and reactivity effects, project staff employed as part of the measurement and survey teams were trained to maintain a neutral attitude and encourage truthful reporting of stove use by participants, emphasizing the importance of collecting accurate information about users' experiences in order to improve the stoves and their usefulness in the future. Thus, during these visits participants were not explicitly encouraged to use their improved stoves, or use them exclusively, and there was no required cooking demonstration or instruction on the use of the stoves. However, it is of course possible that reporting biases remain in our data despite these efforts. In particular, we hypothesize that households in the intervention groups given stoves would tend to overreport their use of new (Gyapa and Philips) stoves, and underreport use of their traditional stoves. Potential for misreporting of stove use seems lower in the control group.

2.4. Results

2.4.1. Stove use across intervention groups

Figure 2-4 shows usage rates for the Gyapa, Philips, TSF, and charcoal stoves (charcoal SUM results are excluded here due to low SUM coverage, but survey results are presented). For these four stove types, the plot shows the rate at which households use any stoves of this type, as measured by SUMs and reported in surveys. For Gyapa and Philips stoves, survey-based estimates of stove usage rates are consistently higher than SUMs-based measurements. This largest discrepancy was 6.8%, for use of Philips stoves in the Philips/Philips group, where 61.5% of survey respondents said they used the stove yesterday, while SUMs showed that they were used on 54.7% of days. The smallest difference, 2.4%, was for Philips use in the Philips/Gyapa group, where surveys showed 51.1% of users used the stove 'yesterday', and SUMs showed use on 48.7% of days. Despite these discrepancies, the surveys and SUMs tell a consistent overall story regarding the relative patterns of use of these two stove types across intervention groups. Specifically, Gyapa stoves are used at substantially higher rates than Philips stoves. Gyapa/Gyapa households used a Gyapa stove on 82.6% of days according to surveys, or 77.9% according to SUMs. The Gyapa use rate in the Gyapa/Philips group was lower, at 61.5% and 54.7% of days per the surveys and SUMs, respectively. In contrast, the

Philips stoves from the Gyapa/Philips group were used on 25.8% and 23.8% percent of days (surveys, SUMs). The Philips/Philips group used a Philips stove on 48.7% and 51.1% of days (surveys, SUMs). This was close to twice the rate of Philips use from the Gyapa/Philips groups, and substantially less than the Gyapa use from the Gyapa/Gyapa group.

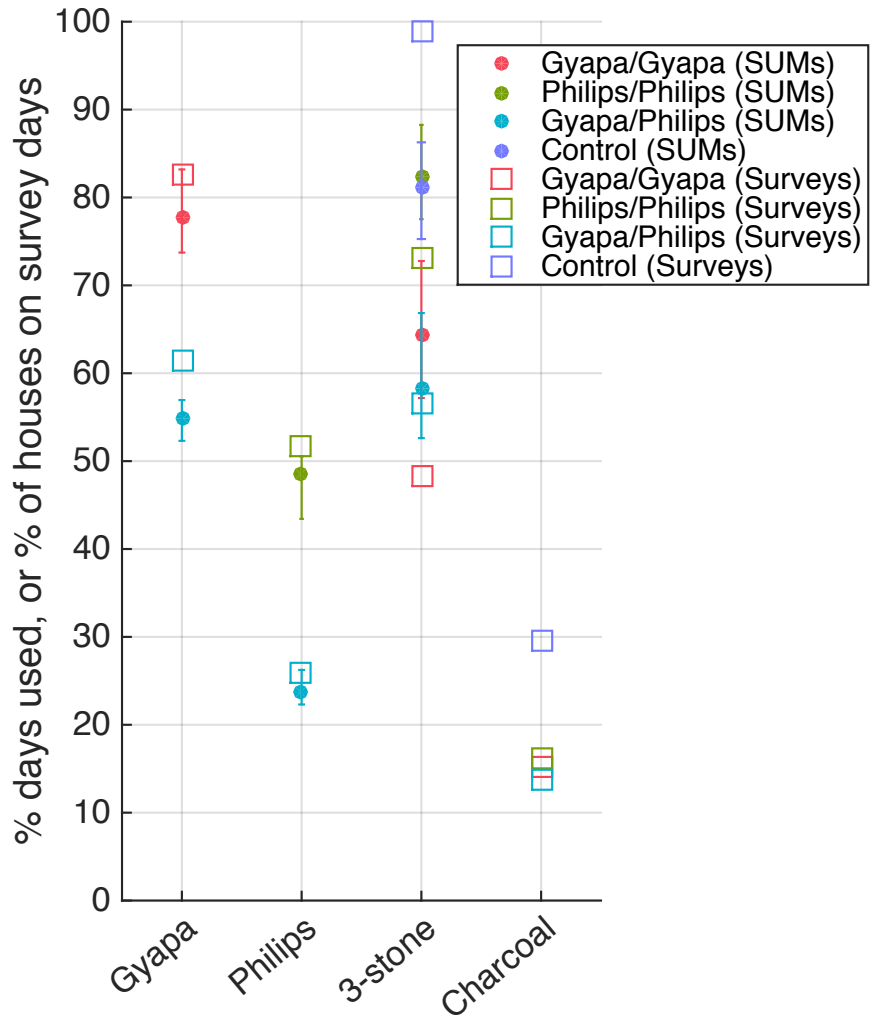

Figure 2-4. Use of different stove types among households in different intervention groups, as measured by SUMs and surveys. Figure shows aggregate results using all data available after stove deployment (Nov '13-Jan '15).

2.4.2. Traditional stove replacement

A primary objective of this and other cookstove interventions is to produce a reduction in use of traditional stoves. Thus, comparing use of TSFs across groups is a key outcome

measure. Unfortunately, the combination of SUMs event detection uncertainty and possible reporting biases leads to difficulty making strong conclusions when it comes to TSF and charcoal stove use. Most puzzling is that while the control group households reported using their TSFs on 98.9% of days measured, SUMs detected TSF cooking events in these households on just 81.3% of days. In this case, the SUMs estimate seems implausibly low for three reasons: 1) these households rely on traditional stoves for all their cooking needs, 2) the control group is expected to have little motivation for overreporting their stove use, and 3) additional TSFs may be in use while not being monitored. Thus, it is likely that the discrepancy between the SUMs and survey results is due to the challenges of measuring TSFs with SUMs.

To add complexity, reporting biases (specifically, underreporting) are expected to be a factor in survey-based measurements of traditional stove use for the other three groups, such that in these groups it is likely that both types of measurement are low. The direction of the survey-SUMs discrepancies is consistent with under-reporting TSF use, though the magnitude of the difference between surveys and SUMs varies from 1.1-15.8%. Surveys and SUMs were in the closest agreement for the Gyapa/Philips group, with 57.7% of days used for the SUMs, and 56.6% of days used according to surveys. The Philips/Philips group had the second best agreement between methods, with 81.5% of days (SUMs) and 73.0% (surveys), while the Gyapa/Gyapa daily use rates were 64.5% (SUMs) and 48.7% (surveys).

Use of TSFs remained high among all three of our stove intervention groups, and was consistently higher in the Philips/Philips group compared with the two groups that received at least one Gyapa stove. Regarding the question of which of the Gyapa groups had the lowest rate of TSF use, surveys and SUMs disagree: reported use is lowest in the Gyapa/Gyapa group, but SUMs-measured use is lowest for the Gyapa/Philips group. Unfortunately, without further assumptions, it is unclear which of these conclusions is correct.

SUM data are extremely limited for the charcoal stove category, so we must rely entirely on survey data. These data showed a strong reduction in charcoal stove use in the intervention groups relative to the control group, with 29.6% of control group homes reporting use ‘yesterday’ compared to 16.3% of intervention group households’.

For a subset of cases, we have both SUMs and survey measurements for the same households on the same days, allowing us to directly observe the method agreement rate for measured vs. reported stove use in each stove group and stove type. Specifically, we compared stove use measured by SUMs 'yesterday', the day before the survey, with survey responses on stove use 'yesterday'. We found moderate agreement on stove use 'yesterday', with error likely due to both misreporting and SUM uncertainty. Method percent agreement was highest for detection of TSF stove use (81.4%, n = 43), and Gyapa stove use (62.1%, n = 58). Method percent agreement here is relative to survey results; in other words, the percent of SUM results in agreement with the survey results. The lowest agreement was when the survey indicated that the TSF was not used on the survey day (25.0%, n = 44). Agreement between all SUMs and surveys declined slightly over the three survey periods, possibly indicative of SUM attrition. There was a higher rate of SUM agreement with survey data for cooking-events than non-events – i.e., when the respondent reported that they used a particular stove yesterday (Figure shown in SI). The fact that we see lower agreement on a case-by-case basis compared to the relatively high agreement between the aggregate measures of use presented in Figure 2-4 is likely due to the low sample sizes for these case-by-case comparisons and the fact that there is measurement error in both data sources (reporting bias on the part of the respondents, and a systematic underestimation of cooking events by the SUMs).

We also analyzed reactivity to enumerator visits in terms of both total number of SUM-detected cooking events with each stove, and number of SUM-detected days each stove was used. Here, reactivity was measured by comparing those metrics in a three-day window before the visit was scheduled, with a three-day window lagged at various intervals post-visit (the 3-day windows of post-visit analysis started 1, 4, 7, and 10 days after the visit). We found subtle increases in intervention stove use for both metrics, with concomitant decreases in TSF use in the windows after the visits, appearing to stabilize in the 7-10 day windows after the visit (Figure shown in the Supplementary Information), indicating that there may have been a subtle enumerator reactivity effect in our study. Further analysis enumerator reactivity, and reactivity due to the knowledge of being monitored by SUMs will be assessed in future work.

2.4.3. Stove stacking

Figure 2-5 shows the percent of days (as measured by both surveys and SUMs) on which households in different intervention groups used any of the 10 possible combinations of two types of stoves among the Gyapa, Philips, TSF, and charcoal stoves. Use of two Gyapas in the Gyapa/Gyapa group occurred more frequently (28.8% and 25.0% for SUMs and surveys) than use of a Gyapa and Philips in the Philips/Gyapa group (20.2% and 14.3% for SUMs and surveys) or use of two Philips in the Philips/Philips group (14.4% and 8.4% for SUMs and surveys).

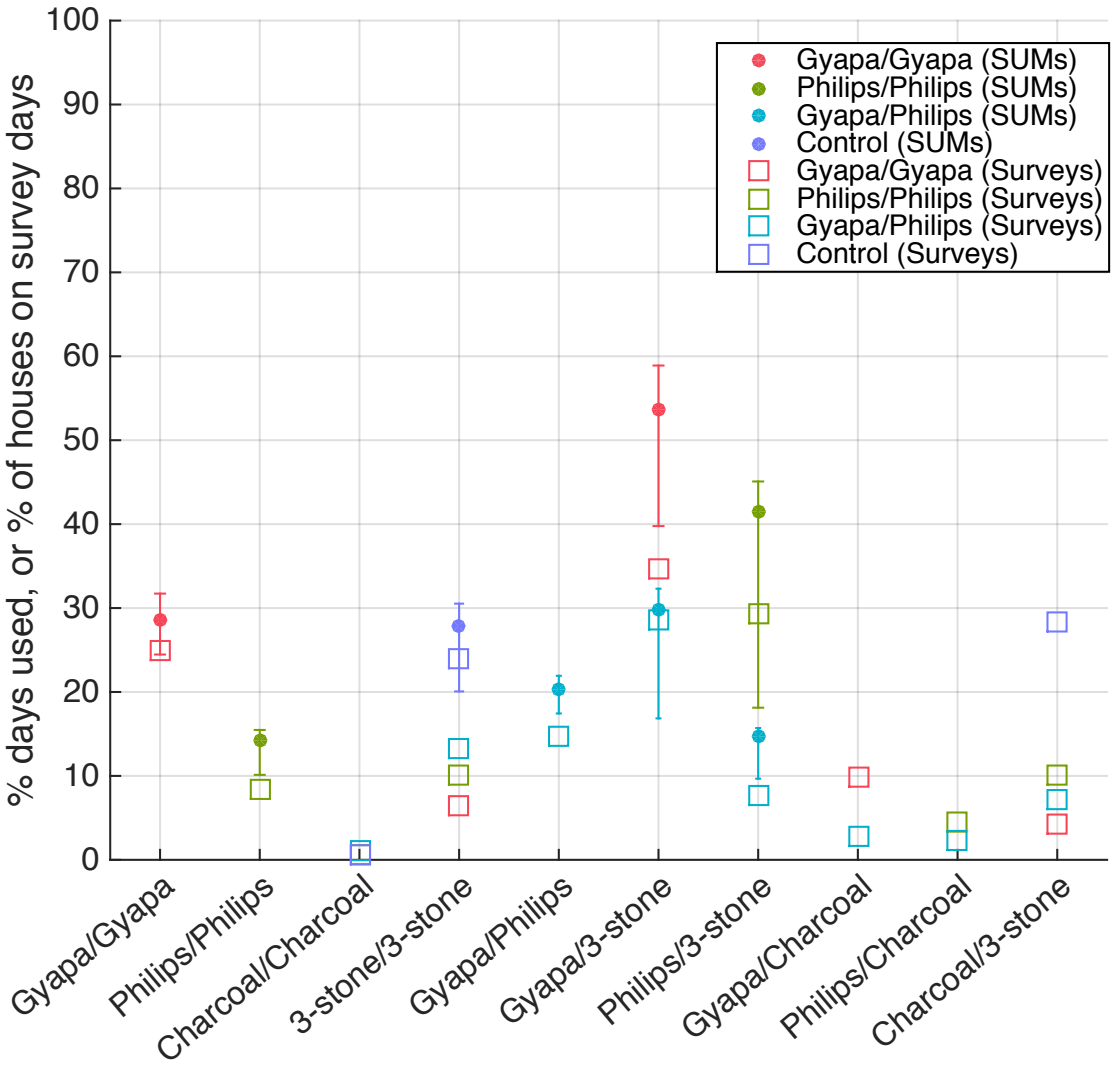

Figure 2-5 Use of different stove combinations among households in different intervention groups, as measured by SUMs and surveys.

Stacking of the Gyapa or Philips stoves with TSFs is much higher from SUM data than survey in the Gyapa/Gyapa (19.6% higher) and Philips/Philips group (12.4% higher). The previously mentioned sources of uncertainty may account for this, and it should be noted that

the estimated confidence intervals on both groups are large due to the low SUM coverage on these groups. We also find a consistently higher use rate from the SUM data than from surveys, which could be due to the sources of our uncertainties. For example, certain climatic conditions could produce false positives in multiple stoves at a household, or alternatively, co-heating could raise the temperature of multiple SUMs in a cooking area, when not all stoves with SUMs are actually used. Again, due to low coverage or failures of SUMs on TSF and charcoal stoves, we rely on survey results for indication of stacking with charcoal stoves. We found substantially similar results for the three non-control intervention groups, with lower stacking rates of multiple traditional stoves than the control group. Less than 13% of homes from those three intervention groups reported stacking multiple TSFs 'yesterday', while 24% did for the control group. Stacking of a charcoal stove and TSF was reported as less than 10.1% of days for all intervention groups, and 28.4% for the control group. Stacking of two charcoal stoves was very uncommon for all groups, less than 1.5% of days. Similarly, stacking of intervention stoves with charcoal stoves was low, with all groups reporting fewer than 9.8% of days used.

This can be contrasted with the work by Loo et al. (2016), who found that users in Kenya preferred the Philips stove among the six ICSs tested, while Lozier et al. (2016) found that even with the preferred Philips stove, it was only used exclusively on 24% days, while traditional methods were used exclusively on 25% of days, and stacking occurred on 45% of days.

2.4.4. Temporal stove usage trends

In the ideal scenario of perfect accuracy from the SUMs and from the surveys, the key benefit of SUMs data is higher temporal resolution. Figure 2-6 shows a time series of stove use for each type of stove, for the daily SUM data (smoothed with a spline and with shaded area 95% CIs) and for the quarterly survey data (dots). The general trends are in good agreement, except for the charcoal stoves, where the SUM data are extremely sparse. Interestingly, the use of most stoves appeared to be in phases, with strong continued use of a stove for a period of time, followed by very little use. There were no obvious patterns or periodicity to this, and may simply have followed each home's fuel collection. There are also large fluctuations in usage for the Gyapa and TSFs from October to December 2014, which partially occur in

between survey periods, and during the end of the rainy season when harvesting typically occurs, which could impact fuel use. The intervention did not introduce any new fuel types to the homes, and homes appear to have continued using wood, charcoal, and millet stalks primarily. Woods commonly used for cooking include Neem, Shea, Mango, and varieties locally known as Zanka, Sesibe, and others. Millet stalks, shown in Figure 2-2, are usually 1-3 cm diameter, often up to 8 feet in length, collected from agricultural byproducts. They have high lignin content, and a higher heating value of 18.05 MJ/kg, similar to agri-wastes like wheat straw and bagasse (Nhuchhen and Salam, 2012). Survey results indicate that the Philips stoves were used with charcoal about 1/3 of the time, with wood making up the remaining fraction. The Gyapa stove has reportedly been used with wood 80% of the time, with millet stalks making up the rest. More detailed analysis of fuels and fuel stacking, a critical component of health and environmental impacts (Masera et al., 2015), will be undertaken when surveying is completed.

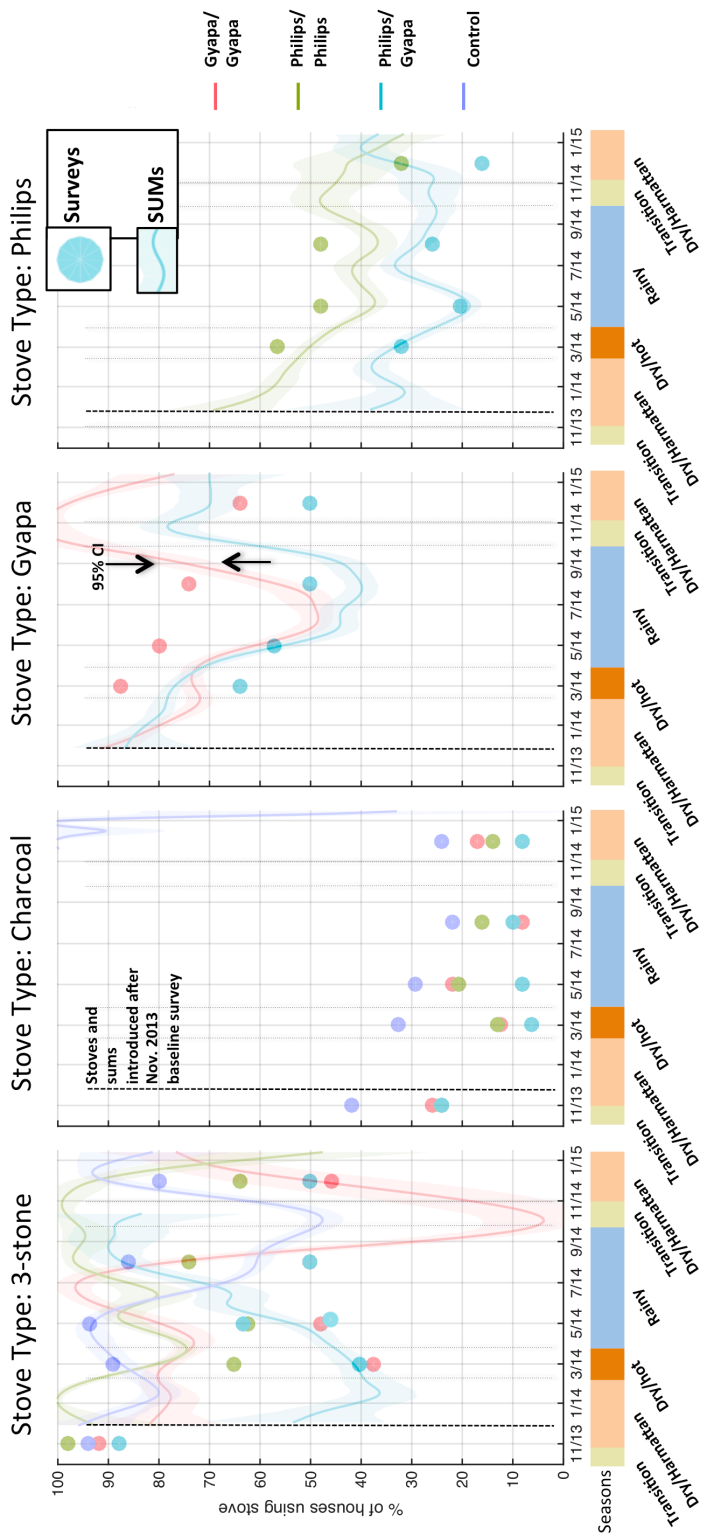

Figure 2-6. Survey and SUM time series of use for the different stove types, by intervention group.

There was no clear initial adjustment period for the intervention stoves, as seen in other studies, but that may have been due to the relatively low sample size for each group and the difficulty of removing seasonal variations in a one-year data set. This will be analyzed formally with the complete two-year data set later, but the survey data do show a lower TSF use rate for the Gyapa/Gyapa and Philips/Philips groups, followed by sustained use on about 50% of days on subsequent surveys. We observed decreasing use of all Gyapa and Philips stove categories over the year, though we have not yet quantified the trend. Charcoal stove use also appeared to decrease over the course of the study, except for the Philips/Gyapa group, though seasonal and adjustment periods may be driving their use as well.

There was no significant day of week trend for cooking with any of the stoves. This was likely because the main temporal periodicity for such rural households only is due to local markets, which cycle locations every 3 days. Cooking peak distributions (Figures in SI) show that the stove groups use different stoves at different times of day, further illustrating the reasons for stacking. The Gyapa stoves, for example, appear to be used preferentially later in the day compared to the others.

2.4.5. Daily stove use durations

Although estimation of cooking time from the SUM data has large uncertainties due to our experimental set up, we did observe it to be lower in our study compared with previous work undertaken in the Sissala district in Northwestern Ghana (Burwen and Levine, 2012). Our control group was found to cook with TSFs 4.6 h/day (we have insufficient data from charcoal stoves to measure cooking time), while in Sissala, the control group was found to cook an average of 10.7 h/day. Burwen and Levine (2012) also found substantial stacking of traditional methods with the brick and clay stove their study introduced. There, the treatment group cooked with the intervention stove 26% of the time, 2.5 hours/day with intervention stoves and 7.1 hours/day with traditional stoves. Stacking ratios were higher in our study, with the intervention stoves accounting for 61.5% of total cooking time for the Gyapa/Gyapa group, 59% for the Gyapa/Philips group, and 32% for the Philips/Philips group. We also found that when two identical stoves were provided, they were used nearly the same amount, likely to prolong

their lives or minimize wear. In the case of the Philips stove, this would also help mitigate downtime caused by the need to charge the stove with solar panels during daylight hours.

2.4.6. Links between stove type and type of food cooked

Figure 2-7 presents survey data on the dishes households in the different intervention groups reported cooking with each stove type during the Dec '14/Jan '15 survey round, approximately one year after receiving their new stoves. At baseline, we noted that TZ was cooked exclusively over TSFs, while vegetable soup and, to a lesser degree, rice, were also cooked over charcoal stoves (Table 2-1). As Figure 2-7 shows, this pattern persists in the control group households. However, there is variation in the extent to which new stoves replaced these existing technologies to cook these three dishes in the intervention groups. Across all groups, TZ continued to be cooked over TSFs in at least two thirds of cases in which this dish was cooked. Use of Gyapas to cook TZ was more common than use of Philips stoves for this dish. Specifically, in this survey round none of the households in the Gyapa/Philips group reported cooking TZ on a Philips stove. This was despite our effort to provide metal support stands for the Philips that we believed would help make them more stable for cooking dishes like TZ, which requires vigorous mixing (Dickinson et al., 2015).

Meanwhile, larger shares of the two other dishes were reportedly cooked over new stoves. Gyapas were used to prepare about 65% of vegetable soup dishes and 50% of rice dishes in the Gyapa/Gyapa group. The shares of these dishes cooked over Philips stoves in the Philips/Philips group are lower: about 40% for vegetable soup and 30% for rice. In the Gyapa/Philips group, Gyapas were used more frequently than Philips stoves to prepare both of these dishes; use of the two new stoves together comprised about 50% of vegetable soup cooking and 70% of rice cooking in these households. Overall, these results indicate that households found both types of new stoves better suited for cooking dishes like vegetable soup and rice than TZ, and even for these less involved dishes, a sizeable fraction of cooking continued to be done over TSFs and charcoal stoves. These usage patterns also suggest that cleaner cooking fuels like LPG or electric stoves could face similar barriers to use, and versatile and heavy-duty designs may be required to better replace traditional stoves and fuels.

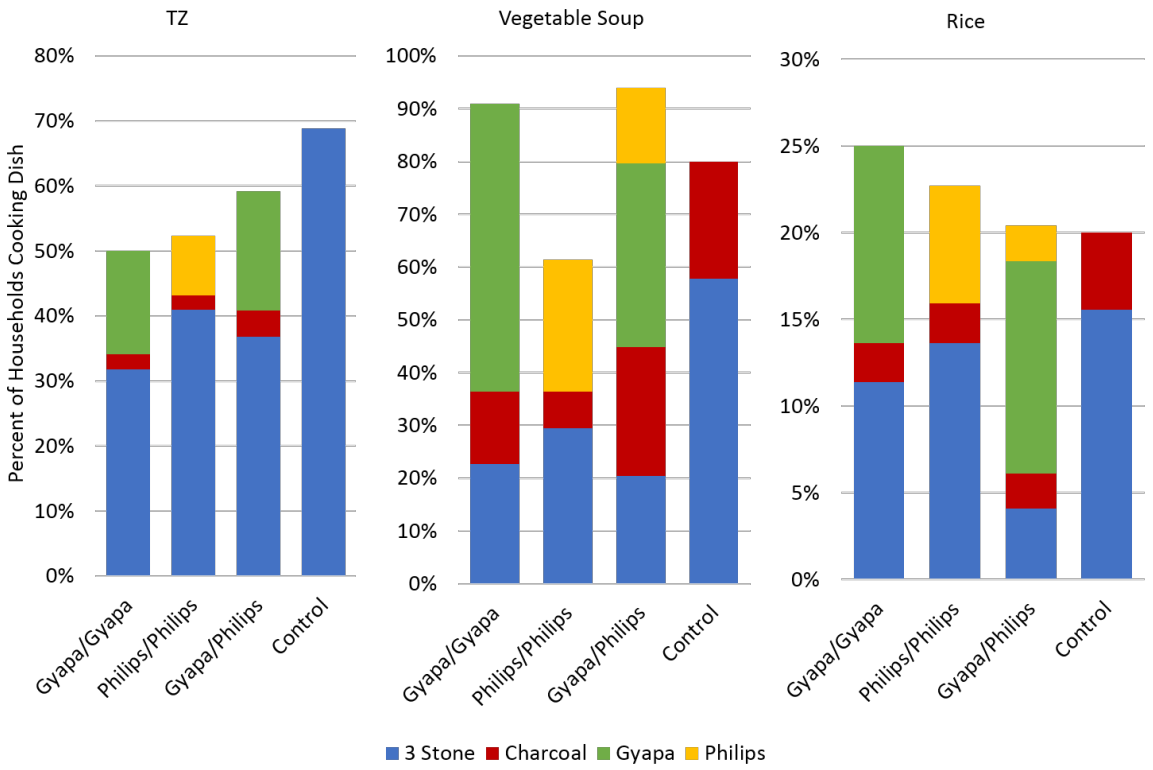

Figure 2-7. Percent of households cooking different dishes with each stove type, one year after intervention (Dec '14/Jan'15), by stove intervention group and type of stove.

2.5. Conclusions

SUM and survey data from a cookstove intervention study in Northern Ghana were combined to assess stove stacking and adoption behavior. Use of multiple methods has previously been identified as a valuable tool for technology and behavior adoption assessment (Stanistreet et al., 2015). Our study provides further evidence of this value. Each of the assessment methods used here faced certain challenges, and there were unexplained discrepancies in results between them. Despite the discrepancies, the two approaches tell a fairly consistent story about patterns of stove use in our different intervention groups. We observed generally high rates of use of the Gyapa stoves, and lower use rates for the Philips stoves. There was substantial continued use of traditional TSF and charcoal stoves among the three intervention groups, though their use was lower than for the control group. This continued reliance on TSF fires is problematic, as Johnson and Chiang (2015) found that use of a TSF for as little as 10 minutes per day was enough to surpass the WHO interim 1 24-hour $PM_{2.5}$

limit of 35μg/m^3 (WHO, 2006). This then suggests that we may not see substantial personal particulate matter exposure reductions with the stove use patterns observed in this study.
To effectively reduce use of TSFs and achieve health improvements, a better understanding of the drivers of traditional and improved stove use will be required. A mixed methods approach will likely be needed to examine these drivers. In our case, information from surveys on the types of foods cooked may help explain the continued reliance on TSF stoves. Specifically, it appears that neither of the new stove models was seen as an adequate replacement for TSFs for the task of cooking the staple porridge TZ. This thinking is brought into focus by Ruiz and Masera (2016), who note that households rely on traditional fires to conduct a wide variety of cooking tasks and meet a diverse set of needs. Generally, any one improved stove may be well suited to some of these tasks, but less so for others, such that stacking to meet household energy needs is inevitable, and potentially optimal if appropriate and efficient technologies are stacked.

Reporting bias is always a concern with survey-based measurement of technology use. However, for the new stove technologies, survey/SUM discrepancies in use rates are not as large in this study as in others. Specifically, our primary metric of interest, use-days, drew on households' reported use of each stove on the day prior to the survey, minimizing recall bias relative to longer measures (e.g., reported uses in the past week). For this primary metric, we find discrepancies between reported and SUMs-measured use of new stoves that range from 2.4% to 6.8%, suggesting that well-designed survey methods may be able to generate estimates of stove use that do not differ greatly from objectively measured results.
Finally, we find that more method validation data is needed to improve cooking event identification from the SUM data. Measurement methods for temperature monitoring of TSFs and frequently moved stoves are needed for more robust and reliable stove usage estimation. Thermocouple or infrared SUMs would provide more accurate results, and this option is becoming increasingly attractive. Thermocouple loggers are available for $25-100 per unit, now comparable to iButtons (Ruiz-Mercado et al., 2008) and Digit-TLs ($18-35 per unit, respectively). Other device manufacturers have also identified and filled this gap with custom configured thermocouples as with the Nexleaf Cookstove Usage Sensor

(http://nexleaf.org/technology/cookstove-usage-sensor), SWEETSense AIR (http://www.sweetsensors.com/applications/energy/), and Berkeley Air Monitoring Group K-SUM.

CHAPTER 3

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

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Abstract

REACCTING (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 ($p <= 0.01$). Both intervention groups given Philips stoves had significantly lower EC exposure than the control group (60.6% reduction, $p<=0.02$). 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 organic $PM_{2.5}$ 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 polyaromatic 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.

3.1. 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 2.5μm ($PM_{2.5}$) 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; Janssen et al., 2011). The steep $PM_{2.5}$ exposure-response curve at low exposures (Burnett et al., 2013) requires drastically cutting $PM_{2.5}$ exposure in order to achieve health benefits. In contexts where cooking-related emissions are a dominant source of $PM_{2.5}$ 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 $PM_{2.5}$ 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 REACCTING study (Research on Emissions Air Quality, Climate, and Cooking Technologies in Northern Ghana; Dickinson et al., 2015). Specifically, we present the results from carbonaceous $PM_{2.5}$ observations from personal, microenvironment, and ambient samples. We focus on EC and OC because they tend to constitute a substantial portion of total $PM_{2.5}$, 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 $PM_{2.5}$ from 191 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., 2012a; 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.

3.1.1. 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 $\mu g/m^3$ and average cooking area BC of 14.5 $\mu g/m^3$. 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 $\mu g/m^3$, 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 3-1). 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.

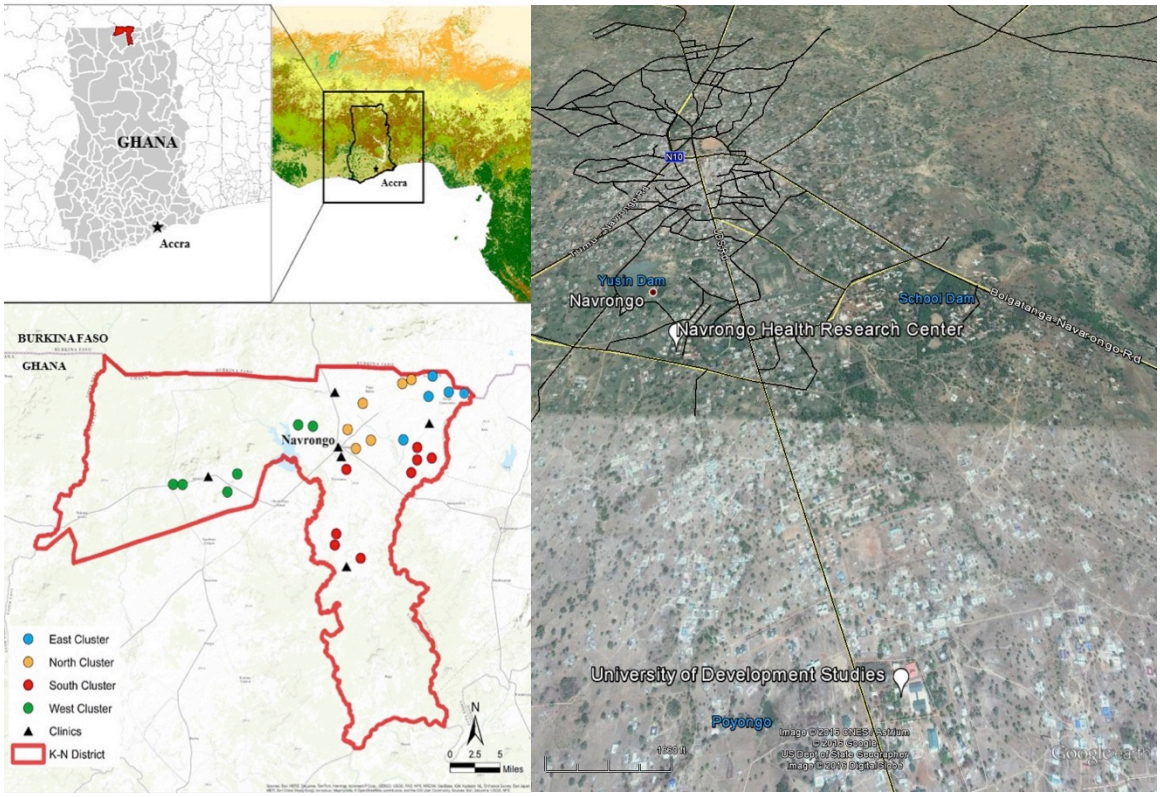

Figure 3-1. Map of the study region at various scales. At right, the town of Navrongo, with markers on the Navrongo Health Research Center, the base of operations and where ambient $PM_{2.5}$ was sampled (Google Maps, 2016).

3.2. 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 3-1) (Dickinson et al. 2015), 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.

3.2.1. 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 3-1. Household and seasonal statistics for the personal exposure (191, 88 unique individuals) and microenvironment samples (127).

		Personal Exposure Samples		Microenvironment Samples		Ambient Samples	
		All samples: 191 samples from 88 unique individuals Organics subset (in parentheses): 37 samples from 37 unique individuals		All samples: 127 samples from 37 unique households Organics subset (in parentheses): 16 samples from 16 unique households		All samples: 51 samples Organics subset (in parentheses): 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	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)

3.2.2. 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, 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 (Figure 3-2). 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 3-2. Ambient sampling inlet shown at the Navrongo Health Research Center. On the right, typical cooking areas, shown with the U-pod air quality samplers.

3.2.3. 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 (Figure 3-2). 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.

3.2.4. 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 cm^2 punch was taken from each quartz fiber filter and analyzed using a Sunset Laboratory EC/OC analyzer operated under NIOSH 5040 thermal optical transmission (TOT) method (Birch, 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.

3.2.5. 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.

3.2.6. 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:

$$ln(Y_{ij}) \sim \beta_0 + \beta_1(stovegroup)_i + \beta_2(\#\ in\ family)_i + \beta_3(gender*age)_i + \beta_4(season)_i + \beta_5(SES)_i + \beta_6(primary\ cook)_i + \alpha_j + e_{ij} \quad \text{Equation 3-1}$$

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 $PM_{2.5}$ 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 3-2). 87 unique household EC and OC samples were available for analysis.

$$ln(Y_{ij}) \sim \beta_0 + \beta_1(stovegroup)_i + \beta_2(\#\ in\ family)_i + \beta_3(season)_i + \beta_4(SES)_i + \alpha_j + e_{ij} \quad \text{Equation 3-2}$$

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 3-3) 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.

$$ln(Y_{ij}) \sim \beta_0 + \beta_1 ln(MicroEnv)_i * \beta_2(stovegroup)_i + \beta_3(\#\ in\ family)_i + \beta_4(season)_i + \beta_5(SES)_i + \beta_6(primary\ cook)_i + \alpha_j + e_{ij} \quad \text{Equation 3-3}$$

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

3.3. Results and discussion

3.3.1. Personal carbonaceous $PM_{2.5}$

Summary statistics for the EC, OC, and organics are shown separately for personal and microenvironmental samples in Table 3-2. Mean personal OC concentration was 38.9 µg/m3 (±54.3 µg/m^3), while mean EC was 2.8 µg/m^3 (±10.5 µg/m^3). 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/m^3. 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 3-3.

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/m^3 (±33.5 µg/m^3). 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/m^3 for the control group, and 102 µg/m^3 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/m^3 in winter, and 55 µg/m^3 in summer. Dionisio et al. (2012a) measured integrated $PM_{2.5}$ on 31 children in The Gambia, reporting average 48-h exposures of 65 µg/m^3.

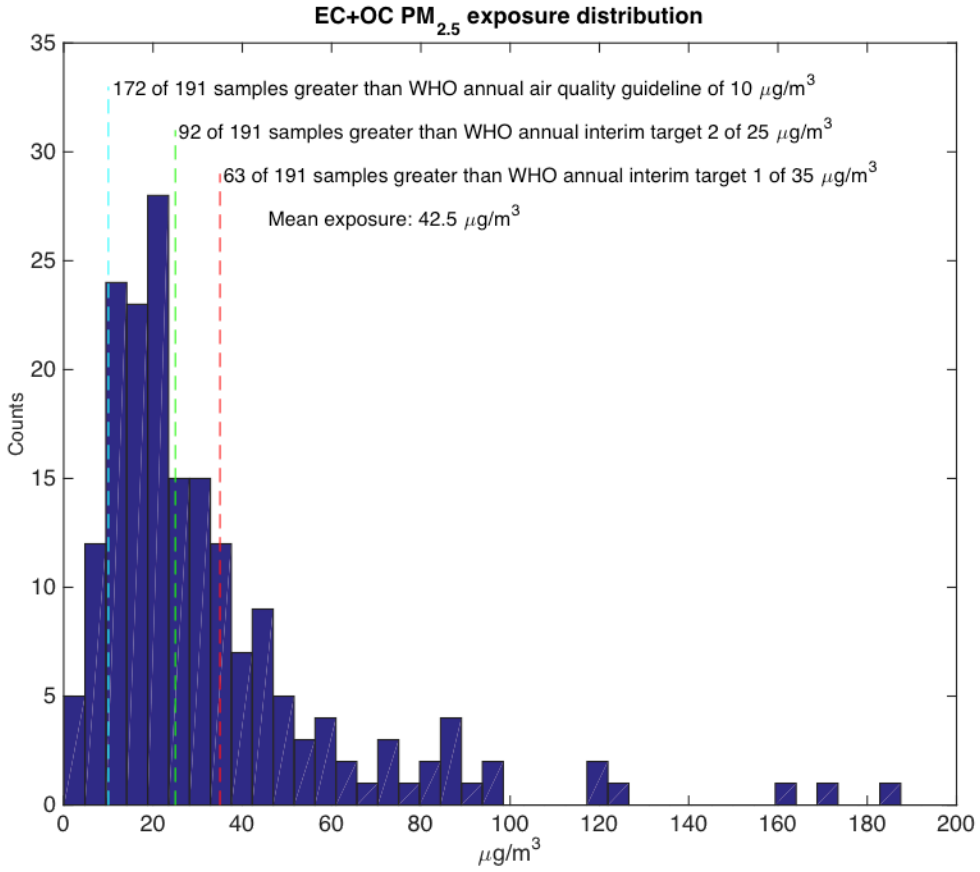

Figure 3-3. Personal exposure distributions for summed EC and OC, compared with relevant WHO standards.

Table 3-2. Summary of personal carbonaceous PM2.5 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/m3)	39.7	23.1	54.5	24.3	0.8	66
EC (µg/m3)	2.8	1.1	10.6	1.0	5.4	66
EC/OC	0.12.8	0.01.1	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/m3)	108.0	54.2	117.9	61.2	0.0	10
EC (µg/m3)	8.3	3.4	12.5	3.3	5.1	10
EC/OC	0.12.8	0.01.1	10.5	1.0	5.4	66

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

*units in ng/m3 unless noted	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
syringealdehyde	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

3.3.2. Personal OC $PM_{2.5}$ modeling results

Results from Equation 3-1 showed significant reductions in personal $PM_{2.5}$ OC exposure for the three intervention groups over the control group (Table 3-3). Full model output is presented in Appendix 2-2). Average OC exposures were 57.3% lower for the Gyapa/Gyapa group (27.8 μg/m^3, 95% confidence interval (15.2, 50.9), p = 0.01), 49.4% lower for the Philips/Philips group (32.9 μg/m^3, (19.6, 55.2), p = 0.01), and 63.2% lower for the Gyapa/Philips group (23.9 μg/m^3, (12.0, 47.7), p < 0.01) relative to the control groups (65.0 μg/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 intercept by individual was significant at 95%, with between-participant variance of 0.86, while the within-participant variance was 0.50.

Table 3-3. 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 (Appendix 2, Figure A2-1), 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.

3.3.3. Personal EC $PM_{2.5}$

Average exposures for $PM_{2.5}$ EC (Equation 3-1) were 44.3% lower for the Gyapa/Gyapa group (1.4 μg/m^3 (0.7, 2.6), p = 0.07), 62.0% lower for Philips/Philips group (1.0 μg/m^3 (0.6, 1.6),

$p < 0.01$), and 59.3% lower for the Philips/Gyapa group (1.0 $\mu g/m^3$ (0.5, 2.1), $p = 0.02$), relative to the control group (2.5 $\mu g/m^3$ (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 3-3, Appendix 2).

Van Vliet et al. (2013) reported average personal exposure to black carbon (BC) of 8.8 $\mu g/m^3$ (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).

3.3.4. Microenvironmental carbonaceous $PM_{2.5}$

Average cooking-area microenvironment $PM_{2.5}$ OC using Equation 3-2 was 68.6% lower for the Gyapa/Gyapa group (33.3 μg/m^3 (16.7, 66.4), $p < 0.01$), 45.5% lower for Philips/Philips group (49.6 μg/m^3 (25.3, 97.4), $p = 0.04$), and 40.4% lower for the Philips/Gyapa group (57.3 μg/m^3 (25.2, 130.7), $p = 0.18$), relative to the control group (100.2 μg/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 μg/m^3 (1.9, 9.4), $p = 0.28$), Philips/Philips (3.3 μg/m^3 (1.5, 7.2), $p = 0.09$), and Gyapa/Philips (5.1 μg/m^3 (2.0, 13.0), $p = 0.6$) groups relative to the control group (6.5 μg/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., 2017 *in preparation*).

The control group results (6.5 μg/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 μg/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 μg/m^3 before the introduction of the Gyapa, and 320 μg/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 $\mu g/m^3$ for the 'poorer' neighborhoods, to 25-33 $\mu g/m^3$ for the 'affluent' neighborhoods, similar to observations presented here.

3.3.5. 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 3-3). 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 3.3.2, with lower expected exposures in the light and heavy rainy seasons ($p = 0.32$, $p = 0.03$) (Appendix 2).

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 (Appendix 2). Van Vliet et al. (2013) reported a similar lack of relationship between cooking area and personal BC in Central Ghana.

3.3.6. Ambient carbonaceous $PM_{2.5}$

Average ambient $PM_{2.5}$ EC and OC concentrations were 0.3 $\mu g/m^3$ and 4.1 $\mu g/m^3$, respectively (Figure 3-4 and Appendix 2). These were lower than those measured in Navrongo by Ofosu et al. (2013), who reported 0.95 $\mu g/m^3$, and 11.36 $\mu g/m^3$. 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/m^3, substantially lower than that measured by the Ofosu study (mean total $PM_{2.5}$ mass concentration of 32.4 µg/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) (Figure 3-1).

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 $PM_{2.5}$ concentrations from 27-164 µg/m^3, and BC concentrations from 1.3-8.2 µg/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 $PM_{2.5}$ concentrations ranging from 63-104 µg/m^3 for the 'poorer' neighborhoods, to 21-55 µg/m^3 for the 'affluent' neighborhoods. They also found seasonal increases in regional and locally resuspended $PM_{2.5}$ and PM_{10}, which were attributed to the Harmattan winds. Van Vliet et al. (2013) reported average ambient $PM_{2.5}$ BC concentrations of 2.0 µg/m^3 (1.1, 2.9) (n = 9), and $PM_{2.5}$ mass concentrations of 20 µg/m^3 (12.1, 27.9).

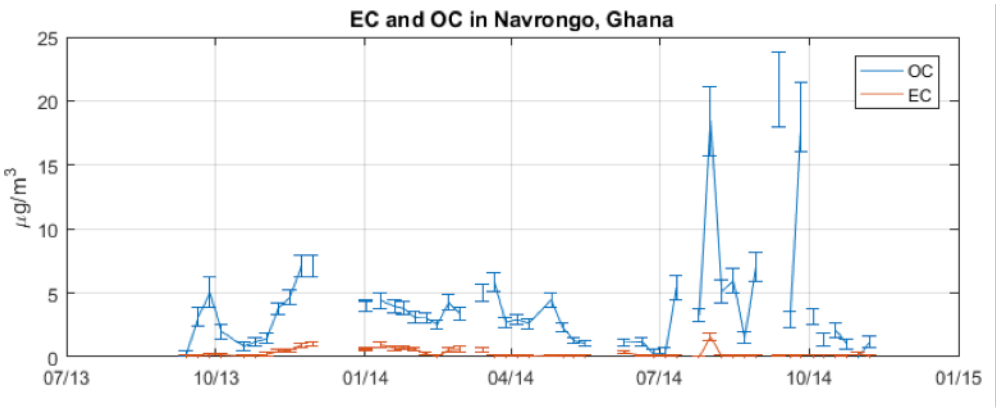

Figure 3-4. Time series of weekly EC and OC $PM_{2.5}$ samples collected in Navrongo from September 2013-November 2014, presented with pointwise uncertainty estimates.

Table 3-4. Summary of ambient carbonaceous PM2.5 EC, OC, and organics samples. Flagged samples were removed from analysis due to various possible issues including sampling duration and contamination.

All Ambient EC/OC Samples (n = 50)

	Mean	Median	Stdev	Geometric Mean	%BDL	
OC (µg/m^3)	4.1	3.2	4.3	2.8	2.0	
EC (µg/m^3)	0.3	0.1	0.3	0.1	47.4	
EC/OC %	6.5%					
PMF Ambient $PM_{2.5}$ Dataset (n = 25)						
*units in ng/m^3 unless noted	Mean	Median	Stdev	Geometric Mean	%BDL	S/N
OC (µg/m^3)	3.65	3.80	1.67	3.13	0.0	6.7
EC (µg/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

3.3.7. Personal $PM_{2.5}$ source apportionment results

A 6-factor PMF solution (Figure 3-5) 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 (Appendix 2). 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.

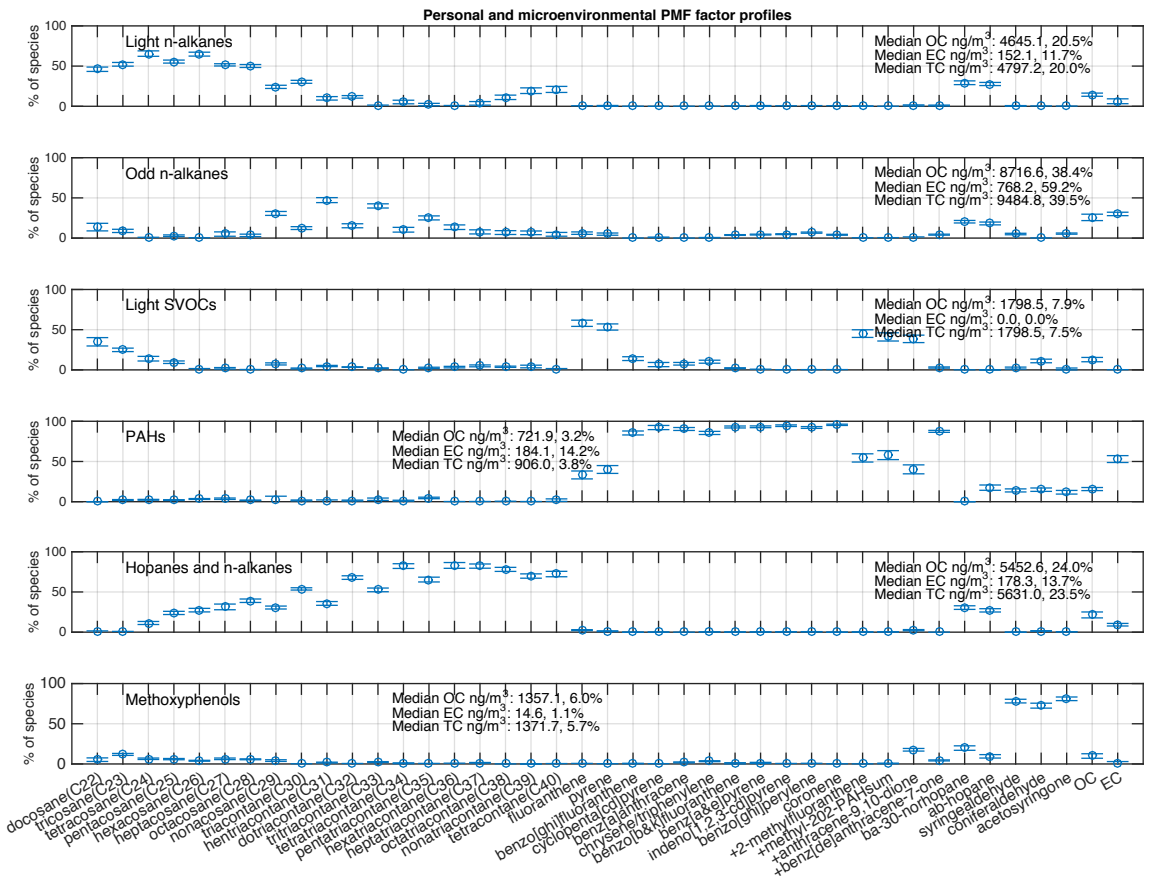

Figure 3-5. 6-factor PMF solution factor profiles for the personal and microenvironmental data, reported as percent of species, along with bootstrapped standard deviations. Mean contributions to OC and EC are shown for each factor in units of ng/m^3, and by the percentage of total apportioned.

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 (Rogge et al., 1993a). 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.

3.3.8. Factor enrichment in personal vs. cooking area samples

Comparing the factor contributions of the personal and cooking area microenvironment samples (Figure 3-6), 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 (Appendix 2 Table A2-1). 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.

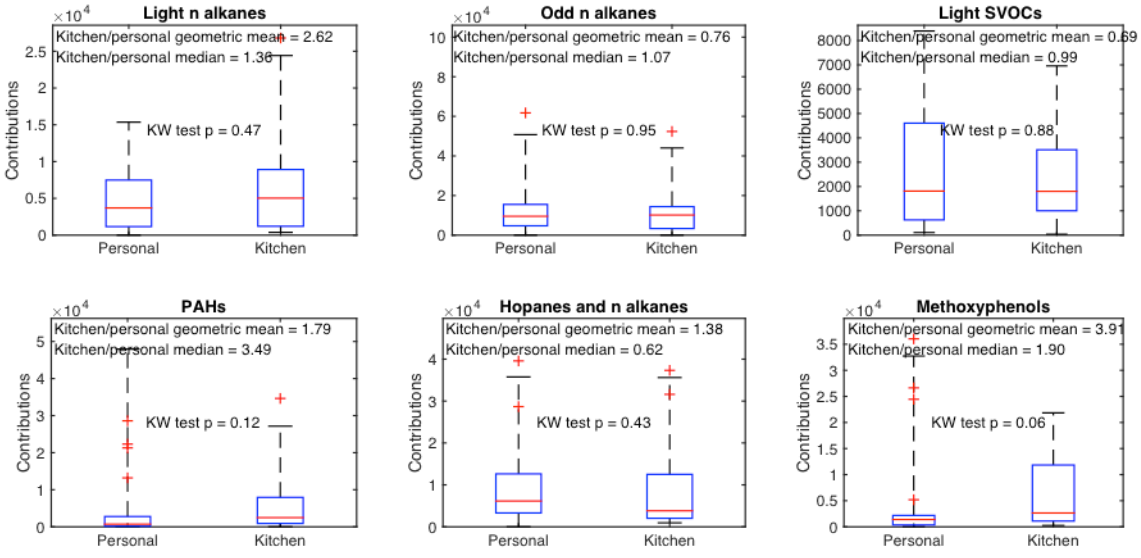

Figure 3-6 Comparisons of factor contributions to personal vs. cooking area microenvironment samples. P-values shown are from the Kruskal-Wallis non-parametric ANOVA test to compare distributions of two or more groups.

3.3.8.1. 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 (Equation 3-2) is again used, with fewer covariates than that specified in Equation 3-1 due to the smaller subset of samples available with speciated organics data (Table 3-4).

$$\ln(Y_{ij}) \sim \beta_0 + \beta_1(\text{stovegroup})_i + \beta_2(\text{family})_i + \beta_3(\text{gender*age})_i + \beta_4(\text{primary cook})_i + \alpha_j + e_{ij} \quad \text{Equation 3-4}$$

First, Equation 3-4 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 (Appendix 2). 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 3.3.1) 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 3-4 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 3-3). 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.

3.3.9. Ambient $PM_{2.5}$ source apportionment

Six PMF factors were identified for the ambient $PM_{2.5}$ data set (Table 2-4). These ambient factor profiles (Figure 3-7) were similar to those of the P-M profiles (Figure 3-5), 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 (Appendix 2, Figure A2-2).

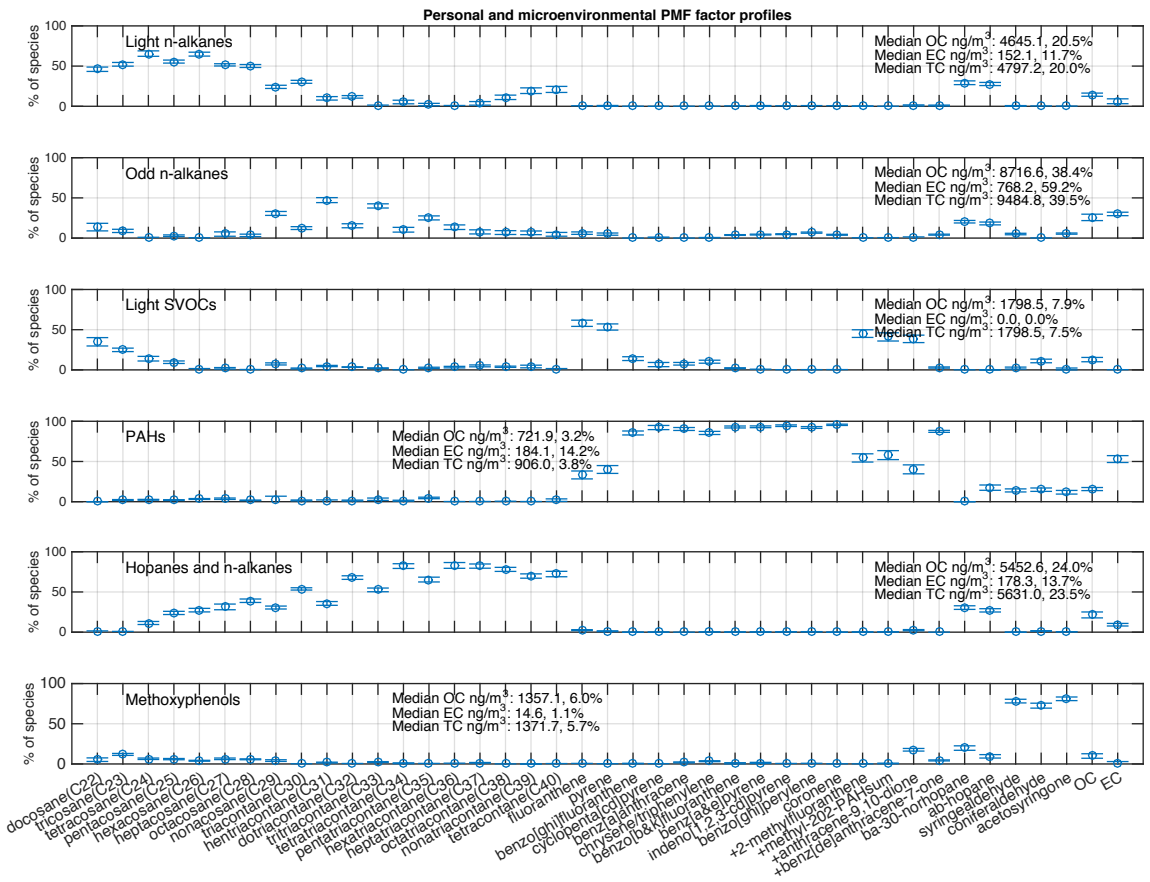

Figure 3-7 Personal and microenvironmental sample factor profiles for the 6-factor PMF solution.

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) 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 3-7). 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.

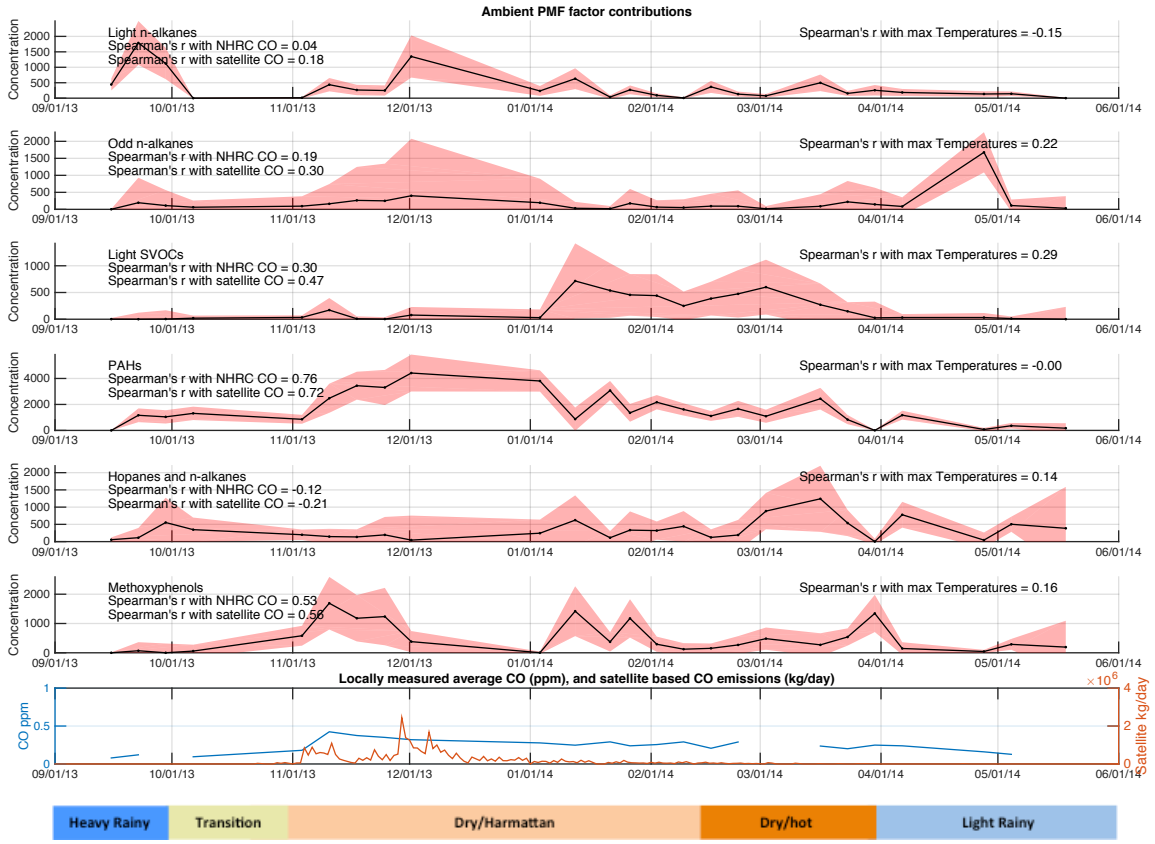

Figure 3-8. Ambient PMF factor contributions for weekly-integrated samples collected at the Navrongo Health Research Center. Red bands shown are standard deviations from the bootstrapped solution. Factor correlations with locally measured CO averages and satellite-measured CO emissions are shown, as well as the respective CO time series.

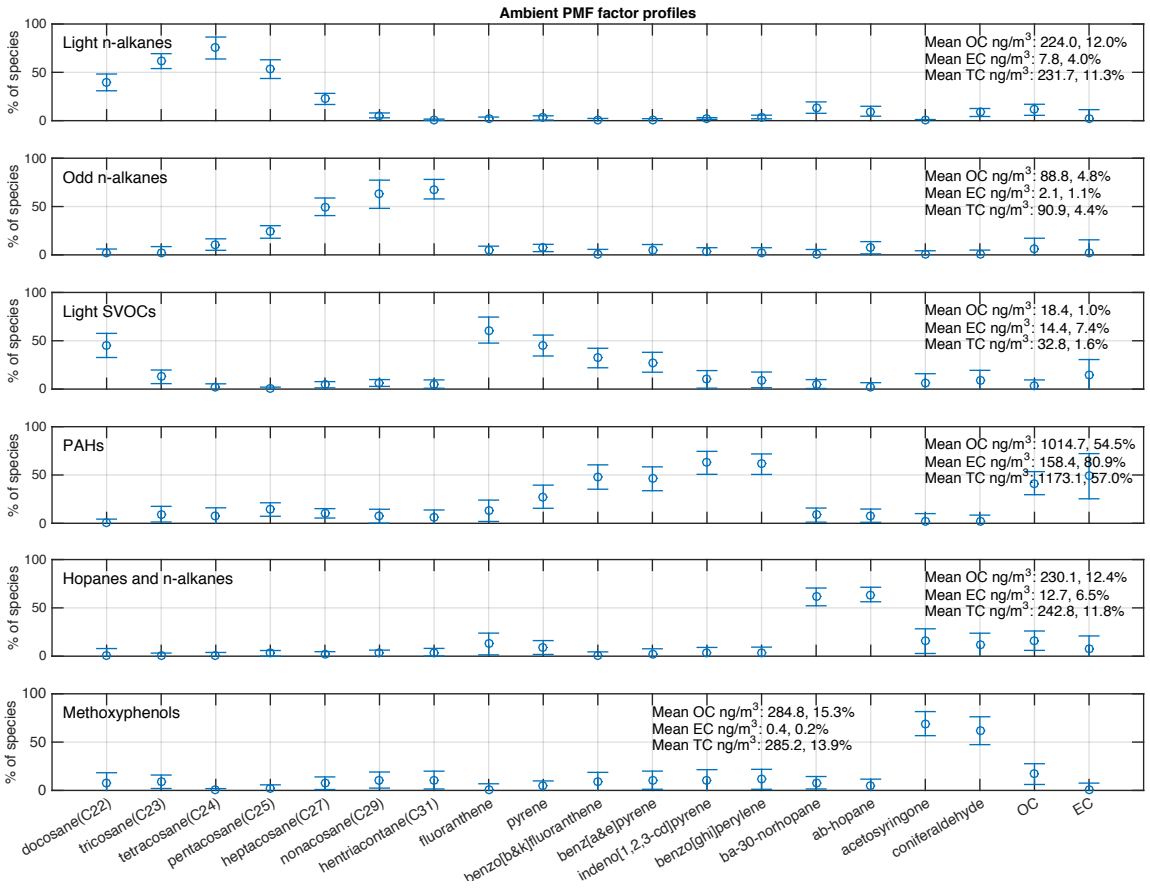

Figure 3-9 Ambient PMF 6-factor solution profiles for the weekly samples collected at the NHRC.

3.4. Comparison with past works and regional implications

The identified ambient source profiles can be somewhat reconciled with the pollution sources in Navrongo described by Ofosu 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 3-8). 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.

3.5. 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/m^3) were much higher than the concentrations of the ambient samples (4.4μg/m^3). 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 in cooking behaviors, cookstoves, fuels, and prevalence of non-cooking PM sources.

CHAPTER 4

Exposures to carbon monoxide in a cookstove intervention in Northern Ghana

Ricardo Piedrahita, Ernest Kanyomse, Evan Coffey, Yolanda Hagar, Christine Wiedinmyer, Katie Dickinson, Abraham Oduro, Michael Hannigan

Abstract

Burning biomass in traditional cookstoves for home energy use is a major health and environmental concern. In our study, two nominally cleaner burning and more efficient stoves were distributed over four study groups in rural Northern Ghana. To measure the effect of these stove interventions on personal pollutant exposures, participants periodically wore real-time carbon monoxide (CO) monitors. Intervention effects on CO exposure were assessed using mixed effects modeling. Relative to the control group, there was a 14.9% reduction in CO exposures in the group given two Philips forced draft stoves ($p = 0.40$), 5.6% reduction in the group given two Gyapa stoves (locally made rocket stoves) ($p = 0.78$), and 0.1% reduction in the group given one Gyapa and one Philips stove ($p = 1.00$). Overall, CO exposure for participants was low given the prevalence of cooking over 3-stone fires, with only 7.5% of daily samples exceeding WHO Interim-1 standards. For the wearable CO monitors used in this study, we present quantification methods and performance of duplicates. Lastly, we analyzed the relationship between personal carbonaceous $PM_{2.5}$ and CO exposure for the data set that included both measurements, finding that there is only a weak relationship, most likely due to the diversity of identified air pollution sources in the region and variability in behaviors.

4.1. Background and motivation

Carbon monoxide (CO) is commonly measured in cookstove and air pollution exposure studies due to its association with adverse health effects and birth outcomes such as low birth weight (Smith et al., 2000; Longo et al., 1977; Astrup, 1972), the low cost of real-time wearable monitors, logistical challenges of measuring other pollutants, and correlation with other co-emitted pollutants. These reasons, along with the well-established health effects associated

with $PM_{2.5}$ (e.g., Naeher et al., 2007), have led to the study and use of CO as a surrogate for $PM_{2.5}$. Previous works have found varied results for this relationship, with differences ascribed to variability in source types like fuels and stoves, behaviors, and home designs, among other characteristics (Naeher et al., 2001; Northcross et al., 2010; McCracken et al., 2013; Dionisio et al., 2012a; Carter et al., 2016 *in press*). Here, we add to the literature on this topic to further the understanding of source types for rural communities in Northern Ghana.

This work is part of the REACCTING study (Research of Emissions, Air quality, Climate, and Cooking Technologies in Northern Ghana), a 200 household cookstove intervention in the Kassena-Nankana districts of Northern Ghana from November 2013 to November 2015 (Dickinson et al. 2015). The 200 study households were divided into four groups, with one group given two locally made Gyapa rocket stoves, one group given two Philips HD4012 LS forced draft stoves, one group give one of each intervention stove, and the last group serving as control until the end of the study, when they were given their choice of two stoves. The study region is described by Oduro et al. (2012) and Dickinson et al. (2015). Piedrahita et al. (2017a) reports personal, cooking area, and ambient carbonaceous $PM_{2.5}$ results from the study. Here, we present personal CO exposure results over the complete study period, focusing on the effects of stove group and other covariates on exposure. Further, we also identify and characterize behavioral characteristics gleaned through the trends observed in the CO exposure daily time series.

4.2. Personal CO sampling methods

Personal CO exposure sampling was performed throughout the REACCTING study, from November 2013 through November 2015, with a total of 751 days of data collected (derived from 48-h samples) and passed through quality checks (Table 4-1, Appendix 3). Lascar USB-CO300 and CO1000 monitors were used to measure 1-minute CO concentrations and were typically worn by the mother and child of the household, as well as other available and consenting family members. Sampling was usually carried out from Monday-Wednesday, and then Wednesday-Friday, at four households during each period, one from each arm of the intervention study. Adult study participants typically wore the monitors either on neck lanyards, or in a waist pack when additional monitoring equipment was included, such as $PM_{2.5}$

filter samplers. Children carried the monitors in chest pockets of custom locally fabricated cotton t-shirts, or in backpacks when carrying other monitors. Field enumerators asked the participants to wear the Lascar monitors as much possible, and instructed them to leave them within arms-reach if they could not wear them or while they were sleeping.

Bluetooth Beacons were deployed during a subset of sampling periods to estimate distance between participants and cookstoves (Piedrahita et al., 2017c, *submitted*). The variability in the distance measurement was used to estimate compliance; no variability was assumed to indicate no motion of the Lascar USB-CO monitor and no motion means the monitoring devices were not being worn. Here, a participant was deemed compliant during time periods in which the standard deviation of the measured Bluetooth signal was greater than 2, on a rolling-hour basis, and during daytime hours (7:00-21:00). The threshold was calculated from the noise during calibrations over multiple distance points (Piedrahita et al., 2017c, *submitted*). The average measured rate of compliance for this data subset was 81.0%, and can be expected to be a lower bound for the sample as a whole because when the Beacons were deployed, they were part of the larger sampling pack, whereas most deployments had only the Lascar USB-CO monitors, which are less intrusive to wear.

Table 4-1. Sample statistics for the personal CO exposure deployments throughout REACCTING.

		Control	Gyapa/Philips	Philips/Philips	Gyapa/Gyapa
Sample overview	Days deployed that failed QA/QC and were removed	42	35	72	52
	Days deployed that passed QA/QC and were retained in analysis	160	136	157	149
	Mean sample duration in hours (stdev)	23.2 (1.3)	23.1 (1.3)	23.1 (1.4)	23.3 (1.1)
	Duplicate days deployed	36	21	48	45.0
	Unique participants	61	73	68	66
Gender and age covariates	Primary cook females >5y	110	94	96	113
	Non-primary cook females >5y	27	24	14	11
	Non-primary cook males >5y	16	24	20	10
	Children <5y	40	49	67	36
	Female over 5y (n, med, stdev, max, min)	137, 30.7, 17., 75.4, 5.	118, 34.4, 13.1, 63.4, 5.8	110, 31.4, 11.6, 53.4, 6.4	124, 34.4, 13.9, 63.4, 5.2
	Female under 5y (n, med, stdev, max, min)	23, 2.1, 1.3, 4.9, 1.2	18, 3.5, .7, 4., 2.4	47, 3.1, 1.1, 4.8, 1.2	25, 3., .8, 5., 2.1
	Male over 5y (n, med, stdev, max, min)	16, 8., 3.1, 13.8, 5.8	24, 14., 20.6, 61.4, 6.1	20, 7.3, 1.3, 11.6, 6.4	10, 6.9, 1.6, 10., 5.6
	Male under 5y (n, med,	17, 2.2, .9, 4.2, 1.3	31, 2.5, 1., 4.6, 1.4	20, 2.8, 1.2, 4.9, 1.1	11, 3.1, 1.3, 3.9, 1.1

	stdev, max, min)				
SES	**Poorest**	22	45	23	43
	Poorer	50	35	57	45
	Poor	39	46	43	28
	Less poor	58	27	33	13
	Least poor	24	38	41	41
Seasons	**Harmattan bush burning**	76	80	108	77
	Hot dry	35	38	30	28
	Light Rainy	18	25	21	31
	Heavy Rainy	58	38	36	26
	Transition	6	10	2	8

4.2.1. Calibration and data preparation methods

CO logger calibrations were performed as often as possible over the 2-year study period, averaging 1.5 calibrations per monitor (range 1 to 7, standard deviation = 1.5) and Pearson's R of 0.99±0.06 from linear calibration functions (Figure A3-1). In most cases monitors were calibrated with certified standards, and in a few, field normalization was performed using a reference monitor (Thermo 48C, Thermo Scientific). On a weekly basis, the loggers were co-located with the reference monitor at the Navrongo Health Research Centre (NHRC) for field normalization. However, the ambient concentrations were usually too low to generate a satisfactory calibration curve in the range desired for the personal exposure measurements.

201 sampling days (25.0%) were removed due to sensor degradation and electronics concerns. Paired CO Lascar monitors were successfully deployed on 150 days, with duplicates showing a Pearson's R of 0.81 for the calibrated and 0.92 for the un-calibrated daily average concentrations (Appendix 3). Lower correlation for the calibrated monitors can be attributed to the general observed trend of decreased sensitivity with Lascar age. When some of the monitors were calibrated and others were not, the improved data quality of the calibrated ones resulted in poorer correlation. As the monitors were randomly distributed among stove groups and individuals, there is minimal risk of systematic bias and the calibrated Lascars should provide more accurate results despite the lower correlation. Calibrations are also important to avoid bias related to the factory calibration of the monitors and to adjust for the Lascar change in sensitivity over time. When the monitors were first calibrated using a linear regression of the form $Lascar\ Signal = p_1 + p_2(Reference\ Concentration) + \varepsilon$, the average slope (sensitivity) was 1.06 ± 0.06 and intercept was 0.08 ± 0.13ppm. While this is reasonably accurate and consistent for the intended use of the monitors, some displayed up to 16% error out of the box, strongly

supporting the need for calibration. When paired monitors were deployed, and both were deemed free of failures, minute average concentrations from the two monitors were used in further analyses.

4.2.2. Personal exposure mixed effects model specification

Mixed effects modeling was performed to ascertain the intervention effects while accounting for repeated measures (Peretz et al., 2002; Burton et al., 1998), in an approach similar to Piedrahita et al. (2017a). A parsimonious model was developed in which daily average log-transformed CO exposure (ppm) was regressed against stove group, season, socio-economic status, and a categorical variable describing primary cook status with gender and age group (Equation 4-1).

$$Log(Personal\ CO_{ij}) = ß_0 + ß_1(Stove\ group)_i + ß_2(SES)_i + ß_3(Primary\ cook*Gender)_i + ß_4(Season)_{ij} + \alpha_j + e_{ij} \quad \text{Equation 4-1}$$

The equation above is slightly simplified, as $ß_2$ represents five categories for socioeconomic status (SES) for each household, 'poorest', 'very poor', 'poor', 'less poor', and 'least poor'. The SES categories were calculated as by Awini et al. (2010) using a principal components analysis of wealth indicators like access to electricity, sanitation, and water. $ß_3$ represents four categories, 'female primary cook' (all over 5 years old), 'female non-primary cook over five years old', 'male non-primary cook over 5 years old', and 'children under five years old', where primary household cook was identified in the baseline survey (Dickinson et al 2015), and there were no male primary cooks. $ß_4$ represents the season categories, defined as 'light rainy' (April-June), 'heavy rainy' (June-October), 'transition' (October), 'Harmattan' (November to mid-February), and 'hot dry' (mid-February to April). The random intercept α_j accounts for the correlation within subjects due to repeated measures and has variance representing the between subject variation, and e_{ij} represents the random error, with its variance representing the within-child variation. A small positive exposure concentration was added to all values so as to include exposures of zero ppm in the log-transformed model. A compound symmetry covariance structure was employed, using the assumption that any repeated measurements of participants had equal correlation regardless of the time between them (Peretz et al., 2002). The AIC and adjusted R^2 were used to select the model and assess the fit after the most relevant variables

were included (stove group, season, SES). Using these model selection methods, the number of family members and other age group variables were not found to have significant effects and did not improve the fit, so were excluded from the final model we present. Residual distributions approached normality, satisfying the assumptions required to employ this modeling technique. We applied Equation 1 separately to the calibrated and un-calibrated Lascar CO data sets in order to compare results and understand the importance of our calibrations over the study.

4.3. Results

4.3.1. Personal CO exposure results

The results from Equation 4-1 are presented here in terms of percent change relative to the reference categories built into the model, as well as expected concentration values (geometric means) at those categories (Table 4-2). The reference for Equation 4-1 were those measurements taken from primary cooks (all female) in the control stove group also in the `poorest' SES category during the Harmattan season. The reference group experienced a daily CO exposure concentration expected value of 0.61 ppm (95% CI 0.38 – 0.95). By holding all variables constant except for stove group, the effect of stove group is isolated relative to the control group. Predicted CO exposure of the Gyapa/Gyapa group was 5.6% lower than control group (0.57 ppm (0.38-0.95), p = 0.78), the Philips/Philips group was 14.9% lower (0.52 ppm (0.35-0.75), p = 0.40), and the Philips/Gyapa group was 0.08% lower (0.60 ppm (0.41-0.89), p = 1.00). Males and females listed as non-primary cooks experienced 33.3% (p = 0.08) and 17.1% (p = 0.42) lower average CO exposures than female primary cooks, respectively. Children experienced 35.1% lower exposures than female primary cooks (p = 0.01). The 'light rainy' and 'heavy rainy' seasons had 49.3% and 42.9% higher expected exposures than the 'Harmattan' (p = 0.03, p = 0.02), while the 'hot dry' season was 31.5% lower (p = 0.02) than the 'Harmattan' season. Relative to the 'poorest' SES group, on average the 'poorer', 'poor', 'less poor', and 'least poor' groups experienced 12.7% 20.8%, 35.8%, and 38.9% lower exposure levels respectively (p = 0.53; 0.29; 0.06; 0.03). The between-subject variation was 0.68 (p < 0.01), and residual error variation was 1.46, giving an intra-class correlation coefficient of 0.32. Most of the unexplained variability observed in daily average CO exposures is thus due to day-to-day

differences within participants rather than between participants. Summary statistics grouped by the same model covariates as Equation 4-1 are presented in Appendix 3.

Table 4-2. Personal CO exposure using Equation 4-1. The reference group from the regression is the control group, for primary cook females, in the poorest SES group in the Harmattan season.

	Equation 1	% change from reference group	Expected exposure (ppm) (95% CI)	P value
Stove groups	Control	*Reference group*	.61 (.38, .95)	0.03
	Gyapa/Philips	-0.1	.6 (.41, .89)	1.00
	Philips/Philips	-14.9	.52 (.35, .75)	0.40
	Gyapa/Gyapa	-5.6	.57 (.38, .85)	0.78
Gender	Primary cook females >5y	*Reference group*		
	Non-primary cook females >5y	-33.3	.40 (.26, .63)	0.08
	Non-primary cook males >5y	-17.1	.50 (.32, .80)	0.42
	Children <5y	-35.1	.39 (.28, .55)	0.01
SES	Least poor	-38.9	.37 (.24, .58)	0.03
	Less poor	-35.8	.39 (.24, .62)	0.06
	Poor	-20.8	.48 (.31, .74)	0.29
	Poorer	-12.7	.53 (.35, .81)	0.53
	Poorest	*Reference group*		
Seasons	Harmattan bush burning	*Reference group*		
	Transition	6.4	.64 (.34, 1.22)	0.85
	Light rainy	49.3	.90 (.63, 1.29)	0.03
	Heavy rainy	42.9	.87 (.63, 1.18)	0.02
	Hot dry	-31.5	.41 (.30, .57)	0.02
		N	Random error variance	Adjusted R-squared
	Model fit statistics	751	.68 (.54, .87)	0.16

Applying Equation 4-1 to the un-calibrated exposure data indicated that the results were stable and consistent with the results presented in Table 4-1, though the 'light rainy' and 'heavy rainy' seasons shifted out of significance, and the effects of all stove groups showed reductions relative to the control group, though still not significant (Appendix 3.5).

4.3.2. Relationship between CO and carbonaceous $PM_{2.5}$

Linear regression modeling was employed to assess the relationship between the log of personal 48-h average EC and OC $PM_{2.5}$ concentration and the log of 48-h average CO concentration, with 108 personal exposure periods available for the comparison. Personal $PM_{2.5}$ EC and OC were analyzed using the NIOSH 5040 method (Birch, 2003), as described by Piedrahita et al. (2017a). A linear regression was employed to assess this relationship (Equation 4-2).

$$Log(Personal\ PM_{2.5})_i = ß_0 + ß_1*(log(CO))_i + e_i \quad \text{Equation 4-2}$$

The R^2 of Equation 4-2 was 0.012 for EC and 0.047 for OC (Figure 4-1; Appendix 3). The weak relationships are likely due to the variability in behaviors and the diversity of air pollution sources in the region (Ofosu et al., 2013; Piedrahita et al., 2017a). The stronger relationship with OC may be due to the higher co-emission of CO than with EC in a typical biomass fire (Andreae et al. 2001; Patterson et al. 1986; Akagi et al., 2011). Additionally, the later smoldering phase of biomass combustion may be a higher source of exposure than other combustion phases, due to behavioral factors. In the smoldering phase of biomass burning combustion, CO and OC are more abundantly co-emitted than with EC due to oxygen deficient combustion.

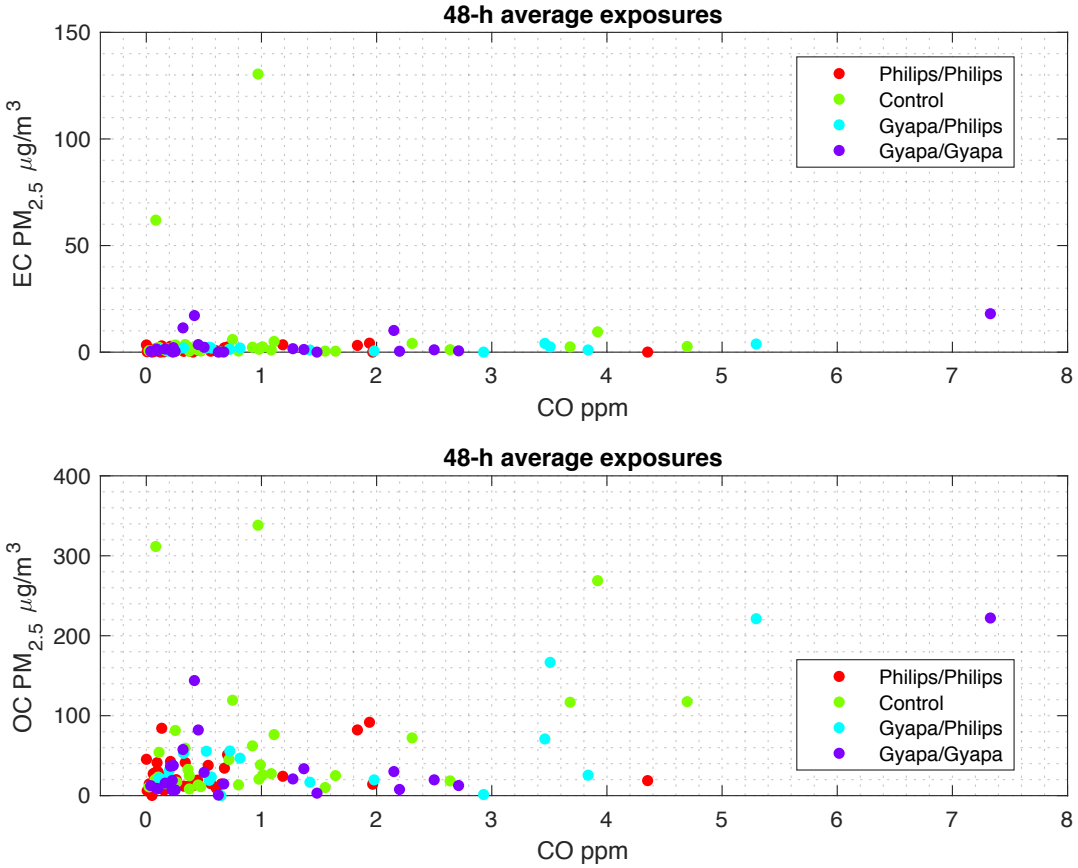

Figure 4-2. $PM_{2.5}$ EC and OC concentration vs. CO personal exposure concentration from the dataset with concurrent measurements. Note the low correlation between pollutants, even when accounting for stove group.

4.4. Discussion

4.4.1. Contributing factors to personal CO exposure

Results from Equation 4-1 showed reductions in CO exposure for two of the three intervention groups, though not substantial or statistically significant, suggesting some reduction in air pollution exposure for the Gyapa/Gyapa and Philips/Philips intervention groups. Reduction in exposure is supported by previous analysis from this study showing that carbonaceous $PM_{2.5}$ was lower for all intervention groups (Piedrahita et al., 2017a). In the $PM_{2.5}$ analysis, OC was found to be significantly lower in the three intervention groups by factors of 49.4% for the Philips/Philips group, 57.3% for the Gyapa/Gyapa group, and 63.2% for the Gyapa/Philips group. As noted in that work, a reduction in exposure was observed despite high continued use of traditional three stone fires (TSFs) in all the intervention groups (Piedrahita et al., 2016), especially the group given two Philips stoves, which here had the biggest decrease in exposure. The Gyapa/Gyapa and the Gyapa/Philips groups had the greatest replacement of TSFs, with TSF use reduced by about half of days cooked as measured with stove use monitors (SUMs) and household surveys.

Seasonal effects for CO exposure were not consistent with the EC and OC results in Piedrahita et al. (2017), but they may be explained by combustion processes. Compared to the 'Harmattan' season, CO exposures during the 'light rainy', and 'heavy rainy' seasons were higher by 49.3% ($p = 0.03$), and 42.9% ($p = 0.02$), while CO exposure during the 'hot dry' season was 31.5% ($p = 0.02$) lower, and there was an increase of 6.4% for the 'transition' season ($p = 0.93$). Lower carbonaceous $PM_{2.5}$ exposures were observed in all seasons relative to the 'Harmattan' season, but the biggest decreases in OC were during the 'light' and 'heavy' rainy seasons (Piedrahita et al., 2017a), in contrast to the results in the CO exposures. This disparity may be connected to the relationship between CO emissions with fuel moisture, with higher CO emissions at higher fuel moisture contents (Bhattacharya et al., 2002), whereas that trend does not appear to be shared with $PM_{2.5}$ at the fuel moisture levels observed here (L'Orange et al., 2012). Dionisio et al. (2012b) also found increased CO exposure during the rainy season in The Gambia, when measuring exposure for children under five years of age. Ambient differences in seasonal $PM_{2.5}$ may also play a role. The decrease in the 'hot dry' season observed in this study

may be due to changes in fuel choices and behaviors, as well as the relative importance of non-cooking ambient pollution sources, such as regional biomass burning that occurs during the Harmattan season (Piedrahita et al., 2017).

The observed exposures may have been affected by measurement error. The RMSE of daily-average duplicate Lascar CO monitors was 1.05 ppm, higher than the expected exposure of any stove group. Differences observed between stove groups may thus have been attenuated. However, the large quantity of samples collected improves the statistical power and allows the identification of differences. Confidence in the results would be bolstered by more frequent calibration and more precise instrumentation, which should be part of future work.

4.4.2. Observations from the real-time CO exposure time series

Temporal patterns from the minute-resolution CO data can illuminate behavioral patterns, and changes therein, due to the intervention.

Figure 4-3 shows CO exposures by time of day for each stove group smoothed using B-splines, which we use because of their efficient computation and calculation of confidence intervals (Eilers and Marx, 1996; Morel, 2016). All groups exhibited high exposure modes in the evening, peaking around 5:00-6:00 pm local time, while the Gyapa/Gyapa, Gyapa/Philips, and control groups had additional peak exposures at different times throughout the morning. The Gyapa/Gyapa group had the lowest morning time exposure, but also the highest evening exposure, while the Gyapa/Philips group had higher morning time peaks than evening peaks. The control and Philips/Philips groups were most balanced in this regard, with about equal CO exposures in the morning and evening. The differences in morning time exposure by stove group likely stem from changes in cooking behaviors, fuel type, or food preparation due to the stove intervention. The diurnal pattern is generally consistent with expected cooking patterns, although analysis of the SUMs showed the highest use for most stoves in the afternoons (Piedrahita et al., 2016).

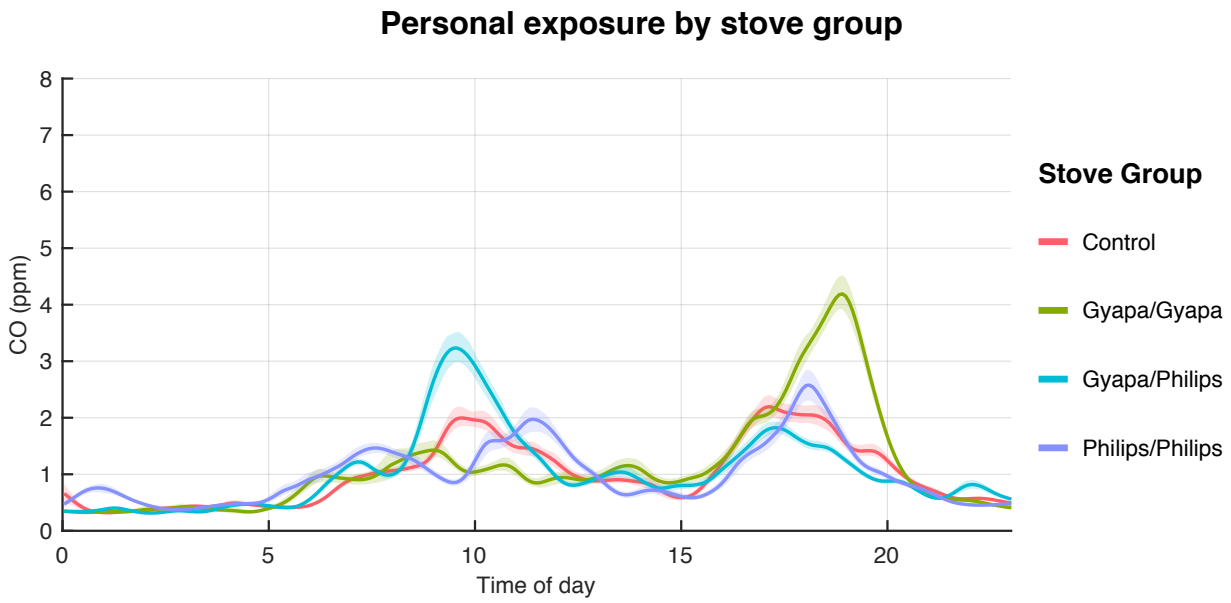

Figure 4-3. Spline-smoothed personal CO concentration by time of day and stove group. The shaded bands represent 95% confidence intervals on the mean estimates.

Aggregate, smoothed time series were also plotted specifically for the primary cooks and for the study participants by gender (Appendix 3). Primary cook females had high morning and evening peaks, with wider morning peaks than male participants, pointing to differences in exposure sources, with those male exposures possibly incurred when away from home, commuting or at work. Males under 5 had very similar exposure trends to the males over 5, and females under 5 had somewhat similar exposure trends to females over 5. These results suggest that gender roles are contributing to exposures as has been widely reported in other locations.

To further investigate personal exposure by stove group, we examined the time series of the CO concentrations measured in the cooking areas, which were available on a subset of the exposure monitoring periods (Figure 4-4). The cooking area CO concentration time series by stove group consistently showed morning and evening peaks, for all the stove groups, and largely matched the personal exposure trends for the earliest and latest peaks. The personal exposure trends showed more pronounced increases later in the morning, when the cooking area CO was not elevated, pointing to non-home CO exposure sources. Cooking area morning and afternoon CO peaks were also substantially higher than personal CO for all groups, especially for the control group, possibly indicative of changes in cooks' behaviors due to

available stove type. Seasonal differences in the diurnal trends in microenvironment CO concentrations were highly variable (Appendix 3), possibly due to lower data density for some of the study groups. This, along with stove use, warrant further exploration as they also hold information about fuel and behavior change with season.

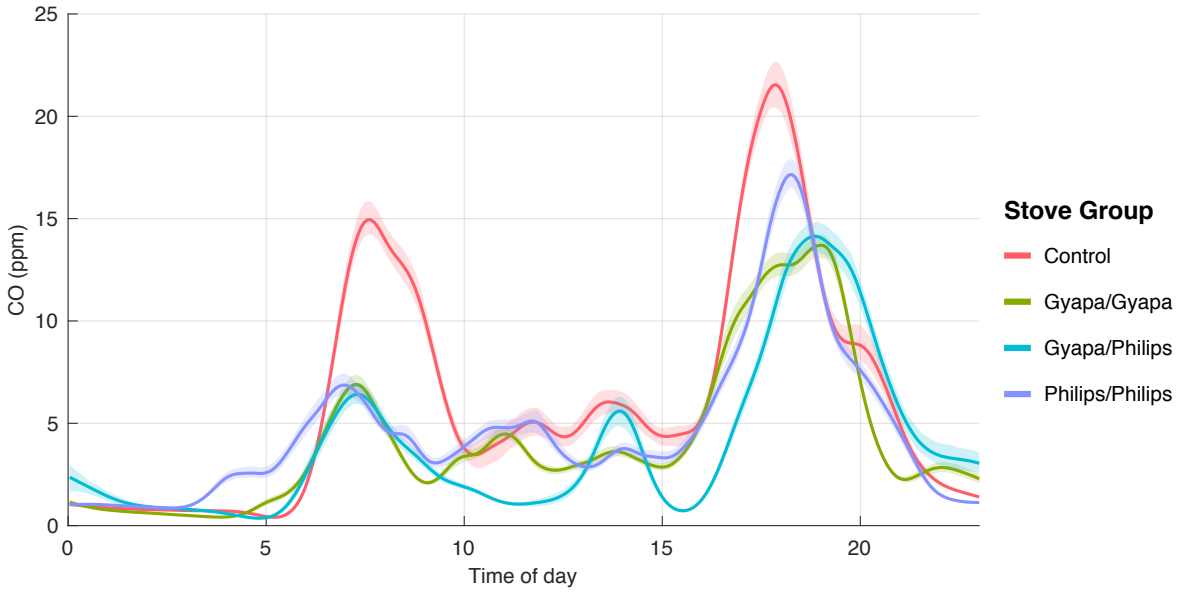

Figure 4-4. Spline-smoothed cooking area CO concentration by time of day and stove group.

4.4.3. Comparisons with previous personal CO exposure results

Exposures in this study were generally low in comparison to past cookstove studies, given the extensive use of biomass fuels in the study households. 7.5% of daily samples exceeded the WHO Interim-1 guideline over the 1-hour maximum averaging periods (35 mg/m^3), and less for the other guideline averaging periods (WHO 2010; Appendix 3). Mean (standard deviation) CO exposure for the control, Gyapa/Gyapa, Philips/Philips, and Gyapa/Philips groups were 0.98 (0.52, ±1.37), 1.10 (0.50, ±1.64), 0.94 (0.37, ±1.60), and 1.09 (0.54, ±2.12) ppm, respectively (Appendix 3 Table A3-1). Key characteristics influencing exposure here include the prevalence of outdoor cooking, where pollutants can disperse at a faster rate, and low dwelling, vehicular, and industrial activity density. In comparison, a study in The Gambia that monitored the CO exposures of children under age 5 found mean 48-h

exposures of 1.04ppm (±1.46ppm) (Dionisio et al., 2012b). In rural Upper West Ghana, a study measuring the effects of a brick and mortar chimney stove intervention found that there was no significant difference in CO exposure compared to the control group, as measured during uncontrolled cooking tests (Burwen and Levine, 2012). Ochieng et al. (2013) found median personal CO concentrations of primary cooks of 6.5ppm in rural Kenyan homes using TSFs, and 4.4ppm in homes using improved mud stoves. In Guatemala, a colder climate with more indoor cooking, adult women cooks in the control group of the RESPIRE study experienced average 48-h exposures of 4.8ppm (±3.6), while the group with an improved plancha stove had an average exposure of 2.2ppm (±2.6). Also in RESPIRE, children under 18 months in the control group had average exposures of 2.8ppm (±2.5), while the intervention group averaged 1.5ppm (±1.9) (Smith et al., 2010).

4.4.4. Relationship between personal CO and carbonaceous $PM_{2.5}$

Strong relationships between CO and either $PM_{2.5}$ EC and $PM_{2.5}$ OC were not expected in this study region, due to the observed variability in cooking areas (often moved indoors and outdoors), and the numerous contributing pollution source types observed both by observation and by source apportionment (Piedrahita et al., 2017a). This is consistent with other studies. Roden et al. (2009) directly measured emission factors from biomass cookstoves in laboratory and field settings, and found correlation coefficients between CO and $PM_{2.5}$ of 0.79 in-lab and 0.30 in-field. Dionisio et al. (2012a) measured CO and $PM_{2.5}$ (N=29) on children under age 5 in The Gambia, and found a correlation coefficient of -0.04. In Kenya, Ezzati et al. (2000) found large variations in the strength of kitchen CO-PM_{10} relationships depending on fuel and stove types. They further noted that the real-time correlation between CO and PM is weak and is thus a concern for the valid use of CO as a PM surrogate. McCracken et al. (2013) observed a strong relationship between CO and $PM_{2.5}$ likely due to the relative contribution of cooking on personal exposure in the Guatemalan study region, consistency in fuels used, and the use of an indoor cooking area with a relatively tight ventilation envelope, that serves as the nucleus of the home. Carter et al. (2016) reviewed studies performing both indoor and personal CO and $PM_{2.5}$ comparisons, and found correlation coefficients (*r*) for personal exposure ranging from 0.22 to 0.97 (median=0.53), and from 0.10 to 0.96 (median=0.71) for cooking areas. That work

discussed the sources of variability contributing to the quality of the relationship, and noted that rigorous validation of the relationship should be performed if its widespread use is intended in a study. Our study confirms this, suggesting that CO is not a good surrogate for PM exposure in our study region.

4.5. Conclusions

This work identified the effects of a cookstove intervention on CO personal exposure in a rural area of Northern Ghana. Modest decreases in exposure in the three intervention groups were observed, with the largest decrease or 14.9% seen in the Philips/Philips group ($p = 0.40$). Average exposure levels for all groups were quite low, with averages for all groups under 1.10 ppm. High acute exposures indicative of cooking events were clearly observed in many exposure time series, especially for women that are primary cooks, but they were usually lower than the WHO Interim-1 guidelines. Seasonal impacts on CO exposure were generally consistent with $PM_{2.5}$ trends identified in previous work, and likely related to fuel type, combustion and weather conditions, and cooking behavior changes. A better understanding of these effects, along with the observed trend of decreasing CO with increasing SES, would provide valuable feedback for study participants. Time of day effects by stove group showed clear differences that also warrant further investigation. Future work will also address the link between cookstove use and CO exposure over time, as stove breakdowns over the two-year study were not uncommon, and may have influenced inter-group comparisons.

The relationship between CO and $PM_{2.5}$ was weak, suggesting that CO should not be used as a surrogate for $PM_{2.5}$ exposure in the region, a finding also supported by a source apportionment analysis of the organic component of $PM_{2.5}$ from personal and cooking area samples.

CHAPTER 5

Bluetooth Beacon proximity sensing to improve exposure assessment

Ricardo Piedrahita, Yolanda Hagar, Ernest Kanyomse, Evan Coffey, Katelin Verploeg, Christine Wiedinmyer, Katie Dickinson, Abraham Oduro, Michael Hannigan

Abstract

Biomass burning for home energy use contributes to negative health outcomes and environmental degradation. In the REACCTING study (Research on Emissions, Air quality, Climate, and Cooking Technologies in Northern Ghana), personal exposure to carbon monoxide (CO) was measured to gauge the effect of introducing two different stove types over four study groups. A novel Bluetooth Low-Energy (BLE) Beacon system was deployed on a subset of those CO measurement periods to estimate participants' distance to the most-used cooking areas at each home over time. In addition to presenting methods and validation for the BLE Beacon system, we present exposure assessment modeling results using two different approaches: the first, in which the proximity data was used to better understand exposure and behaviors within and away from homes, and the second, in which the proximity data was used to improve the prediction of personal exposure via microenvironment exposure measurements.

5.1. Background and motivation

REACCTING (Research on Emissions, Air quality, Climate, and Cooking Technologies in Northern Ghana) was a 200-home cookstove intervention study in the Kassena-Nankana districts of Northern Ghana from November 2013 to February 2016. The intervention consisted of 50 households given two locally made rocket stoves (Gyapa), 50 households given two Philips HD4012 LS stoves, 50 households given one Gyapa stove and one Philips stove, and 50 control households generally using traditional 3-stone fires (TSFs) and coalpots with biomass and charcoal, respectively. The control was given their choice of stoves after the study. The study protocol is presented in Dickinson et al. (2015), while detail on the study region and population is presented in Oduro et al. (2012). Stove usage trends over the first year of the intervention

were reported in Piedrahita et al. (2016), and $PM_{2.5}$ results for personal, cooking area microenvironment, and regional measurements were reported in Piedrahita et al. (2017a). Personal CO exposure results over the duration of the entire study are presented in Piedrahita et al. (2017b, *submitted*).

In this work, we present results on carbon monoxide (CO) exposure of intervention groups, and on the relationship between personal and cooking area microenvironment CO, for the deployments that also included a novel Bluetooth Low Energy (BLE) Beacon system. We developed this BLE Beacon system using commercially available Beacons, to estimate the user's distance to the cooking area (time-activity data) and therein refine our exposure assessment and improve personal-to-cooking area CO modeling. There is substantial interest from health-related fields in acquiring detailed spatial information for personal exposure monitoring (Jerret et al., 2015).

5.1.1. Proximity monitoring background

A Bluetooth Low Energy (BLE) Beacon proximity monitoring system fills two gaps in the exposure assessment toolbox. First, the time-location data provided by the system allows for separate analysis of exposure from sources at home vs. away from home. Second, adding a significant predictor of distance within the home could allow better estimates of personal exposure from microenvironment measurements. Personal exposure measurements of air pollution have traditionally been difficult to collect due to instrument size, power consumption, operating noise, and high costs. Such issues can lead to non-compliance of protocols by users. Modeling personal exposure from microenvironment measurements is thus an attractive proposition, and past cookstove studies have done this using time-location budgets from surveys (Brauer et al., 1996; Albalak et al., 1999; Zuk et al., 2007; Cynthia et al., 2008; Dionisio et al., 2012a; Balakrishnan et al., 2002, 2004). Although regional differences make it difficult to compare across studies, other cookstove studies have used similar time-activity budgets and area monitors to predict personal exposure. Baumgartner et al. (2011) found a correlation coefficient of 0.58 (0.34, 0.75) between home and personal $PM_{2.5}$ for adult women over 24h measurements, though measurements for children were not correlated. Cynthia et al. (2008) assessed the quality of this relationship for $PM_{2.5}$ in Michoacán Mexico, and found a weak

relationship. They noted that exposures from short duration visits in rooms can be important for exposure (like walking into a smoky kitchen for a moment), and can be difficult to record using traditional methods. They proposed that until a better system is developed for recording presence in rooms, the approach should not be attempted in their study location due to important source of variability that were not being captured.

Self-reported time-activity measurement approaches (Freeman and Tejada, 2002) are resource intensive and can result in misclassifications (Clark et al., 2013). New developments in wireless technologies such as Wi-Fi allow precise indoor location estimates, but resources are required to train the identification system and a high density of Wi-Fi access points is required. Global Positioning System (GPS) devices can be used to assess location (Elgethun et al., 2003; Rooney et al., 2012), but these tools tend to have a relatively high power consumption and accuracy can suffer in regions with certain geographic characteristics and, perhaps more crucially, indoors. Radio Frequency Identification (RFID) tags can be used as binary room-location indicators, but users must place their small 'passive' type badges close to the RFID receiver, making compliance a concern. Larger 'active' RFID badges that use a battery to increase transmission power have been shown to perform well in indoor location testing (Ni et al., 2004), and would be a viable technology if the additional logging capabilities conferred by the phones are not needed. Costs are also generally higher than Beacon systems.

Allen-Piccolo et al. (2009) introduced an ultrasound-based time-activity monitoring platform (UCB-TAMS) for cookstove applications that displayed promising results. Ultrasound has lower attenuation than Bluetooth, improving signal consistency in difficult geometries or crowded spaces, but such systems have not come into widespread use. As used in their study, one receiver is placed in each room of interest, and the users wear the ultrasonic transmitters on the outside of their clothing. A receiver and three transmitters were reported to cost $80 when purchased at scale, very similar to the cost of the system presented here. We were unable to perform a direct comparison between the systems due to UCB-TAMS unavailability.

BLE Beacon technology appears well suited for indoor localization in cookstove studies as it offers a simple measurement principle, system flexibility, commodity pricing for the hardware, and the ability to use the phone for additional monitoring tasks, such as acceleration

(compliance monitoring, and potentially activity classification), real-time data sharing, and GPS, which can help identify important non-home pollution source locations. Previous versions of Bluetooth have been studied extensively for indoor localization (e.g. Bandara et al., 2004; Hossain and Soh, 2007), but with substantially different goals of high accuracy and precision, usually involving more Bluetooth transmitters and well-characterized spaces. In this work, we show that a BLE Beacon system contributes substantially to personal exposure monitoring with only zonal time-location information, which is simpler to obtain. First, Beacon system performance validation results are presented along with relevant metrics. We then show that distance categorization improves exposure model performance and adds valuable insights to the subset of personal CO exposure samples available with Beacon data from the REACCTING study.

5.2. Methods

5.2.1. Sampling system overview

A total of 71 48-h personal CO exposure samples were collected, along with BLE Beacon measurements. Primary cook females were targeted for participation in the BLE Beacon measurements to better understand the exposures and activity patterns of those spending the most time in cooking areas (Table 5-1). Thirty-eight of those samples also had cooking area microenvironment CO measurements taken (see configurations as shown in Figure 5-1). In this work, we present results for a subset of personal exposure CO measurements that have corresponding BLE Beacon measurements, but a full analysis of all personal CO measurements is presented Piedrahita et al. (2017b, *submitted*).

Table 5-1. Sample statistics for the home monitoring deployments that included BLE Beacons. These include number of deployments, average sampling duration of 48 and 24 h, number of samples removed due to faulty Lascars CO monitors.

		All available days with personal CO and beacon data	'Home cooking by stove group' vs. 'home not cooking' vs. 'away' data set (Eq. 1)	Personal vs. cooking area CO by zones (Eq. 2)	Daily average personal vs. cooking area CO (Eq. 3)
Duration	Compliant and non-flagged periods deployed	279 (time-activity periods)	107 (time-activity periods)	123 (zone-days)	38 (days)
	Daily compliant duration in hours (mean (stdev))	19.9 (3.25)	20.28 (3.6)	20.94 (3.48)	20.22 (3.81)
	Unique participants	31	22	21	22

Gender covariates	Primary cook Females	228	101	115	36
	Non-primary cook females	51	6	8	2
	Males	0	0	0	0
	Female over 5y (med, stdev, max, min)	38.4, 12.9, 12.3, 73.4	39.4, 14.2, 73.4, 12.3	39.4, 14.2, 73.4, 12.3	39.4, 14.8, 73.4, 12.3
	Female under 5y (med, stdev, max, min)	2.1, .9, 1.9, 4.2	3.3, .5, 3.8, 2.9	3.3, .5, 3.8, 2.9	3.3, .6, 3.8, 2.9
SES	Poorest	48	24	30	30
	Poorer	72	24	26	26
	Poor	69	12	15	15
	Less poor	27	20	23	23
	Least poor	63	27	29	29
Seasons	Harmattan	150	37	47	13
	Hot dry	23	11	15	4
	Light Rainy	31	20	25	4
	Heavy Rainy	75	39	36	14
	Transition	0	0	0	0
Stove Group	Control	31	14	15	6
	Gyapa/Philips	48	27	28	9
	Philips/Philips	110	29	31	10
	Gyapa/Gyapa	90	37	49	13

Microenvironment air quality monitors (G-Pods, Boulder CO, mobilesensingtechnology.com) were placed in the two most used cooking areas in each home, and were equipped to measure CO and CO_2, and in some cases, integrated $PM_{2.5}$, and total VOCs (Appendix 4; Dickinson et al., 2015). Study participants carried personal CO monitors along with Bluetooth-logging Android phones. BLE Beacons were adhered to the G-Pods to provide distance estimates between participants and the two primary cooking areas. Any personal location tracking device introduces serious ethical questions, as the potential for misuse and exploitation exists (Benatar et al., 2002), so verbal consent was obtained from study participants, after explaining the operation of each instrument they carried, and participants were also given the option to have their data deleted at the end of the sampling period if they so desired.

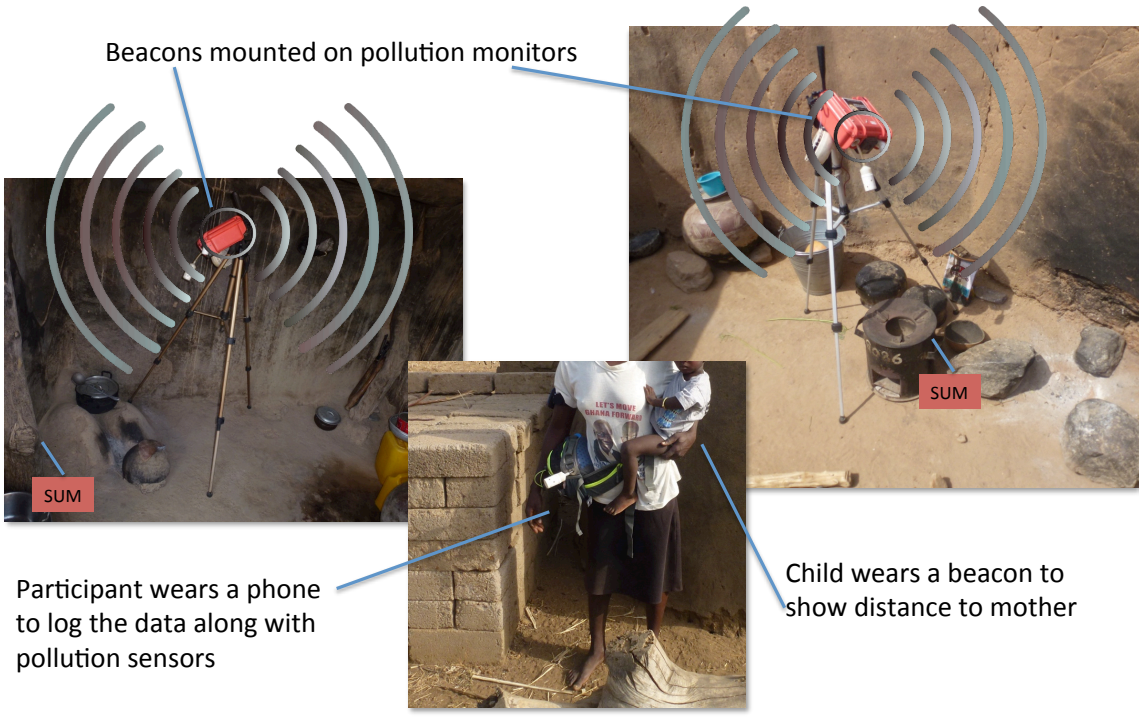

Figure 5-1. Example deployment diagram. Beacons were mounted on the cases of the orange G-Pod pollution monitors. Logging Android phones were worn by participants along with personal air quality monitors.

5.2.2. BLE Beacons

BLE Beacons are small battery powered devices that periodically broadcast their MAC addresses and other unique identifying information. They have found use in a variety of commercial applications requiring location-based services, such as advertising, where a phone would perform a task upon receipt of a BLE Beacon signal, like offering an in-store coupon. Roximity Model X Beacons (Roximity, Denver, CO) were used in our study due to their small size (6.4 *x* 6.4 *x* 2.5 cm), claimed battery life of 5 years, and cost of $12 (USD) per Beacon. They employ the Apple iBeacon protocol to transmit data, but in our application, only the Beacon MAC addresses and received signal strength indicator (RSSI) are recorded, so this approach can be used with both iOS and Android devices. The phones record the identifying information and RSSI from any Beacon within range (generally less than 100m in open space). RSSI is then converted to a distance measure, providing an estimate of distance from phone to Beacon.

5.2.3. BLE Beacon Receivers

Phicomm C230w Android phones served as Bluetooth receivers and data loggers ($56 (USD) per unit). A custom Android application was written and installed on each phone to log the Beacon address data, Beacon RSSI, as well as acceleration, GPS, and GPS accuracy (GPS can be manually disabled). Data was logged every six seconds to the phone's microSD card in the JSON format. The app can be configured to upload data to a remote server, but data was downloaded manually in our study. The phone battery was swapped for an external 6.6Ah li-ion battery pack, yielding 50-60 hours of continuous use. Phones with battery packs weighed 280g, and were consistently placed in the outer pockets of the personal sampling pack.

5.2.4. BLE Beacon Data Processing

RSSI is sensitive to path effects like room geometry, and obstructions in the measurement area, including people, since water is a strong signal attenuator for Bluetooth, transmitting on the 2.4GHz band. Considering such limitations, many applications use distance categories. Here, we used zones defined as 'near' (<15m), 'medium-near' (15-30m), 'medium-far' (30-50m), 'far' (50-90m), and 'within signal range' (>90m).

To understand signal measurement uncertainty, we can first look at the results of a simple test we conducted outdoors with one phone and one Beacon. When the body is directly between a phone and Beacon, the signal attenuation is equivalent to predicting a change in distance from ~1 meter to ~10 meters. When performed indoors, the results are usually less pronounced, due to signal reflectance aiding the Beacon signal to reach the phone. The two primary modes of localization miscategorization are 1) high frequency attenuation, or a 'teleportation' effect, where phones appear to jump between distances faster than physically probable, and 2) sustained attenuation that consistently places the user farther from the Beacon than they are. The first issue can be mitigated with algorithms (Madhavapeddy et al., 2005; Zanca et al., 2008). The second issue was not addressed in this work, which may have resulted in bias, but this effect can be mitigated with more BLE emitters or receivers throughout the study area, or if other types of sensors are also used.

We developed a filtering algorithm to reduce the high-frequency attenuation effects, as previous works have mainly focused on precise within-room location or room categorization (Zhou et al., 2006; Dahlgren and Mahmoud, 2014) rather than distance time series

categorization. Our approach, the 'maximum velocity' (MV) filter, assigns greater weight to higher signal strength data by defining a maximum change in distance over time (an 'expected walking velocity'), and recursively adjusts the signal strength values *i* according to the previous value *i-1*. The expected velocity here was set to $ß = 1\ m/s$, and with Δt the time between samples, the predefined maximum distance is then $d_{max} = ßΔt$, giving $d_i = d_{i-1} + ßΔt$. For example, as we collected six-second data, if consecutive distance readings are 5m and 25m, the second data point would be modified to $d_2 = 5m + 1m/s*6s = 11m$. Minute medians are then extracted from this data to further reduce noise and align with other minute-data. Participant compliance, meaning the daytime hours when participants were predicted to be wearing the phone and air sampling equipment was estimated at 81.9% using the variability in the Bluetooth signals over time (Piedrahita et al., 2017b, *submitted*; Appendix 4).

5.2.5. Calibration and Validation

In July, 2016, the Beacon RSSI values were calibrated against distance, and validation testing took place. We first performed testing in an open field as a base case to assess calibration reliability and the effectiveness of our classification scheme. Two phones were placed in the center of a set of concentric circles at radii of 2m, 5m, 10m, 20m, and 40m, and a person wearing two Beacons on either hip walked slowly and randomly throughout each zone for 20 minutes. These zone categories were different (smaller) than those used in the models for the field data because we found that in the field, sustained attenuation interference effects observed in typical homes resulted in unbalanced distributions over the distance categories, and we wished to balance them. A second validation test was later performed in a location with additional obstructions (Appendix 4).

Before the start of each 20-minute testing period, there were periods when the tester stood still at the intersections of the areas, in order to generate known calibration data. The data from both Beacons and both phones was aggregated to generate a single calibration function for all data collected during validation testing as well as throughout the study deployments in Ghana. Details on BLE Beacon distance calibrations are presented in Appendix 4.

Classification into zones was performed using a single Beacon, as the deployment in Ghana did. However, we also tested the performance of a merged signal that combined the two Beacons worn on the hips, by selecting the stronger signal at each time point, then applying the MV filter. This approach could be used to reduce the multi-path and attenuation effects in future deployments, though if participants carry the Beacons rather than the phone, they would miss out on benefits of wearing the phone, such as user GPS and acceleration logging. Classification performance was assessed using the matching success rate and the rate at which the predicted classification was within one zone of the correct zone, for all available combinations of phones and Beacons.

Validation testing for the 'open field' deployment and all combinations of phones and Beacons, when using the MV filter and data from a single Beacon, showed correct classification of zones on 34.7% of observations, and 65.3% of observations were within one zone of the correct zone. In the later validation test with additional obstructions, those classification rates were 28.2% and 68.6%, respectively. The classification rates when using merged data from both Beacons on the hips were 53.2% and 89.5% for correctly classified, within-one classification for the 'open field' test, and a similar 46.0% and 91.3% for the validation set with obstructions (Appendix 4, Figures A4-2 and A4-3).

5.2.6. CO exposure as a function of time-activity category

The inclusion of time-activity data allows us to gain additional information about how different cookstoves and cooking behaviors impact CO exposure. To help illustrate this, a time series of Beacon proximity data along with personal and cooking area CO is presented in Figure 5-2. The data show a clear relationship between personal CO exposure and cooking area CO when the user is at home, and sharp reduction of personal CO when the user leaves home at 13:00, as the home CO level remains elevated. The home also appears to use the Philips stove more than the TSFs throughout the day. This user is unique in that they do not spend the night within range of the cooking area, either spending it in another home, or obstructed enough to be out of range. It can also be seen that periods spent near the stove, as defined by the nearest proximity values, have high variability. It is difficult to discern whether this is due to real movement or teleportation effects, and additional Beacons could reduce this uncertainty.

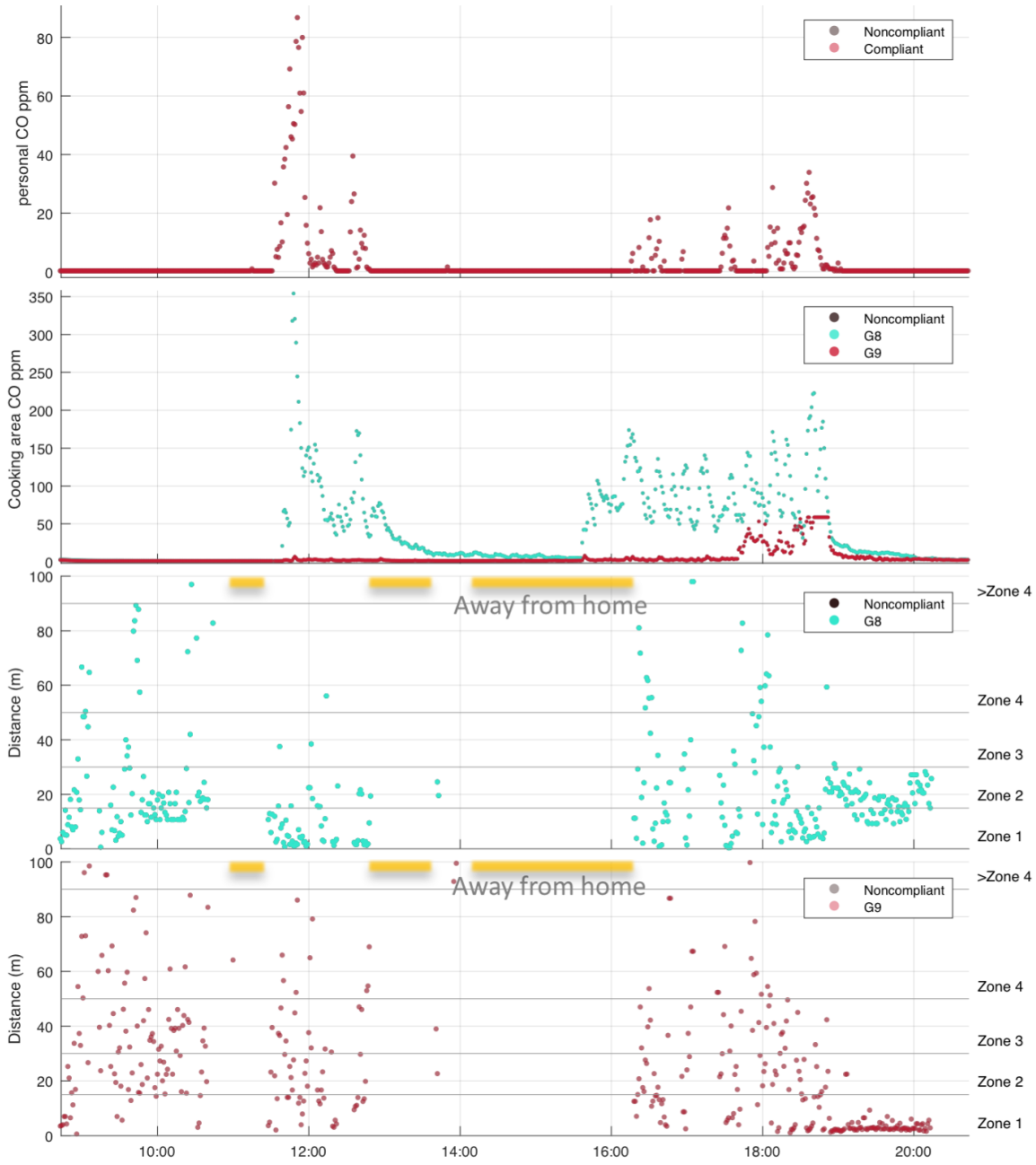

Figure 5-2. Time series showing personal and cooking area CO concentration with Beacon proximity data to each cooking area. The cooking area monitor and their respective Beacons were named G8 and G9. Lower plots show proximity to the given cooking areas, and the number of samples observed in each zone, or in the case of >zone 4, when signal is weak or lost. Zone thresholds were defined as 15m, 30m, 50m, and 90m.

To quantify some of these features, a mixed effects regression model was used to determine the effect of the intervention on CO exposure for various time-activity categories. Specifically, in Equation 5-1, log(Mean personal CO_{ijk}) represents the log-transformed average

CO concentration on day *i* for individual *j*, in time-activity *k*. The 'time-activity' categorical variable is defined using the Beacon proximity data in conjunction with available microenvironment CO data, and has six possible states: State 1 ("Away From Home"): participant is considered 'away from home' if more than 90m away from both cooking areas; State 2 ("Home Not Cooking"): participant is within 90m of any cooking area but no cooking appears to be in progress; and States 3-6 ("Home Cooking"): for each of the four stove groups, participant is within 90m of any cooking area in the home, and any cooking area CO measurement in the home is above 10ppm.

To determine the effect of stove group on personal exposure at home when the stoves are in use, the 'home cooking' category is interacted with stove group giving categories of 'control group home cooking', 'Gyapa/Gyapa group home cooking', 'Philips/Philips group home cooking', and 'Gyapa/Philips' home cooking. Covariates such as socioeconomic status and season were too sparse in some categories to include. The individual random intercept α_j accounts for the correlation within subjects due to repeated measures, and e_{ij} represents the random variation from subject to subject. More than 12 hours of data was required of the daily data (50% data completion) for inclusion in the model, as the primary goal of these models was to assess the system rather than assess exposure over the entire intervention.

$$Log(Mean\ personal\ CO_{ijk}) = ß_0 + ß_1(TimeActivity_{ijk}) + \alpha_j + e_{ijk} \qquad \text{Equation 5-1}$$

This model is useful for identifying time-activity categories with high average exposures. However, it is also beneficial to also consider time spent in each time-activity category to identify where the most exposure comes from each day. Equation 5-1 was thus modified by changing the dependent variable to total exposure by daily time-activity (ppm-hr) (Equation 5-2). This approach highlights nuances in different cooking behaviors. For example, if one stove group has very high average exposures near the stove, but does not spend as much time near that stove, total exposure levels may be different than stoves that have the opposite effect.

$$Log(Total\ personal\ CO_{ijk}) = ß_0 + ß_1(TimeActivity_k) + \alpha_j + e_{ijk}$$ Equation 5-2

5.2.7. Personal vs. microenvironmental CO modeling

In addition to the exposure assessment models described in Equation 5-1 and Equation 5-2, we also investigated average personal CO exposure as a function of the user's distance away from each cooking area and the cooking area CO measurements (see Equation 5-3). This approach reflects previous efforts to estimate personal exposures by assigning mean area concentrations from different areas in a home, using time-activity budgets (Dionisio et al., 2012a; Zuk et al., 2007; Cynthia et al., 2008; Özkaynak et al., 1999). Such an approach would be expected to perform better with precise time-activity budgets from a BLE Beacon system. Here, we show that this is the case when looking at the cooking area microenvironment by isolating exposures when the users are at home. The dependent variable was the log transformed average personal CO exposure for each user deployment *i*, at each distance zone *j* from a cooking area, using only observations when the participant was within zone 4 (based on the BLE Beacon distance data). The independent variable was cooking area CO, linearly scaled by distance zone so as to account for dispersion (e.g. 100% of the cooking area CO was applied if the user was in the nearest zone to the cooking area, and 80% if in the second nearest), and then log transformed. An exponential weighting scheme was also tested to reflect the Gaussian dispersion of CO through the cooking environment, but resulted in no significant difference in performance, likely due to the naturally high variability in the environment. If multiple cooking areas were monitored, we used a weighted average of the cooking areas based on the participant's proximity to each. It is implicitly assumed that concentrations within each zone are uniformly distributed, and average exposures within each zone are independent of one another.

$$Log(Personal\ CO_{ijk}) = \beta_0 + \beta_1(weighted\ cooking\ area\ CO_{ijk}) + \alpha_j + e_{ijk}$$ Equation 5-3

As a contrast, we also examine how the exposure assessment model results change if the Beacon proximity data were removed (Equation 5-4). To do so, log transformed daily average personal CO concentration was regressed against log transformed daily average cooking area CO concentration. Thirty-eight daily samples were available for analysis in which Beacon data was also collected.

$$Log(Personal\ CO_{ij}) = \beta_0 + \beta_1(Daily\ average\ cooking\ area\ CO_{ij}) + \alpha_j + e_{ij}$$ Equation 5-4

5.3. Results

The BLE Beacon system allowed us to assess and compare participant time-activity characteristics. On average, participants spent 8.5% (± 7.9%) of their sampling days at home while cooking, 51.4% (± 30.5%) of their time at home and not cooking, and 40.1% (± 32.1%) of their time away (recall that 'away' is defined as beyond zone 4 (90m), and can still be within signal range). These results were calculated using the data for which cooking area CO was monitored. For the data set in which only CO and Beacon data was collected (33 additional days, presented as 'all available data' in column 1 of Table 5-1), 24.9% of the day was spent within zone 1, 14.4% was spent in zone 2, 13.4% was spent in zone 3, 14.8% was spent in zone 4, and 39.3% was spent beyond zone 4 (Figure 5-3). We observed some participants with over 90% of their time spend in zone 1, which seems unreasonable, and could be indicative of non-compliance, suggesting that additional compliance filtering steps may be appropriate in future work. Time-activity variability among users was high, changing with tasks and behaviors depending on household needs. Figure 5-3 shows that 32.6% of total daily exposure (ppm-hr) was experienced within zone 1, 30.7% was experienced in zones 2-4, and 36.7% was incurred beyond zone 4.

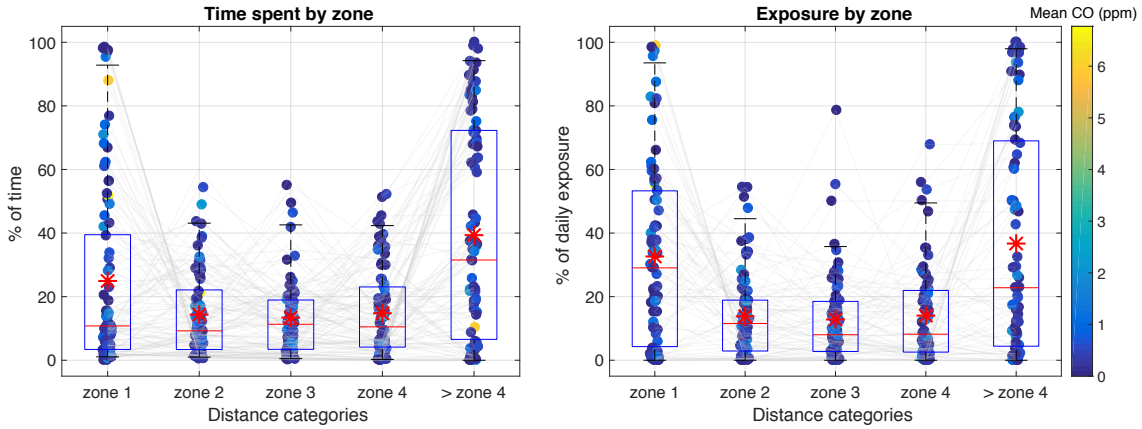

Figure 5-3. Time spent in each zone as percentage of the day. Marker colors indicate the day's mean exposure to CO. Some participants spent nearly the entire day within zone 1, leading to questions about compliance. Additional sensor streams could improve our measurement of compliance in future work.

Directly analyzing average exposure by the zone, the daily median and average exposures were highest in the near-cookstove regions, decreasing with increasing distance from the cooking areas, although this decreasing trend was not statistically significant (Figure 5-4).

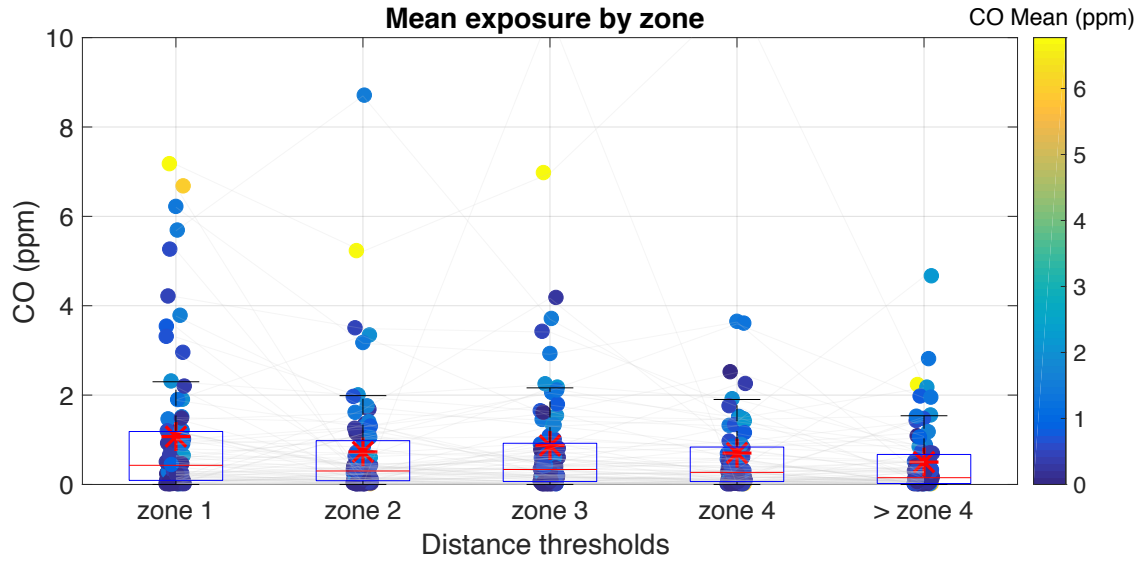

Figure 5-4. Mean exposure distributions categorized by zones. Marker colors indicate the participant's average exposure from the entire day, and red stars represent means by zone. Slope of decreasing average exposure by zone was not found to be statistically significant by univariate linear regression.

5.3.1. CO personal exposure results using home vs. away categorization

The reference category from Equation 5-1, which is the control group at home and cooking, had expected exposure levels of 3.62ppm (0.78, 16.75ppm). Relative to the reference group, the three intervention groups had large but not statistically significant reductions in exposure during cooking periods at home. The Gyapa/Gyapa group had 82.4% lower exposure (0.64ppm, (0.10, 4.05), p = 0.07), the Philips/Philips group had 62.4% lower exposure (1.36ppm, (0.20, 9.20), p = 0.31), and the Philips/Gyapa group had 81.1% lower exposure (0.69ppm, (0.10, 4.62), p = 0.23). Exposures in the 'home not cooking' and 'away' categories accounted for 95.0% (0.18 ppm (0.03, 0.94), p = 0.00) and 96.5% (0.13ppm (0.02, 0.65), p =0.00) lower exposure than the reference group, respectively. While some of these differences are not significant statistically, the dramatic difference in estimates is notable, and may be significant with larger sample sizes. See Table 5-2 for results.

Modeling integrated exposure by time-activity category (Equation 5-2) yielded similar results, indicating that the categories with the highest average exposures were also contributing to most of the personal exposure (Table 5-2). The Gyapa/Gyapa, Philips/Philips, and Gyapa/Philips homes were respectively responsible for 94.5% (p = 0.01), 71.8% (p = 0.30), and 92.7% (p = 0.01) lower integrated exposures relative to the control group, who experienced 2.45ppm-hr of integrated exposure (0.53, 4.37), while cooking at home. 'Away' and 'home not cooking' had integrated exposure contributions that were 89.0% and 96.0% lower than the control group's total daily cooking exposure.

Table 5-2. Summary of results from Equation 5-1 and Equation 5-2, modeling personal CO exposure by time-activity categories.

	Average personal exposure vs. 'home cooking', 'home not cooking', and 'away' (Eq. 5-1)				Total integrated personal exposure vs. 'home cooking' , 'home not cooking', and 'away' (Eq. 5-2)			
	Expected value ppm (95% CI)	Coefficient (95% CI)	% change (95% CI)	P-value	Expected value (ppm*hr)	Coefficient (95% CI)	% change (95% CI)	P-value
Intercept (control group home cooking)	3.62 (.78, 16.75)	1.29 (-0.24, 2.82)	NA	0.10	11.57 (1.69, 79.07)	2.45 (.53, 4.37)	NA	0.01
Gyapa/Gyapa Home cooking	.64 (0.10, 4.05)	-1.74 (-3.58, 0.11)	-82.4 (-97.2, 11.7)	0.07	.64 (.06, 6.45)	-2.9 (-5.22, -.58)	-94.5 (-99.5, -44.2)	0.01
Philips/Philips Home cooking	1.36 (0.20, 9.2)	-0.98 (-2.89, 0.93)	-62.4 (-94.4, 153.8)	0.31	3.26 (.30, 35.89)	-1.26 (-3.66, 1.13)	-71.8 (-97.4, 210.3)	0.30
Gyapa/Philips Home cooking	0.69 (0.10, 4.62)	-1.67 (-3.57, 0.24)	-81.1 (-97.2, 27.6)	0.09	.84 (.08, 9.28)	-2.62 (-5.01, -.22)	-92.7 (-99.3, -19.7)	0.03
Home not cooking	0.18 (0.04, .94)	-2.99 (-4.62, -1.35)	-95.0 (-99.0, -74.2)	0.00	1.27 (.16, 9.83)	-2.21 (-4.26, -.16)	-89.0 (-98.6, -15.0)	0.03
Away from home	0.13 (0.02, 0.65)	-3.36 (-4.99, -1.72)	-96.5 (-99.3, -82.2)	0.00	.46 (.06, 3.59)	-3.22 (-5.27, -1.17)	-96.0 (-99.5, -69.0)	0.00
	Eq. 1a				**Eq. 1b**			
Random effect by individual variance	0				0			
Random error variance	2.98 (2.28, 3.89)				4.69 (3.59, 6.14)			
Adjusted R-squared	0.20				0.08			
N	107				107			

5.3.2. Personal CO exposure assessment using cooking microenvironment CO

Model results using Equation 5-4 indicate that on a daily average basis, the log of cooking area microenvironment CO is a significant predictor of the log of personal CO exposure ($p < 0.01$), accounting for 28% of within-subject variability ($R^2_{adjusted} = 0.28$). The coefficient on the log of weighted area CO was 0.81 (CI = 0.40, 1.21), corresponding to a 124.3% (49.9%, 235.6%) increase in personal CO for an increase of one on the log of the weighted area CO.

The model from Equation 5-3 (Appendix 4, Table A4-1), fitting the log of average personal CO exposure with the BLE Beacon zone-weighted cooking area CO measurements (Equation 5-3, Figure 5-5), was much improved over the daily average model, accounting for 63% of within-subject variability. With Equation 5-3, there was a 173.5% (124.8%, 232.9%) increase in personal at-home expected CO for every decrease of 1 on the log-transformed

weighted cooking area CO (as the participant got closer to the cooking area). The random intercept variance was 0.35, and intra-class correlation coefficient was 0.26.

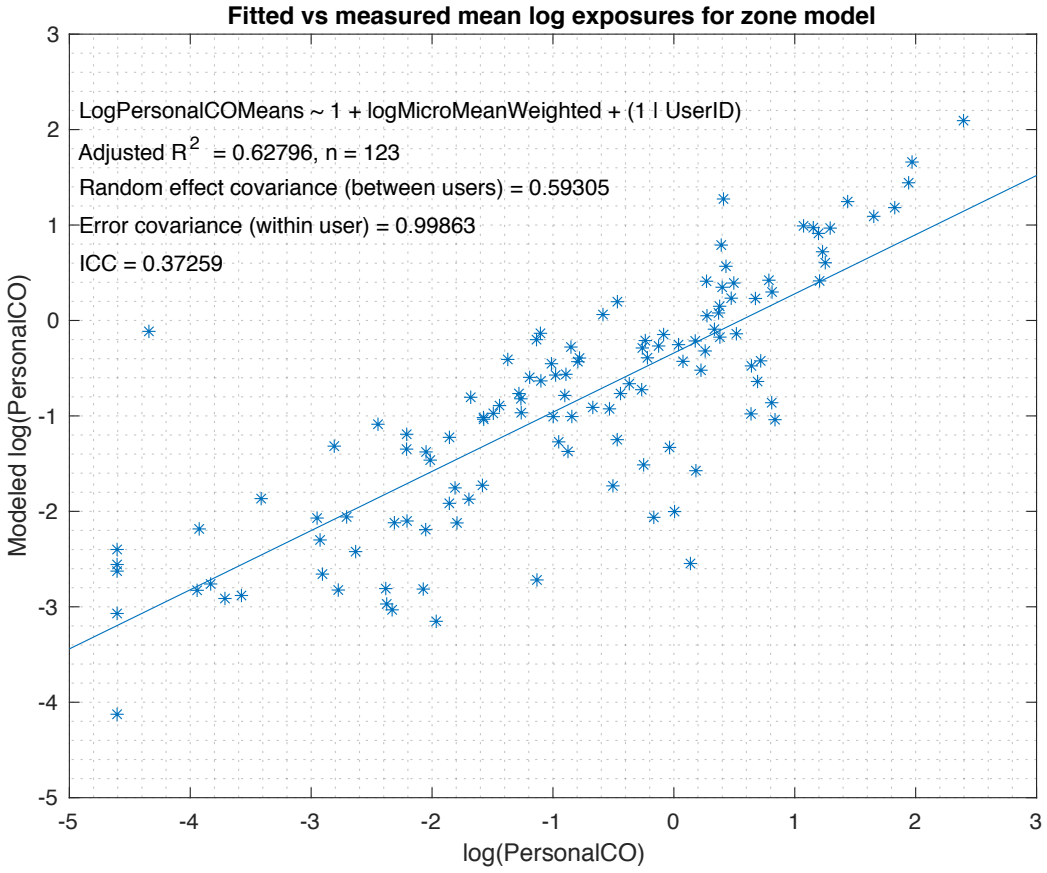

Figure 5-5. Relationship between at-home personal and cooking area CO using Beacon weight-derived cooking area CO, using Equation 5-3.

5.4. Discussion

5.4.1. Personal exposure models

Model results using Equation 5-1 (Table 5-2) showed significantly higher average exposures during 'home cooking' in the control group relative to the 'home not-cooking' and 'away' categories. No significant differences were found between the control group and the intervention groups for the 'home cooking' data, though the effect estimates showed

substantially lower exposure for all groups relative to the control group (at least 62.4% lower). This suggests that the intervention may have served to reduce CO exposure, and low statistical significance is due to low sample sizes and high variability. Analysis of the daily average values for this same data set showed lower exposure for the intervention groups, but by smaller margins, and still not significant (Appendix 5). Analysis of the complete CO exposure data set carried out in Piedrahita et al. (2017b, *submitted*), again with daily averages, showed lower exposure for the three intervention groups, where the largest reduction was for the Philips/Philips group at 14.9% ($p = 0.40$), the Gyapa/Philips group was 0.1% lower ($p = 1.00$), and the Gyapa/Gyapa group was 5.6% lower ($p = 0.78$). Modeling the effect of stove group using daily averages with only the data available with Beacons yielded much larger reductions relative to the stove group, so the magnitude of effects seen in the results from Equation 5-1 are likely also a consequence of the small sample size (Appendix 4.7). The intervention cookstoves may have also decreased in performance over the course of the intervention, perhaps reflected in the more modest exposure reduction in the Philips/Philips group here, as the Beacon-supported measurements were collected in the latter half of the study period.

The importance of home-level air pollution sources thus appears to be quite substantial, but it is important to consider that the average exposures were based on different time durations, and cooking takes up less time than the 'home not cooking' and 'away' categories. Model results using Equation 5-2 reflect this, and summary statistics presented in Figure 5-3 indicate that 36.7% of average daily exposure is experienced beyond zone 4. Source apportionment of $PM_{2.5}$ in the same study showed that two cooking-related sources accounted for a median 15.3% of elemental carbon (EC) and 9.2% of organic carbon (OC) in personal and cooking area concentrations, with other important sources including a biomass combustion source that appeared to be more regional in nature, and vehicular combustion (Piedrahita et al., 2017). It should be noted that neither total daily $PM_{2.5,}$ nor EC and OC have been found to be well correlated with CO in rural settings in personal or microenvironmental measurements in the region (Dionisio et al., 2012a; Piedrahita et al., 2017b, *submitted*).

Results from the models using Equation 5-3 and Equation 5-4 show that microenvironment CO measurements coupled with cooking area proximity data can

substantially improve prediction of personal exposure using area measurements, even in areas with high variability in cooking location, ventilation, and cooking area geometry.

While the system we have developed shows promising results, further testing should be performed to assess performance and limitations in other regions, household member types, seasons, and with other pollutants. Additionally, the relationship is likely to vary based on regions and cooking behaviors, so pilot studies should always be performed to determine model coefficients and quality of fit.

5.5. Conclusions

With a relatively low budget of $600 for 5 sets of equipment, we were able to add time-activity data that improved our understanding of personal CO exposure in REACCTING. There were significant inter-user differences by exposure location, and thus exposure sources. One mode of user had CO exposure tightly correlated with distance from the main cooking area, while another experienced the majority of their exposure outside the home. The results presented here demonstrate the ability to bypass potential sources of error and cofounding, and more accurately measure CO exposure differences due to the intervention. Such a system could even be used to customize exposure reduction strategies to different types of users.

We find that even in the dynamic and predominantly outdoor homes in Northern Ghana, using the time-activity data provided reasonably good performance in predicting personal exposure. We would expect improved performance in places with tighter building envelopes, more time spent in the main cooking area, and fewer sources of combustion emissions. In addition to improved exposure modeling and time-activity apportionment, variability within individuals can be explored in detail. While so much information may be difficult to synthesize into a model due to the high variability, and need for much more data to do it well, having such data provides great potential going forward.

CHAPTER 6

Laboratory assessment of electrochemical carbon monoxide monitors

Ricardo Piedrahita, Yolanda Hagar, Michael Hannigan

Abstract

Lascar USB-CO monitors have been widely used in recent personal exposure studies due to their low cost, low-power consumption, ease of use, simple calibration, and stability over time. However, in some field trials, we observed changes in sensor sensitivity and increases in response time as the sensors aged. In that study, we were interested in peak mixing ratios and average exposures over sampling periods. Changes in response time can affect the average exposure substantially, and as such we sought to understand the causes and magnitudes of such changes in sensing characteristics. We thus conducted laboratory experiments during an attempt to accelerate the aging of new sensors using high CO exposure and drying of the electrolyte. We found significant changes in response times with cumulative exposure of the sensor, and differences between individual sensors.

We also used the laboratory data to compare three approaches for generating and applying calibrations to inform expected performance in the field. The first method, for optimal measurement accuracy, and ideal for a study with ample resources, was a multi-point calibration on the day of every deployment. The second method, useful for studies with more limited resources, used only calibrations from the start and end of the five-month period, and applied an interpolated sensitivity to the data in between. The final approach used the raw value directly generated by the Lascar CO monitor. We found average exposure errors of 1.9% and 1.2% for the first and second calibration approach, respectively, and 19.1% error using the raw data.

6.1. Background and motivation

Electrochemical gas sensors are widely used in personal exposure monitoring, owing to their low power operation, small size, relatively low cost ($20-100, typically), linear response, calibration stability over time, and fast response times. Sensing challenges can include cross-

sensitivities to non-target gases, and non-linearity with temperature and humidity. Practical challenges include a lifetime on the order of one to two years (though the sensor used in this work lists a lifetime of seven years; Nemoto, 2016) due to degradation of sensor electrodes and drying of the electrolyte solution, which can also change sensing properties nonlinearly over time. Previous works using real-time and dosimeter tube CO measurements abound in the cookstove literature, but there has been limited work published on sensor characteristics, like calibration of real-time monitors, agreement of duplicates, sensor response times, and response linearity over time. Edwards et al. (2007) and Smith et al. (2010), presented calibration methods for the HOBO CO logger (Onset Computer Corp) similar in measurement principal to the Lascar USB-CO used in this work. Cheng et al. (2010) and Young and Jones (2014) have characterized the response and decay times of CO data loggers (Langan, and Lascar USB-CO, respectively) as a means of correcting the time series, especially in response to short duration peaks that can be underestimated due to sensor dynamics. Here, we assess the stability of characteristic response times as well as calibration coefficients with a batch of ten Lascar USB-CO1000 monitors tested from March 31, 2016 to October 7, 2016, under laboratory conditions.

Changes in sensitivity and baseline drift can be adjusted for using linear regression calibrations. However, measurement of sensor response times requires a more involved experimental setup. We present the extent of the potential error derived from these effects as a function of total previous exposure experienced by the sensor, assess inter-monitor variability, and offer practical advice for maintaining data quality in challenging study conditions.

6.1.1. Electrochemical sensor operation

The Nemoto NAP-505 sensor used in the Lascar CO-USB allows gas to diffuse through a filter membrane, then through a capillary, a charcoal filter, and finally to the working electrode where CO is oxidized with H_2O to form CO_2, $2H^+$ and $2e^-$. The $2H^+$ migrates into an acidic electrolyte solution bathing the electrodes, while the electrons are measured with a specially designed potentiostat circuit. This circuit then provides electrons to a counter electrode to maintain an equal potential between both electrodes. At the counter electrode, atmospheric

oxygen reacts with hydrogen and electrons to form H_2O, replenishing the electrolyte. A reference electrode is maintained at a constant potential to help regulate the working electrode. The manufacturer states that the time to normal operation is under two minutes for a sensor stored for six months (Nemoto, 2016). The manufacturer and peer reviewed works have demonstrated high linearity for various types of electrochemical CO sensors, within mixing ratios of interest for personal exposure measurements (0 to over 100 ppm) (Buck et al., 2013). Response times were characterized for three different electrochemical sensor types in that work, with T_{90} times averaging 11.5 to 33.8 seconds. They found no correlation between T_{90} response times, and the set point mixing ratios they were approaching. However, they found a strong linear correlation between the slope of the initial sensor response and the mixing ratio set point in the chamber, suggesting that predicting the actual mixing ratio should be possible from the initial slope of a signal rise. Decay times were not investigated in that work.

6.1.2. Electrochemical sensor performance limitations

Sensor response times are governed by both gas diffusion rates and reaction rates. Reduced diffusion rates could be caused by befouling of the primary filter, increasing the time needed for the target analyte to reach the working electrode. Reduced reaction rates could be due to damage or corrosion of the working electrode, leaving fewer available reaction sites, or due to evaporative loss of electrolyte fluid over time, which would result in slowed H^+ ion transport through the electrolyte so that the reduction reaction at the counter electrode could not occur as quickly. Inspecting previously used Lascar USB-CO monitors it is clear that both occur, with visible fouling of the filter surface, as well as dried out electrolyte baths, though the relative importance of these is not determined in this work.

6.1.3. Our testing approach

Our research group first used the Lascar USB-CO monitors during a cookstove intervention study in Ghana (Dickinson et al., 2015; Piedrahita et al., 2017b *submitted*). The monitors generally worked well and exhibited good stability, but we observed a rate of sensor and logging failures that prompted us to investigate further. Monitor failure was presented with four different symptoms: 1) failure to log data, often due to damage to the electronics from moisture or impact; 2) highly sporadic and fluctuating data, which may be due to electrical

issues with the embedded system or sensor; 3) slow response and recovery times due to CO spikes; 4) no sensor response to CO. In the REACCTING study, 25.0% of sample days (201 days) were removed due to the last three issues. To further investigate symptoms three and four, controlled laboratory tests were performed at the Hannigan Lab. The main modes of electrochemical sensor degradation were expected to be damage to the sensor electrodes, drying of the electrolyte solution, and fouling of the sensor's particle filter. We sought to determine the importance of the first two causes, as the last is dependent on many possible types of random befouling events.

6.2. Methods

6.2.1. Laboratory set up

Laboratory testing was conducted at the Hannigan Lab at the University of Colorado, Boulder. A LabVIEW controlled gas delivery system was employed (Piedrahita et al., 2014). To achieve the desired CO mixing ratios between 0-700 ppm, two different CO gas standards were used; 1.) A certified 1010 ppm standards from Air Liquide, injected through a 0-1000 sccm MFC, and 2.) A pure CO standard from AirGas, injected through a 0-20 sccm MFC. A 0.94L chamber was used to house the 5-10 Lascar monitors used at any time during testing. With 10 Lascars and dead-volume blocks, the empty volume remaining in the chamber was 0.3L. A calibrated mass flow controller (MFC, Tynan Corp.) flowed zero-grade air at a constant rate of 3 $lmin^{-1}$, resulting in a T_{90} time of 24 seconds in the selected chamber (6 seconds/air exchange). A pre and post-calibrated API 300 CO monitor provided the reference data with which to compare the Lascar measurements. The API monitor sampled directly from the testing chamber at a rate of 0.790 $lmin^{-1}$. The sensors were tested at room temperature (mean 26.6 ^{o}C, $\pm$2.4 ^{o}C) and high temperature tests were conducted (35 ^{o}C) to assess changes in sensitivity and response time. A temperature probe inserted into the chamber provided feedback for the temperature controller, and a heat lamp was used to heat the chamber uniformly. In this work, only the room-temperature tests are analyzed.

6.2.2. Accelerated aging test procedure

Ten new and unused Lascar USB-CO monitors were set to log at 10s intervals. An initial test was performed with all 10 units over a 22-hour period. Two identical 11-step calibrations

were repeated consecutively, alternating clean air and a CO spike for 30 minute periods as follows: clean air - 18 ppm CO - clean air - 39 ppm CO - clean air - 81 ppm CO - clean air - 220 ppm CO - clean air - 400 ppm CO - clean air, though there was some variability in the mixing ratio set points due to system variation. After this, a two-hour simulation of a typical cooking event CO time series was performed, with CO values ranging from 0-500ppm. Then, the chamber was heated to 35 ^{o}C, and the 'base' calibration was performed again. A time series of a calibration is shown in (Figure 6-1). Five of the units (the 'fresh controls') were stored in a sealed bag between trials to serve as controls and used for comparison at the end of the tests. The 'fresh' control units were tested four times over the 26 weeks (on the 1st, 15th, 16th, and 26th week), while the 'used' batch of was tested 23 times over that period, with most calibration occurring in the first two months of testing. To accelerate the aging process, dry air was used in the tests, and before each test, a four-hour exposure to elevated mixing ratios (400-600 ppm) was carried out.

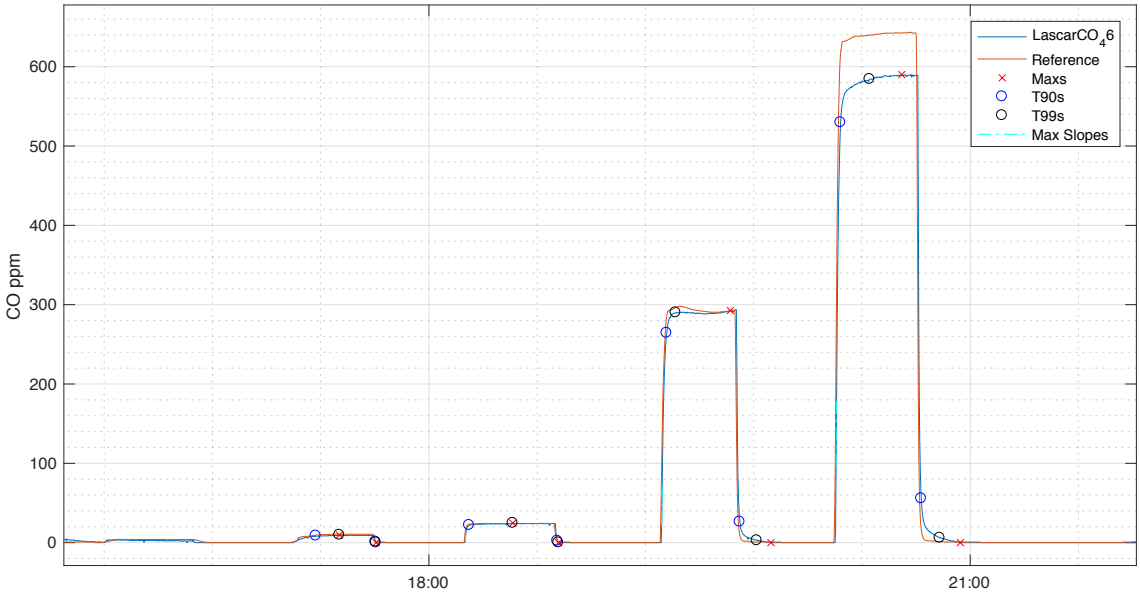

Figure 6-1. Typical calibration time series, shown along with the calculated response times to CO inputs.

The Lascar monitors were weighed three times over the test period to identify whether a significant change in electrolyte mass had occurred in the aged group vs. the control group. A Mettler AE163 balance with 0.1mg resolution was used to weigh the monitors after removing the batteries, USB dust shields, and external housings. No significant changes in sensor weights

were detected for either group, suggesting that there was no significant drying of the electrolyte throughout our testing period.

6.3. Results

6.3.1. Calibration stability over time

Stability of calibration slopes (Figure 6-2) was assessed with a linear mixed effects model as shown in Equation 6-1 (Burton et al., 1998). S_i represents the calibration curve slope for Lascar *i*, at time point *j,* regressed against the log cumulative exposure *CE* (in ppm-hours), where β_0 is an overall intercept corresponding to the 'fresh' batch of monitors and B represents the 'used' monitors, which are interacted with cumulative exposure here. Cumulative exposure is collinear with time, so it cannot be ruled out that relationships identified with that variable herein are in fact related to a different temporal characteristic that we were unable to control for. Average chamber temperature during testing was not a significant predictor in any of the models presented in this work, so were not kept in the final models. The model used a random intercept α_i on the individual sensors, and a random slope φ_i by cumulative exposure to account for individual variations between units and repeated measurements. Throughout this paper, cumulative exposure is calculated in units of ppm-hours, using the raw Lascar signal to maintain independence from calibration approach. Linear modeling assumptions of homoscedasticity, independence, and normality of residuals were met for this and the other models presented in this paper.

$$S_{ijk} = \beta_0 + \beta_1 log(CE_{ij})B_k + \varepsilon_{ijk} + \alpha_i + \varphi_i \qquad \text{Equation 6-1}$$

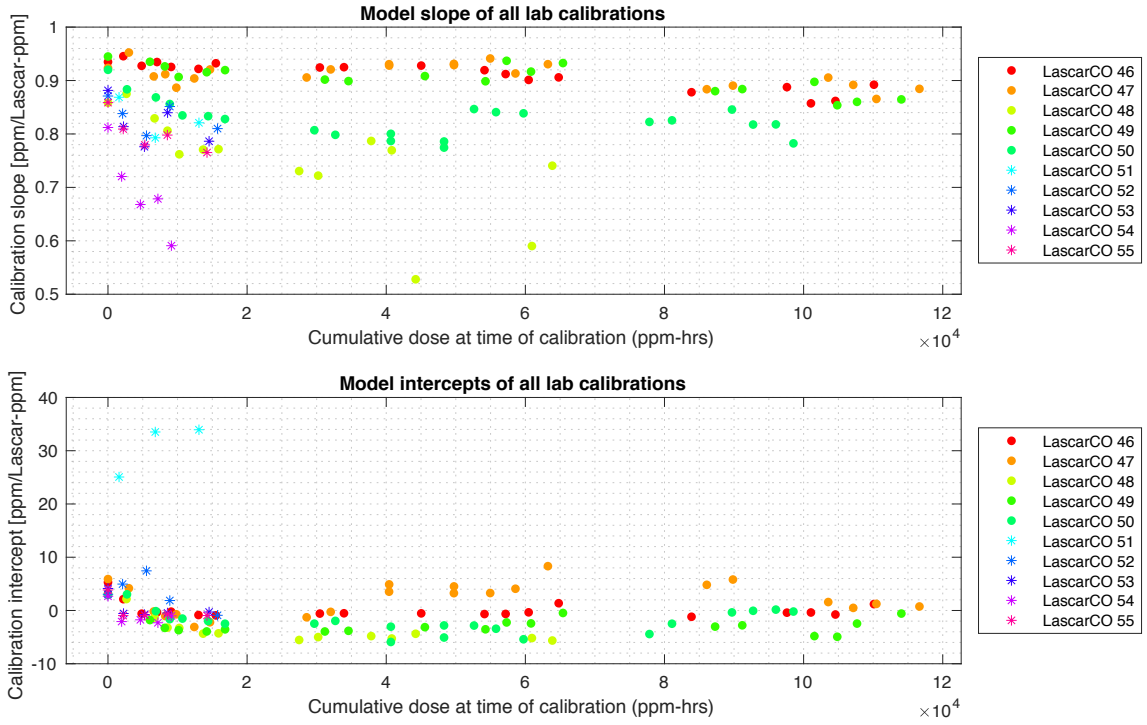

Figure 6-2. Calibration sensitivities (regression slopes) for all Lascar monitors plotted against cumulative exposure experienced by the Lascar sensors in their lifetimes. The star symbols represent Lascar monitors from the 'fresh' batch.

Results from Equation 6-1 indicated that overall, there was a significant reduction in sensitivity over time of 1.63e-6 [ppm/Lascar ppm] per unit of cumulative exposure in ppm-hr (Appendix 5 Table A5.1). The 'used' set of five Lascar monitors (used more frequently) exhibited better calibration sensitivity stability than the 'fresh' batch (-1.01e6 vs. -5.75e6 [ppm/Lascar ppm] per unit of cumulative exposure), and the offset was greater for the 'fresh' batch.

Calibration intercepts are also presented in Figure 6-2, showing that there can be substantial offset, both positive or negative. This variability is indicative of low sensitivity at low CO levels, and could lead to substantial relative error in clean environments. The intercepts of Lascar 51 are very high, and represent poor fits from calibrations as the later measurements were very noisy. Of all laboratory calibration tests, 89% of trials successfully recorded data, with 55% of failures coming from a single unit, Lascar 48.

6.3.2. Average exposure error over time

To simulate a real-world sensor deployment, we modeled average percent error (*PE*) of average exposure using sensor data prepared with three different calibration approaches, while controlling for the log of cumulative sensor exposure and sensor batch (Equation 6-2).

$$PE_{ijk} = \beta_0 + \beta_1 log(CE_{ij})(B_k) + \varepsilon_{ijk} + \alpha_{i+}\varphi_i \qquad \text{Equation 6-2}$$

Average percent error was calculated as error in the mean calibrated sensor mixing ratios relative to the mean reference monitor mixing ratios using an 'exposure test period' from each calibration test. The first calibration (Figure 6-3) approach compared the reference mean exposure with the sensor data calibrated using the nearest calibration available, which represents the gold standard for personal exposure measurement calibration. The second approach compared the mean reference data with mean sensor data calibrated using only the first and last available calibration ('pre/post calibration'), and a linear interpolation between them, correcting for total cumulative exposure for the given monitor, as measured by the data directly output by each monitor (raw data). The third approach directly compared the mean reference data with the raw and uncorrected Lascar data. In all three cases, the baseline was shifted to zero, reflecting a practice that can be performed in the field even without gas standards, and can reduce bias. A random intercept and slope was included to account for inter-sensor variations and repeated measures.

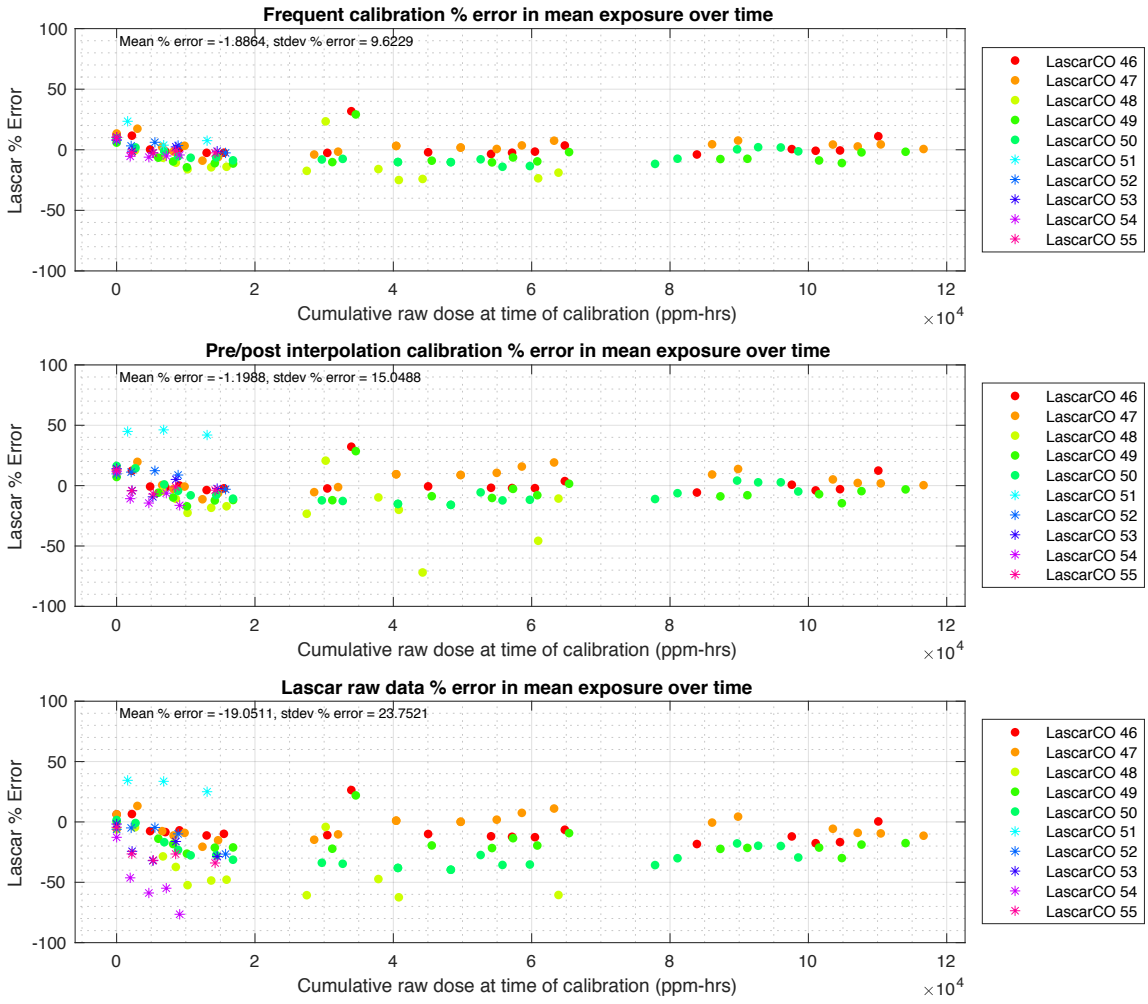

Figure 6-3. Percent error in average exposures using three different calibration approaches, plotted against the cumulative exposure the Lascar has experienced in its lifetime. Stars represent the 'fresh' Lascar batch, and dots represent the 'frequently used' batch.

The 'nearest calibration approach' yielded an overall average of -1.9% in average error during the exposure periods (standard deviation = 9.6%). From the results of Equation 6-2, the 'fresh' batch had -5.8% average error ($p<0.05$), and the 'used' batch had an average error of -+1.8% ($p<0.05$). There was no significant change in percent error with cumulative exposure, indicating that average changes in sensor behavior over time were corrected for by using frequent calibration. However, the random intercept and slopes by Lascar monitor were significant, showing that between-monitor variability of calibration drift must be considered when calibration devices. Complete model output for all models is presented in Appendix 5 Table A5.1.

The 'pre/post' approach had an average percent error of -1.2%, nominally better than the previous approach, but at a cost of higher variability (±15.1%). Using Equation 6-2, there was no significant slope in the error over time, showing that this approach corrected for calibration drift satisfactorily, but there was a significant offset of -10.7% for the 'fresh' batch ($p<0.05$), indicative of an exposure overestimation for that batch. The 'used' batch error was +0.8% relative to the reference data.

The raw Lascar data error was -19.1% on average (±23.8%), relative to the reference data. The error over time slope was -1.6e-3 [% error/cumulative ppm-hr] ($p = 0.09$). There were no significant differences by batch, likely due to the high variability in the data.

6.3.3. Transient responses characteristics

There were three dynamic parameters of interest in our work, 1) rise time to respond to an input of CO, 2) decay time going from elevated CO to clean air, and 3) the maximum slope when stabilizing in response to an input of CO. Rise and decay response times to 90% of the steady state target mixing ratios (T_{90}) were calculated at four mixing ratios for each calibration (5-15ppm, 20-40ppm, 250-350ppm, 550-650ppm). Response times were calculated as the time to 90% of the maximum observed peak value, or zero value, starting when four consecutive slopes were positive, or negative. Response rise and decay times, with means of 183.8s (±91.2s) and 103.2s (±52.9s), respectively, were generally higher than the '<30s' listed in the sensor's data sheet (Nemoto, 2016), and higher than those seen in Cheng et al. (2010) or Buck et al. (2013). This likely due to the 24-second time to steady state of our chamber, so this data does not represent a true measurement of response time. Rather, because our experimental set up

remained constant over time, we can make claims about this pseudo-response time.

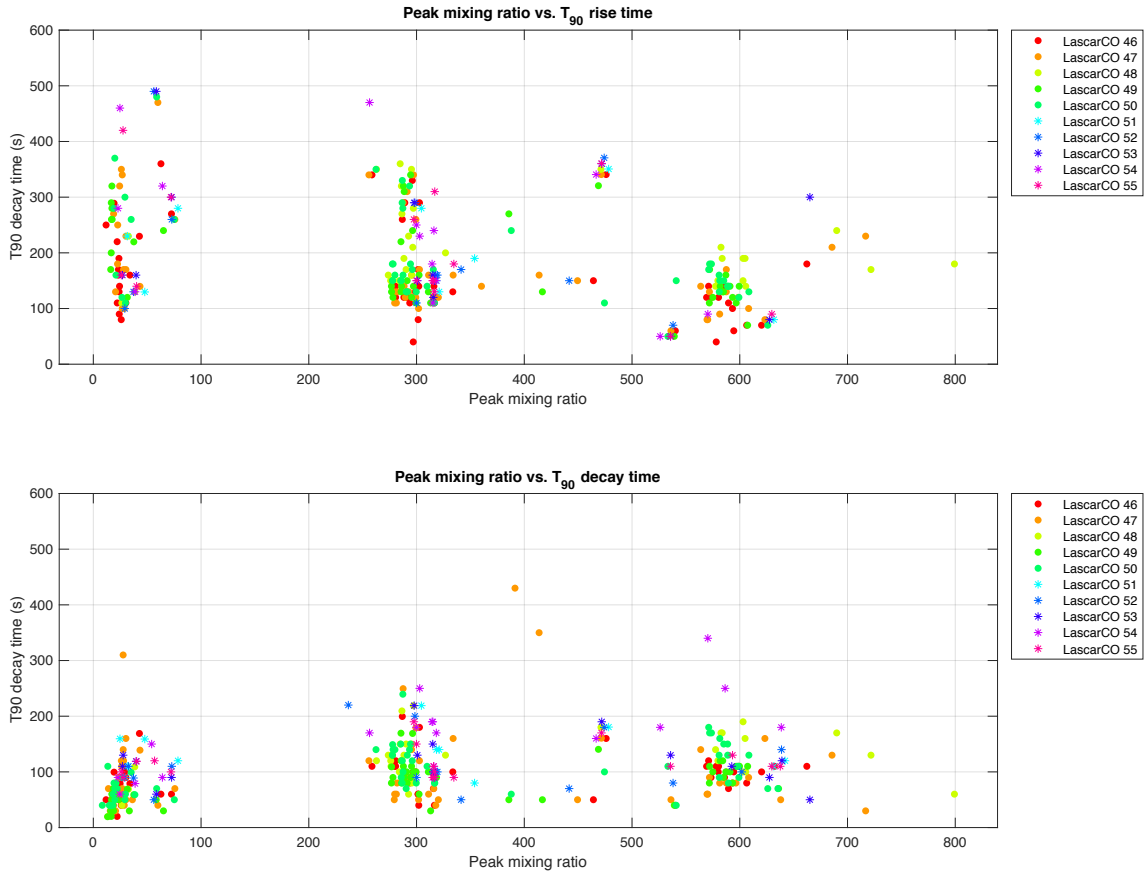

Figure 6-4. Time to steady state for the Lascar sensors, plotted against the peak mixing ratio being approached or departed. Rise time had less structure and more variability than decay time.

Using Equation 6-3, the log of rise times and decay times τ for sensor *i* on trial *j* were regressed against the cumulative exposure *CE* for sensor *i* on trial *j* interacted with the log of the mixing ratio peak *CP* that the measurement was rising to or decaying from, once again controlling for batch *B* and including a random intercept and slope for the individual sensors. The rise and decay was analyzed separately to determine if there was any change in sensitivity as the sensors experienced more exposure.

$$log(\tau_{ijk}) = \beta_0 + \beta_1 log(CP_{ij})(CE_{ij})(B_k) + \varepsilon_{ijk} + \alpha_l + \varphi_i \qquad \text{Equation 6-3}$$

The model for rise time using Equation 6-3 had a -13.7% decrease in response time with an increase of one in the log of peak mixing ratio ($p < 0.01$, an increase of 0.0006% ($p = 0.046$) with cumulative exposure at the time of testing, and a slight (0.0002%, $p < 0.01$) decrease in

response time due to the interaction between peak exposure and with cumulative exposure (Appendix 5 Table A5.2). There was no effect by batch, though the random intercept and slope by sensor were both significant.

The model for decay time using Equation 6-3, had the opposite response with respect to peak mixing ratio. The response time was 11.1% greater for every increase in the log of peak mixing ratio of one ($p = 0.055$), and was 0.0004% greater for every decrease of one in cumulative exposure in ppm-hrs. The 'used' batch had an 83.5% faster response time than the 'fresh' batch. The average daily CO cumulative exposure seen in the REACCTING study for participants cooking over traditional biomass fires was 25.2 ppm-hr (Piedrahita et al., 2017b, *submitted*). At this rate, even 100 days of such exposures should only result in an increase of 1.5% in the response time of the sensors. This is substantially less than we observed in some calibrations, suggesting that the primary driver was not drying or exposure to high concentrations as we have exposed the sensors to in these tests, but rather particulate filter fouling affecting diffusion rates, or sensor damage that slowed the reaction rates in ways we have not observed here.

The characteristic times of the rise were higher than those of the decays here, especially for the lower concentration peaks. We have not identified any reason for our gas delivery system to be responsible for this, and believe it to be a real effect. This effect would be expected to thus reduce the average exposure estimate to an extent. It would be of great future interest to apply the temporal correction of Cheng et al. (2010) to identify the magnitude and change in average exposure over time due to these time responses.

We also investigated the relationship between the maximum slope observed during rise due to a step in the mixing ratio the sensors were exposed to, and the magnitude of that step (Figure 6-5). Maximum slope was calculated as the largest consecutive difference between the 10-second samples during the approach to those mixing ratios. This was modeled using Equation 6-4, regressing the maximum slope (S_{max}) against the mixing ratio of the peak *MRP* that was being approached, interacted with cumulative exposure *CE* and batch *B*. Characteristic time responses have been used to correct for time lags in electrochemical CO sensors

previously (Cheng et al., 2010), and could improve exposure estimates if implemented, if the characteristic times are known.

$$S_{max,ikj} = \beta_0 + \beta_1(MRP_{ij})(CE_{ij})(B_k) + \varepsilon_{ijk} + \alpha_l + \varphi_i \qquad \text{Equation 6-4}$$

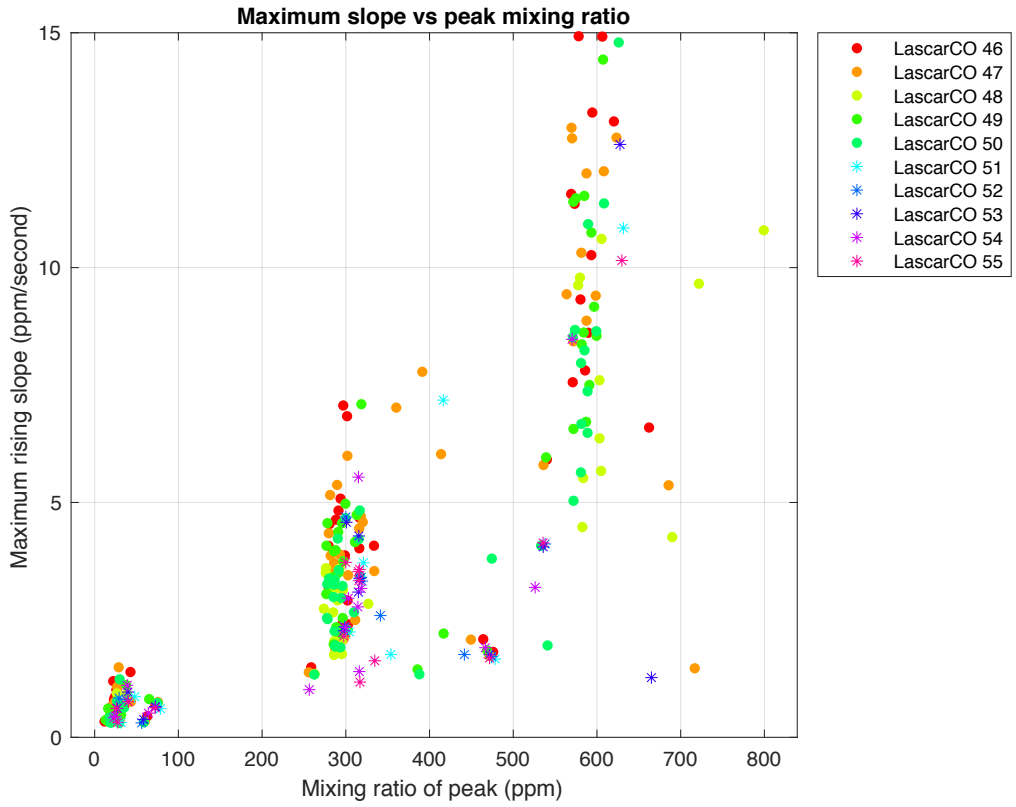

Figure 6-5. Maximum slope [ppm/second] during approach to an elevated CO mixing ratio.

Similar to Buck et al. (2013), we found a significant association between maximum slope during the approach to a peak, and the magnitude of the peak that was being approached ($p<0.01$, 104.6% higher maximum slope per unit log(mixing ratio peak). The log of the cumulative exposure experienced by the sensor was independently responsible for -0.0005% decrease in maximum slope per ppm-hr, but this was not significant ($p = 0.1$), corroborating the observed reduction in response times from Model 2. There was no difference by batch.

6.4. Conclusions

The identified sensor behaviors may help improve personal exposure assessments, at varying costs. Calibrating as frequently as possible is ideal, because although a pre/post

calibration performed quite well relative to the daily calibrations, using only 2/23 of the calibrations, there would be risk of losing a post-calibration due to instrument failure. Frequent calibration can also help identify when a sensor stops responding sufficiently quickly, and may be introducing bias to the measurement, or if it is simply logging zero data erroneously. Implementing a system to correct for time responses will improve accuracy, but may be too burdensome to be practical in the field. It may be preferable to simply stop use of Lascars after a preset period, or cumulative exposure, and remove them from circulation in a piecewise manner so as to reduce any longitudinal bias that could be introduced by swapping entire batches. We were not able to identify a maximum cumulative exposure value, or lifetime in this work, and as of now, this value simply depends on calibration and responsiveness performance when the sensor is being used for exposure measurements in the field. Indeed, the manufacturer claims a sensor lifetime of 7 years, which most instruments used in the field would never reach intact.

CHAPTER 7

CONCLUSIONS

This work helped assess key links of the causal chain (Figure 7-1) from a cookstove intervention to human and environmental health, and developed improved methods to do so. Measurement of cookstove use with electronic monitors, in concert with survey data, frames the discussion of replacement and expected uptake in the event of more widespread distribution of portable improved cookstoves. The accompanying air quality measurement campaigns helped gauge the intervention effectiveness, and increased our understanding of the importance of different air pollution sources to personal exposure, and to regional air quality.

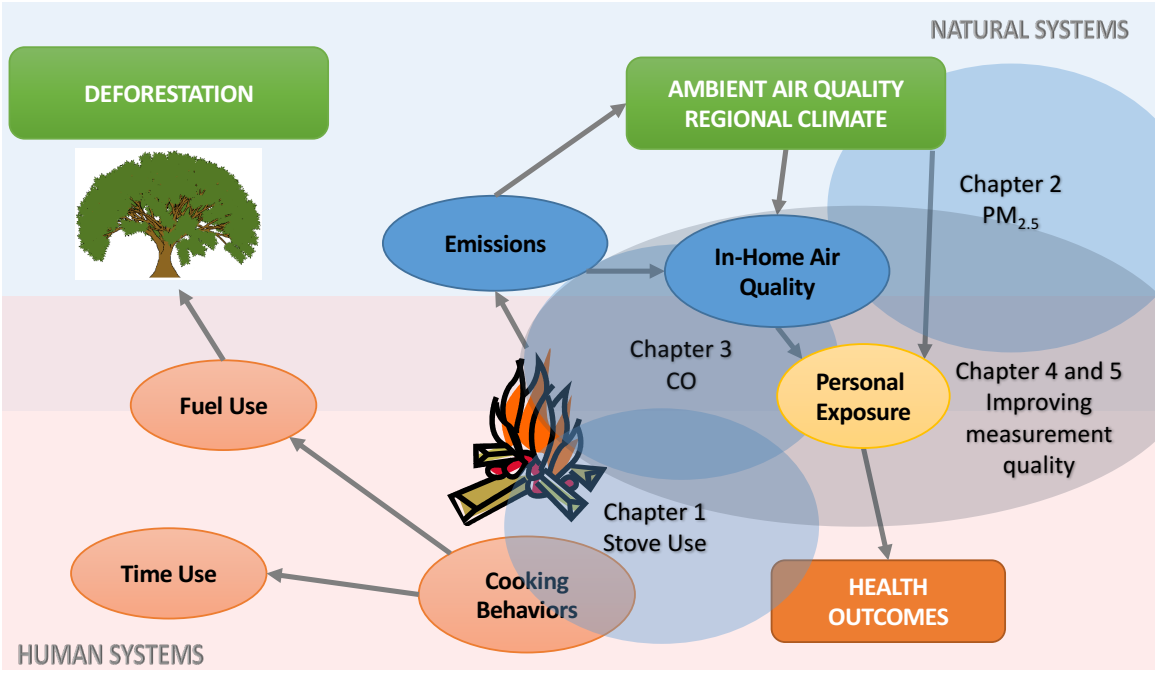

Figure 7-1. Causal chain from cooking behaviors to health and environmental outcomes, updated with regions of the relationship addressed by this dissertation.

Returning to the questions posed in the introduction: Will the population consistently and exclusively use an improved cooking technology if given the opportunity? We observed substantial use of the intervention stoves but only moderate reductions in traditional cookstove use, supporting the non-linear view of the energy-ladder theory. Given this, will real-world use of the improved cookstoves result in exposure reductions of components of woodsmoke? We

did observe reductions in CO and carbonaceous $PM_{2.5}$ exposure at varying magnitudes and significance levels, providing evidence that the selected cookstoves may be beneficial to the region and may be in precisely the regime of dose-response relationships that may provide significant reductions in health effects such as pneumonia.

Is woodsmoke from cooking in fact the most significant source of pollution impacting personal exposure, or are there other important sources and behaviors associated with exposure? Evidence on this is mixed, as source apportionment of $PM_{2.5}$ found that sources related to biomass combustion made up only 9.2% of OC and 15.3% of EC to personal and cooking area microenvironment samples, while 64.5% of CO exposure was incurred at home, according to the Beacon data. These results are from different stages of the intervention, and are certainly not in conflict, and may suggest that exposures at home from sources unrelated to biomass are important, and that exposure when away from home is substantial.

Future work should consider real-time personal $PM_{2.5}$ measurement, in conjunction with BLE Beacon measurement. $PM_{2.5}$ remains the most-studied pollutant from biomass cookstove smoke, due to its strong causal links with negative health effects. Additionally, PM measurement instrument sizes and costs have dropped since the start of the REACCTING project, and they may present fewer logistical challenges than gas measurement, due to more stable calibrations. Most wearable PM monitors rely on optical measurements, which are more easily maintained, only requiring cleaning and zeroing periodically, whereas gas sensors need periodic multipoint calibrations to known standards, often challenging and costly to do in rural areas. The simultaneous measurement of real-time PM and Beacon data, ideally in addition to thermocouple stove usage monitoring, would lend high confidence to exposure study results at a relatively low cost. Such a data stream would provide a rich vein of information on behaviors and exposures that this thesis has provided a path to analyze.

Further work to measure fuel use over time using kitchen performance tests would be beneficial to understand the environmental co-benefits of the intervention. Also, although improved fuel efficiency was observed during uncontrolled cooking tests as part of this study, the sample size was small and not necessarily representative of standard home use. Despite this worry, a household level reduction in average fuel use is supported by the observed

reductions in exposures in the intervention households due to the magnitude of the effects of OC for example, vs. the observed efficiency improvements. Further economic assessments that help build a foundation for a cookstove market to develop in the region are eagerly awaited, and stand on firm footing as to the potential benefits to the community.

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APPENDIX 1

Assessment of cookstove stacking in Northern Ghana using surveys and stove use monitors

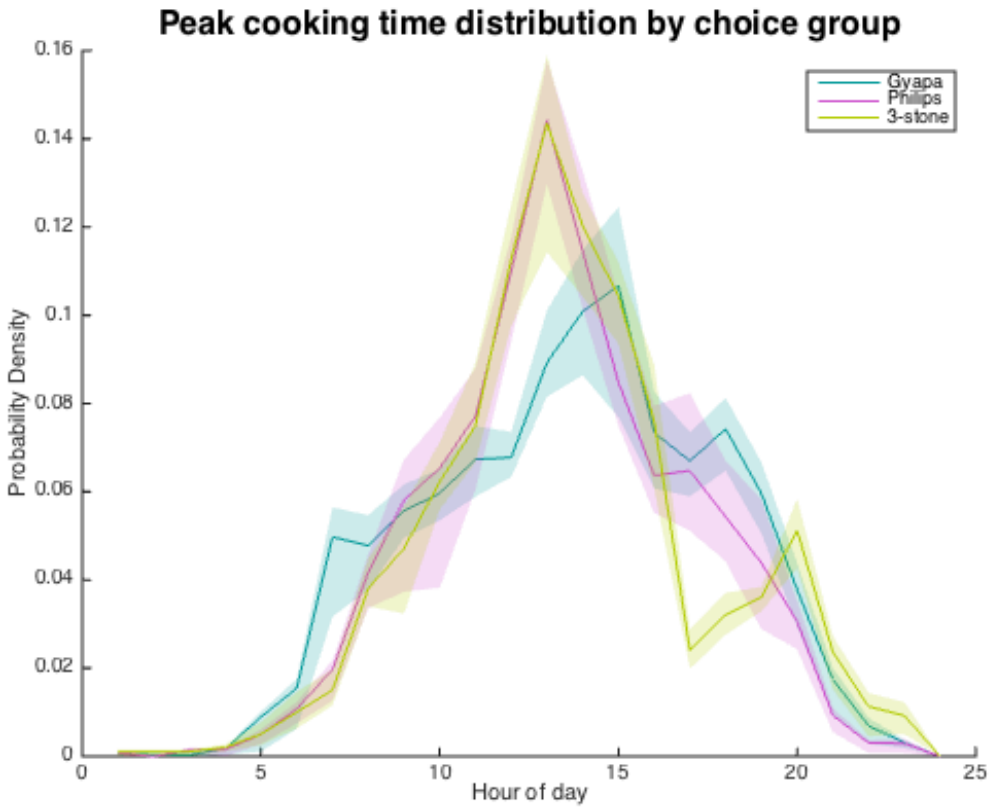


Figure A1-1 Gyapa/Philips group

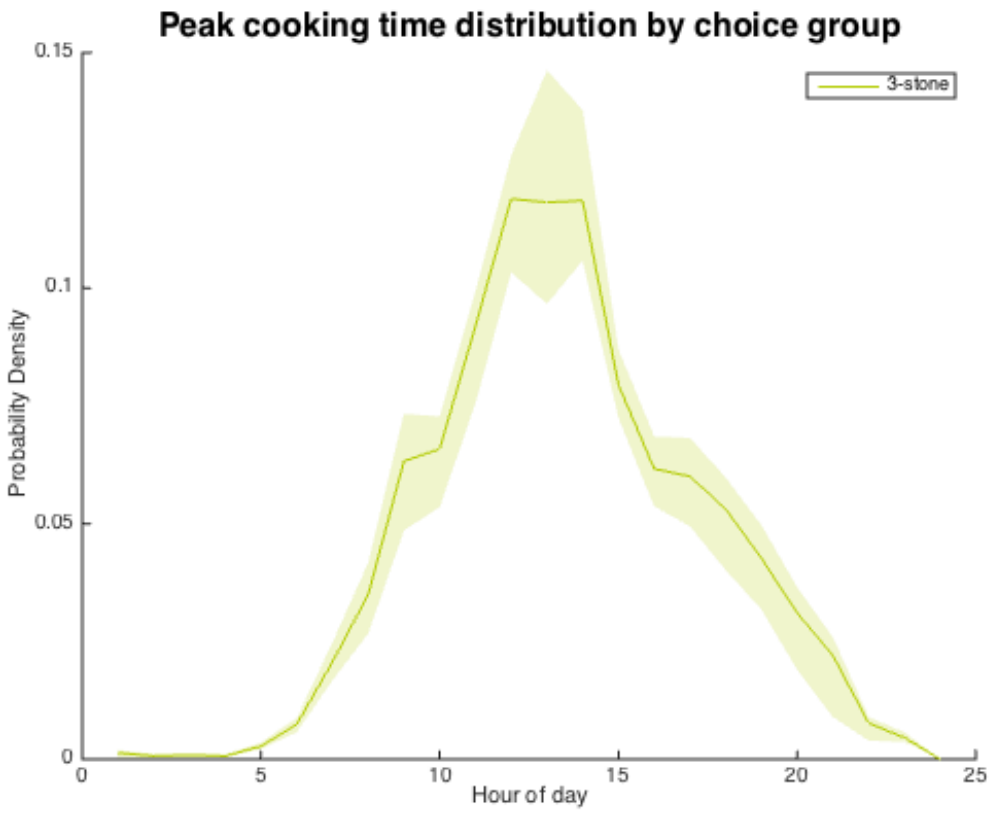


Figure A 1-2 Control group.

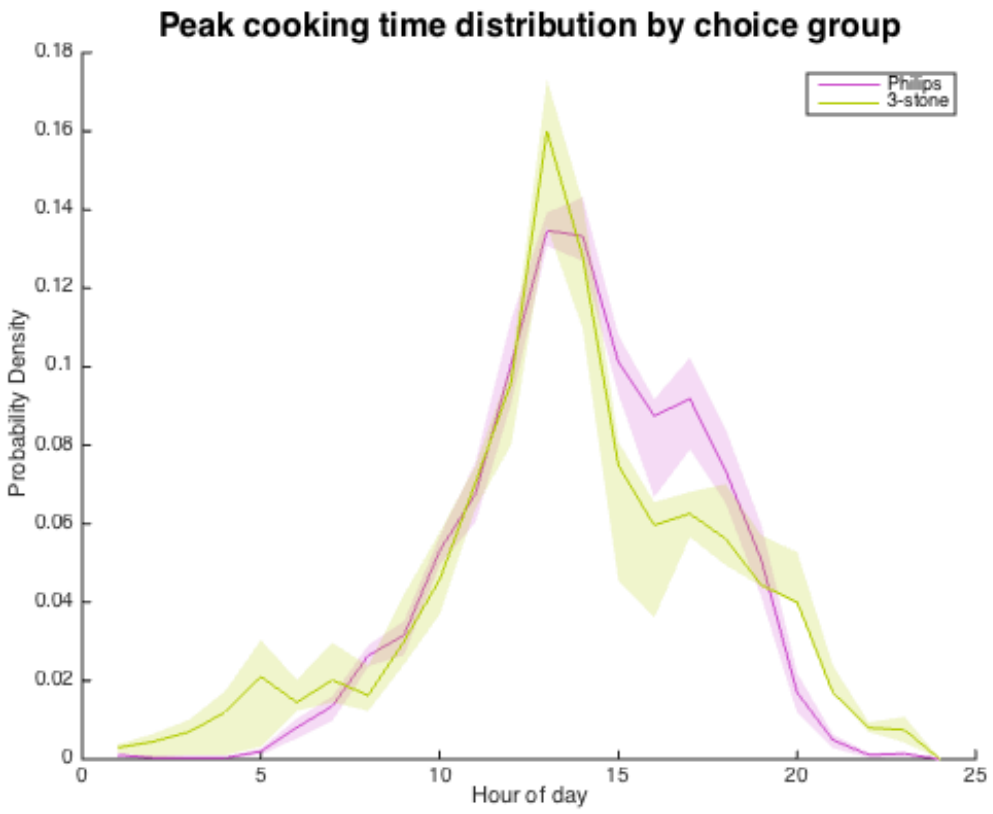


Figure A 1-3. Philips/Philips group

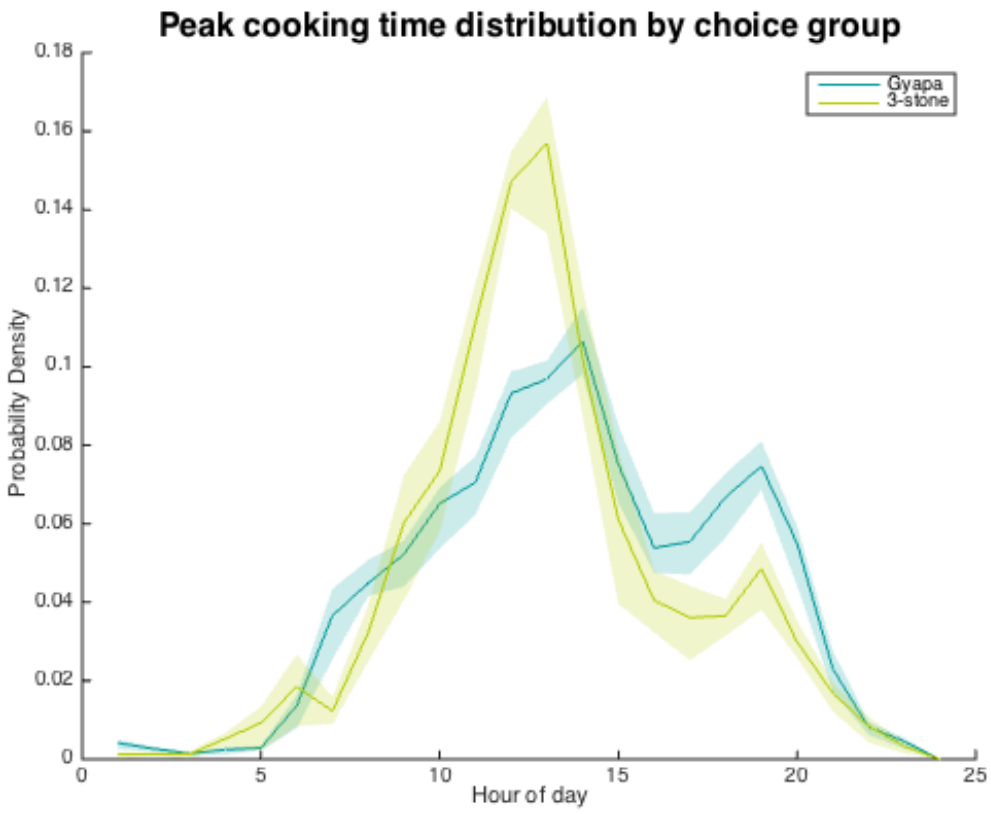


Figure A 1-4. Gyapa/Gyapa group.

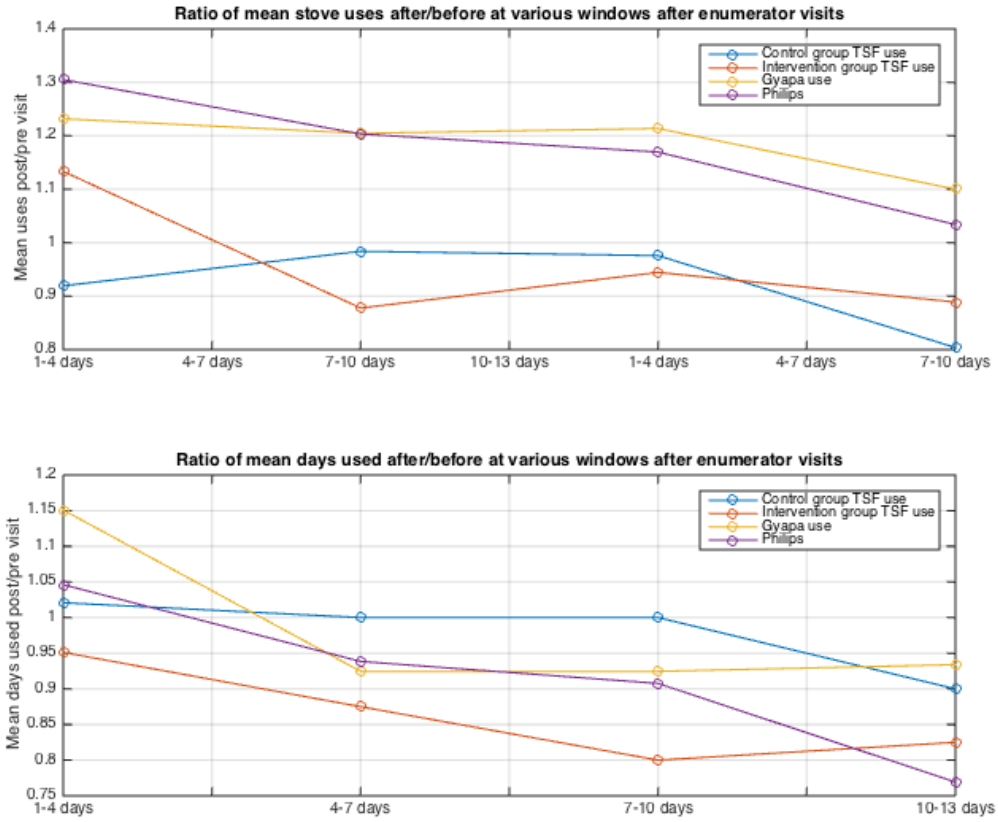


Figure A 1-5 Reactivity to enumerator visits. Ratio of use in sliding 3-day windows after the visit over the 3 days before the visit was scheduled. Above shows this data by number of uses in the 3-day window, while below shows ratio by days used.

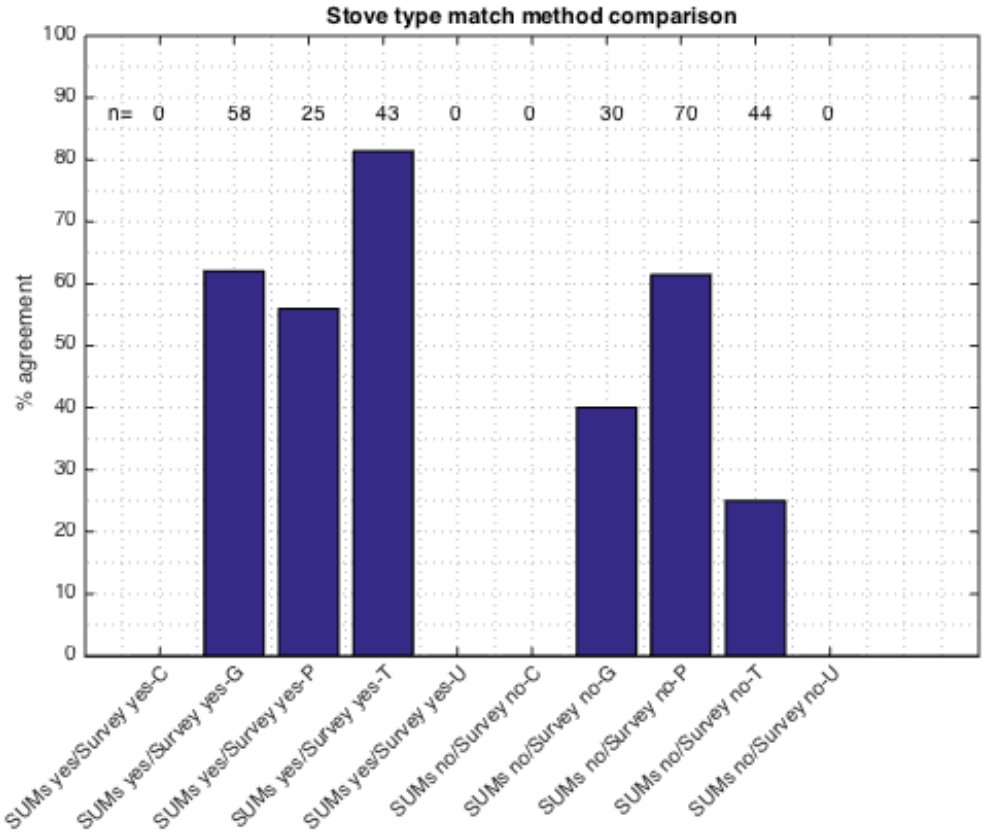


Figure A 1-6. Agreement between SUM and survey results on whether a stove was used 'yesterday'.

APPENDIX 2

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

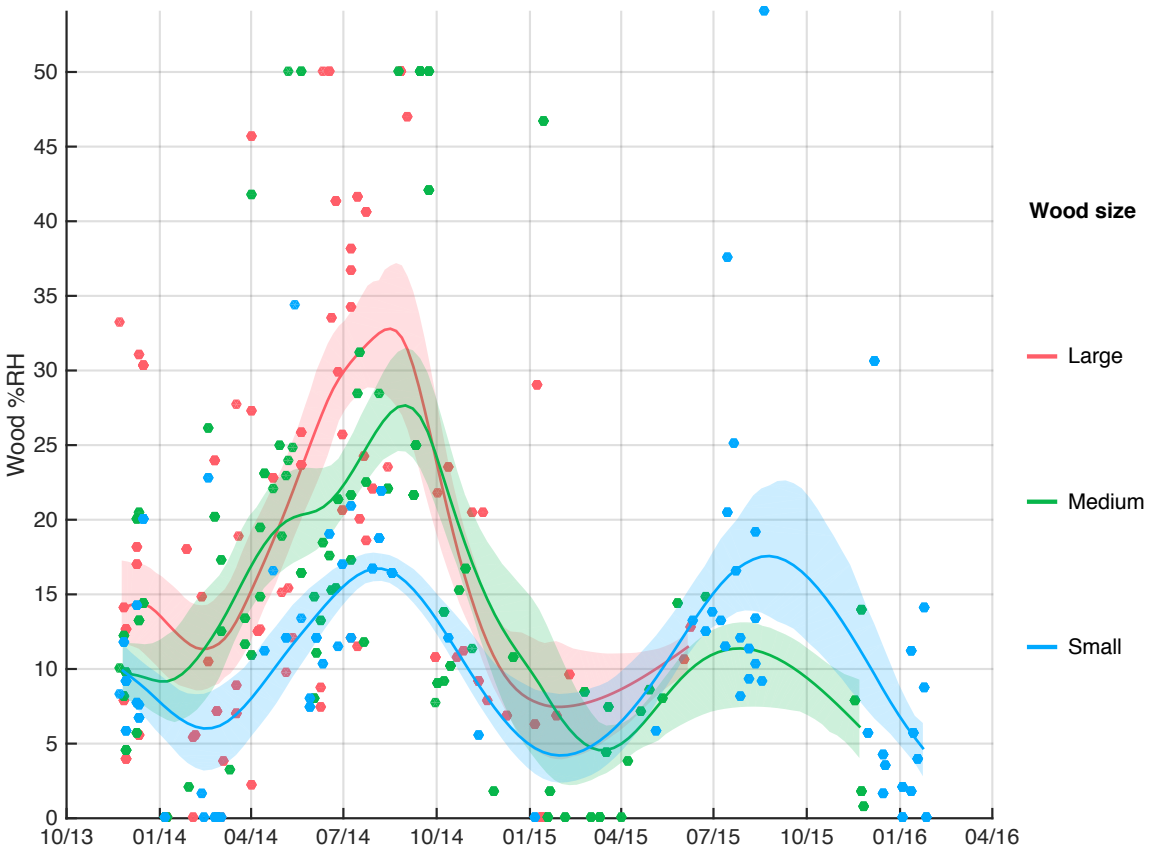


Figure A 2-1. Wood fuel moisture content as measured at households during personal exposure sampling periods. Note the elevated during the rainy seasons from April to November on both years. The trend line is a penalized b-spline with 95% confidence intervals, computed with the GRAMM package for MATLAB.

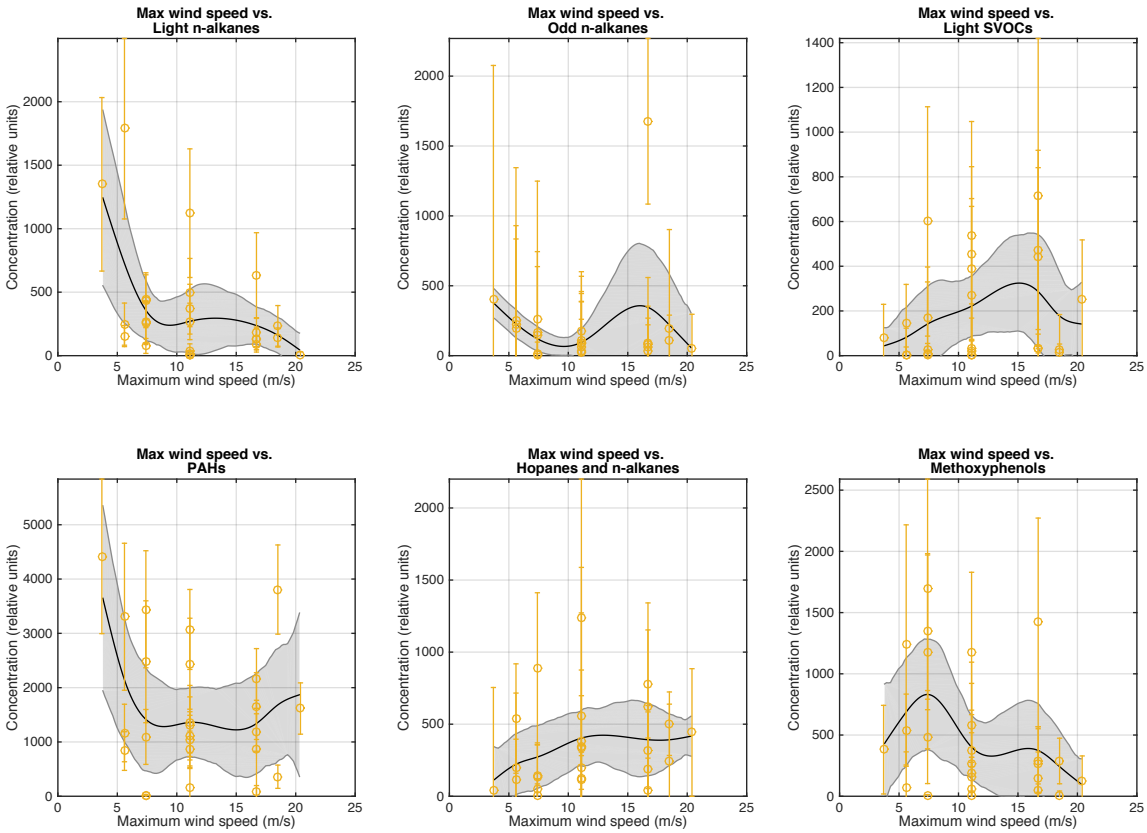


Figure A 2-2. Mean wind speeds from the PM2.5 sampling periods vs. the factor contributions in relative concentration units. The trend line is a penalized b-spline with 95% confidence intervals, computed with the GRAMM package for MATLAB.

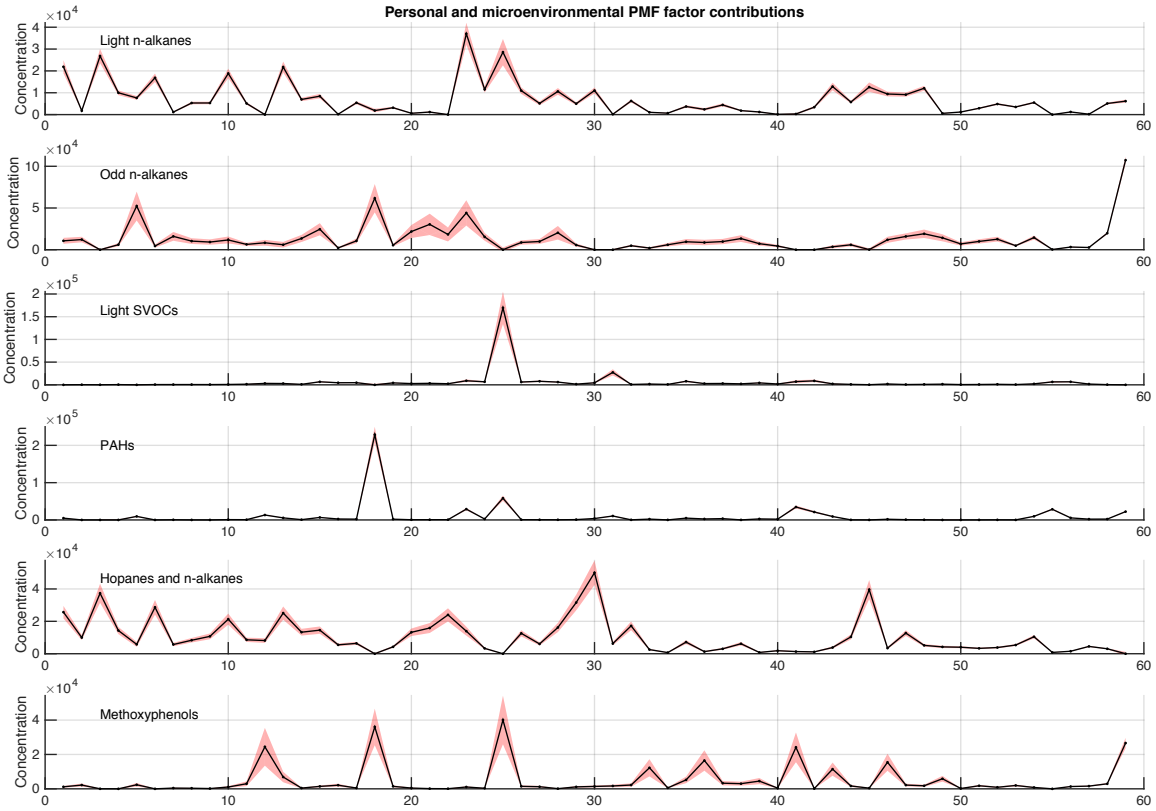


Figure A 2-3. 6-factor solution contributions for the personal and microenvironmental EC/OC and organics data set.

A2.1. Mixed effects model results for personal exposure to OC

Linear mixed-effects model fit by ML

Model information:

Number of observations	190
Fixed effects coefficients	17
Random effects coefficients	88
Covariance parameters	2

Formula:

OCLOG ~ 1 + StoveGroup + familymembers + primarycook + GENDERAGECAT + season + SES + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
566.57	628.26	-264.28	528.57

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	4.1748	0.41592	10.038	173	5.6796e-19	3.3539	4.9958
'StoveGroup_C'	-0.99978	0.34955	-2.8602	173	0.0047556	-1.6897	-0.30984
'StoveGroup_B'	-0.68192	0.26237	-2.5991	173	0.010154	-1.1998	-0.16407
'StoveGroup_A'	-0.85045	0.30684	-2.7716	173	0.0061881	-1.4561	-0.24482
'familymembers'	-0.015671	0.025041	-0.6258	173	0.53227	-0.065096	0.033754
'primarycook_1'	0.47211	0.27575	1.7121	173	0.088675	-0.072163	1.0164
'GENDERAGECAT_F1'	-0.70665	0.36753	-1.9227	173	0.056159	-1.4321	0.018766
'GENDERAGECAT_M0'	-0.30457	0.35375	-0.86096	173	0.39045	-1.0028	0.39366
'GENDERAGECAT_M1'	-0.25163	0.46087	-0.54598	173	0.58578	-1.1613	0.65803
'season_Heavy_Rainy'	-0.42401	0.16544	-2.563	173	0.01123	-0.75054	-0.097472
'season_Hot_dry'	-0.13148	0.22652	-0.58041	173	0.56239	-0.57858	0.31562
'season_Light_Rainy'	-0.4229	0.23805	-1.7765	173	0.0774	-0.89276	0.046949
'season_Other'	-0.37198	0.25927	-1.4347	173	0.15317	-0.88372	0.13976
'SES_Poorer'	0.52092	0.25798	2.0192	173	0.04501	0.011719	1.0301
'SES_Less_poor'	-0.23748	0.3233	-0.73454	173	0.46361	-0.87559	0.40064
'SES_Least_poor'	0.37819	0.31037	1.2185	173	0.22469	-0.23441	0.99079
'SES_Poorest'	0.78795	0.37097	2.124	173	0.03509	0.055738	1.5202

Random effects covariance parameters (95% CIs):

Group: INDIVID (88 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.50395	0.2796	0.90832

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.8621	0.73314	1.0137

A2.2. Mixed effects model results for personal exposure to EC

Linear mixed-effects model fit by ML

Model information:

Number of observations	190
Fixed effects coefficients	17
Random effects coefficients	88
Covariance parameters	2

Formula:

ECLOG ~ 1 + StoveGroup + familymembers + primarycook + GENDERAGECAT + season + SES + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
630.33	692.02	-296.16	592.33

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	0.9109	0.44235	2.0592	173	0.040972	0.037796	1.784
'StoveGroup_C'	-0.89909	0.36838	-2.4406	173	0.01567	-1.6262	-0.17198
'StoveGroup_B'	-0.967	0.26596	-3.6359	173	0.00036523	-1.492	-0.44205
'StoveGroup_A'	-0.58453	0.31816	-1.8372	173	0.067895	-1.2125	0.043449
'familymembers'	0.026129	0.026525	0.98507	173	0.32597	-0.026225	0.078483
'primarycook_1'	0.27462	0.28926	0.94938	173	0.34375	-0.29632	0.84556
'GENDERAGECAT_F1'	-0.24108	0.38745	-0.62223	173	0.53461	-1.0058	0.52365
'GENDERAGECAT_M0'	-0.22794	0.37618	-0.60594	173	0.54535	-0.97043	0.51455
'GENDERAGECAT_M1'	0.36721	0.4902	0.7491	173	0.45481	-0.60033	1.3347
'season_Heavy_Rainy'	-0.70584	0.20645	-3.4188	173	0.00078427	-1.1133	-0.29834
'season_Hot_dry'	-0.79192	0.28078	-2.8204	173	0.0053564	-1.3461	-0.23772
'season_Light_Rainy'	-1.9591	0.29334	-6.6788	173	3.1604e-10	-2.5381	-1.3801
'season_Other'	-0.67674	0.31655	-2.1379	173	0.03393	-1.3015	-0.051949
'SES_Poorer'	0.32202	0.26567	1.2121	173	0.22713	-0.20236	0.8464
'SES_Less_poor'	-0.32176	0.33258	-0.96747	173	0.33466	-0.97819	0.33467
'SES_Least_poor'	0.19924	0.32307	0.61671	173	0.53824	-0.43843	0.8369
'SES_Poorest'	0.69911	0.39533	1.7684	173	0.078753	-0.081181	1.4794

Random effects covariance parameters (95% CIs):

Group: INDIVID (88 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.3097	0.087633	1.0945

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.1105	0.97324	1.267

A2.3. Mixed effects model results for cooking area OC

Linear mixed-effects model fit by ML

Model information:

Number of observations	86
Fixed effects coefficients	9
Random effects coefficients	34
Covariance parameters	2

Formula:

microOCLOG ~ 1 + StoveGroup + familymembers + season + (1 | COMPOUND)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
253.39	280.39	-115.69	231.39

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	4.6067	0.4397	10.477	77	1.7936e-16	3.7312	5.4823
'StoveGroup_C'	-0.55785	0.41372	-1.3484	77	0.18149	-1.3817	0.26598
'StoveGroup_B'	-0.7018	0.33828	-2.0746	77	0.04136	-1.3754	-0.02821
'StoveGroup_A'	-1.1012	0.3468	-3.1753	77	0.0021528	-1.7918	-0.41064
'familymembers'	0.0012941	0.03317	0.039014	77	0.96898	-0.064756	0.067344
'season_Heavy_Rainy'	0.56002	0.29149	1.9213	77	0.058398	-0.020398	1.1404
'season_Hot_dry'	-0.5673	0.37416	-1.5162	77	0.13357	-1.3123	0.17775
'season_Light_Rainy'	-0.21004	0.33942	-0.61882	77	0.53786	-0.8859	0.46583
'season_Other'	1.0003	0.36075	2.7728	77	0.0069683	0.28193	1.7186

Random effects covariance parameters (95% CIs):

Group: COMPOUND (34 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.33235	0.12007	0.91992

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.87536	0.72641	1.0549

A2.4. Mixed effects model results for cooking area EC

Linear mixed-effects model fit by ML

Model information:

Number of observations	86
Fixed effects coefficients	9
Random effects coefficients	34
Covariance parameters	2

Formula:

microECLOG ~ 1 + StoveGroup + familymembers + season + (1 | COMPOUND)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
300.16	327.16	-139.08	278.16

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	1.876	0.53597	3.5001	77	0.00077667	0.80872	2.9432
'StoveGroup_C'	-0.24647	0.46945	-0.52501	77	0.60108	-1.1813	0.68834
'StoveGroup_B'	-0.67518	0.38951	-1.7334	77	0.087022	-1.4508	0.10042
'StoveGroup_A'	-0.44522	0.40709	-1.0937	77	0.27751	-1.2558	0.3654
'familymembers'	-0.058029	0.039532	-1.4679	77	0.1462	-0.13675	0.020688
'season_Heavy_Rainy'	0.63824	0.39711	1.6072	77	0.1121	-0.15249	1.429
'season_Hot_dry'	-0.81885	0.50787	-1.6123	77	0.11098	-1.8301	0.19244
'season_Light_Rainy'	-1.0274	0.45452	-2.2603	77	0.026629	-1.9324	-0.12228
'season_Other'	1.0593	0.48149	2.2001	77	0.030796	0.10057	2.0181

Random effects covariance parameters (95% CIs):

Group: COMPOUND (34 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	3.1419e-09	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper

'Res Std' 1.2193 1.05 1.4158

A2.5. Mixed effects model results for personal exposure vs. cooking area microenvironment OC

Linear mixed-effects model fit by ML

Model information:

Number of observations	141
Fixed effects coefficients	13
Random effects coefficients	73
Covariance parameters	2

Formula:

OCLOG ~ 1 + familymembers + GENDERAGECAT + season + microOCLOG + StoveGroup:microOCLOG + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
376.8	421.03	-173.4	346.8

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	2.2674	0.4499	5.0397	128	1.554e-06	1.3772	3.1576
'familymembers'	0.028669	0.022446	1.2773	128	0.20382	-0.015744	0.073082
'GENDERAGECAT_F1'	0.063429	0.27036	0.23461	128	0.81489	-0.47152	0.59838
'GENDERAGECAT_M0'	-0.12801	0.31694	-0.40389	128	0.68697	-0.75512	0.4991
'GENDERAGECAT_M1'	0.3582	0.37367	0.95861	128	0.33956	-0.38116	1.0976
'season_Heavy_Rainy'	-0.43406	0.19701	-2.2032	128	0.029371	-0.82388	-0.044232
'season_Hot_dry'	0.048375	0.26809	0.18044	128	0.85709	-0.48209	0.57884
'season_Light_Rainy'	-0.25541	0.25386	-1.0061	128	0.31625	-0.75771	0.24689
'season_Other'	-0.51697	0.27623	-1.8715	128	0.063555	-1.0635	0.029597
'microOCLOG'	0.22606	0.077792	2.906	128	0.0043157	0.072136	0.37999
'StoveGroup_C:microOCLOG'	0.046827	0.05705	0.8208	128	0.41329	-0.066057	0.15971
'StoveGroup_B:microOCLOG'	-0.040036	0.046039	-0.86962	128	0.38614	-0.13113	0.05106
'StoveGroup_A:microOCLOG'	-0.036951	0.05568	-0.66363	128	0.50812	-0.14712	0.073221

Random effects covariance parameters (95% CIs):

Group: INDIVID (73 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.18847	0.013128	2.7058

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.80681	0.6755	0.96364

A2.6. Mixed effects model results for personal exposure vs. cooking area microenvironment EC

Linear mixed-effects model fit by ML

Model information:

Number of observations	141
Fixed effects coefficients	13
Random effects coefficients	73
Covariance parameters	2

Formula:

ECLOG ~ 1 + familymembers + GENDERAGECAT + season + microECLOG + StoveGroup:microECLOG + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
452.82	497.05	-211.41	422.82

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	-0.36866	0.45686	-0.80693	128	0.4212	-1.2726	0.53533
'familymembers'	0.055599	0.030245	1.8383	128	0.068342	-0.0042466	0.11544
'GENDERAGECAT_F1'	0.28751	0.37008	0.77688	128	0.43867	-0.44476	1.0198
'GENDERAGECAT_M0'	0.0078422	0.43144	0.018177	128	0.98553	-0.84583	0.86151
'GENDERAGECAT_M1'	0.63216	0.50833	1.2436	128	0.21592	-0.37365	1.638
'season_Heavy_Rainy'	-0.6299	0.25833	-2.4384	128	0.016126	-1.141	-0.11875
'season_Hot_dry'	-0.58253	0.35002	-1.6643	128	0.098502	-1.2751	0.11004
'season_Light_Rainy'	-1.7442	0.35996	-4.8457	128	3.5836e-06	-2.4565	-1.032
'season_Other'	-0.5769	0.36662	-1.5736	128	0.11806	-1.3023	0.14853
'microECLOG'	0.25854	0.14547	1.7772	128	0.077903	-0.029302	0.54639
'StoveGroup_C:microECLOG'	-0.0056563	0.17262	-0.032768	128	0.97391	-0.34721	0.33589

'StoveGroup_B:microECLOG'	-0.12607	0.15423	-0.81737	128	0.41524	-0.43124	0.17911
'StoveGroup_A:microECLOG'	-0.35947	0.18143	-1.9814	128	0.049694	-0.71845	-0.00048764

Random effects covariance parameters (95% CIs):

Group: INDIVID (73 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.37198	0.096852	1.4287

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.0237	0.8477	1.2363

A2.7. Mixed effects model results for personal exposure to PMF factor contributions

A2.7.1. OC mixed effect personal exposure model with subset of data having organics available

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

OCLOG ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
82.074	96.572	-32.037	64.074

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	4.2577	0.36939	11.526	30	1.5156e-12	3.5033	5.0121
'StoveGroup_C'	-0.71634	0.32002	-2.2384	30	0.032765	-1.3699	-0.062776
'StoveGroup_B'	-0.4743	0.22839	-2.0767	30	0.046492	-0.94074	-0.0078622
'StoveGroup_A'	-0.48396	0.30587	-1.5822	30	0.12409	-1.1086	0.14072

'familymembers'	-0.025682	0.03057	-0.84009	30	0.4075	-0.088115	0.036751
'primarycook_1'	-0.20473	0.25667	-0.79764	30	0.43135	-0.72891	0.31946
'AGECAT_1'	0.087943	0.28173	0.31215	30	0.75708	-0.48743	0.66332

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.40671	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.40671	NaN	NaN

A2.7.2. EC mixed effect personal exposure model with subset of data having organics available

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

ECLOG ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
107.3	121.8	-44.652	89.303

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	0.87036	0.51946	1.6755	30	0.10423	-0.19051	1.9312
'StoveGroup_C'	-0.59011	0.45003	-1.3113	30	0.19971	-1.5092	0.32897
'StoveGroup_B'	-0.54799	0.32118	-1.7062	30	0.09831	-1.2039	0.10795

'StoveGroup_A'	0.086896	0.43014	0.20202	30	0.84127	-0.79157	0.96536
'familymembers'	0.018299	0.04299	0.42565	30	0.6734	-0.069499	0.1061
'primarycook_1'	-0.11542	0.36094	-0.31977	30	0.75136	-0.85256	0.62173
'AGECAT_1'	0.25735	0.39619	0.64956	30	0.52092	-0.55178	1.0665

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.57194	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.57194	NaN	NaN

A2.7.3. 'Methoxyphenol' factor personal exposure mixed effects model

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

logMethoxyphenols ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
187.94	202.43	-84.968	169.94

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	5.8786	1.5444	3.8063	30	0.00064829	2.7245	9.0328
'StoveGroup_C'	-1.5264	1.338	-1.1408	30	0.26298	-4.259	1.2062

'StoveGroup_B'	-1.5694	0.95492	-1.6435	30	0.11073	-3.5196	0.38084
'StoveGroup_A'	-0.50355	1.2789	-0.39375	30	0.69655	-3.1154	2.1083
'familymembers'	0.051569	0.12782	0.40346	30	0.68947	-0.20947	0.31261
'primarycook_1'	1.5355	1.0731	1.4308	30	0.16281	-0.65615	3.7272
'AGECAT_1'	0.46315	1.1779	0.39319	30	0.69696	-1.9425	2.8688

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	1.7005	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.7005	NaN	NaN

A2.7.4. 'PAH' factor personal exposure mixed effects model

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

logPAHs ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
192.6	207.1	-87.302	174.6

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	6.4429	1.645	3.9167	30	0.00047989	3.0834	9.8024

'StoveGroup_C'	-0.27025	1.4251	-0.18963	30	0.85088	-3.1808	2.6403
'StoveGroup_B'	-4.0027	1.0171	-3.9354	30	0.00045589	-6.0799	-1.9255
'StoveGroup_A'	0.0985	1.3622	0.072312	30	0.94283	-2.6834	2.8804
'familymembers'	0.14731	0.13614	1.082	30	0.28786	-0.13073	0.42534
'primarycook_1'	0.33506	1.143	0.29313	30	0.77144	-1.9993	2.6694
'AGECAT_1'	0.18056	1.2546	0.14392	30	0.88653	-2.3818	2.7429

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	1.8112	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.8112	NaN	NaN

A2.7.5. 'Light n-alkane' factor personal exposure mixed effects model

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

logLight_n_alkanes ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
198.8	213.3	-90.399	180.8

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper

'(Intercept)'	7.0069	1.7886	3.9174	30	0.00047885	3.354	10.66
'StoveGroup_C'	-0.52218	1.5496	-0.33698	30	0.73848	-3.6868	2.6425
'StoveGroup_B'	2.0892	1.1059	1.8891	30	0.068579	-0.16939	4.3477
'StoveGroup_A'	0.28696	1.4811	0.19375	30	0.84768	-2.7378	3.3118
'familymembers'	-0.041089	0.14803	-0.27758	30	0.78324	-0.3434	0.26122
'primarycook_1'	1.3805	1.2428	1.1108	30	0.27549	-1.1577	3.9187
'AGECAT_1'	-1.2146	1.3642	-0.89037	30	0.38035	-4.0007	1.5714

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	1.9694	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.9694	NaN	NaN

A2.7.6. 'Odd n-alkane' factor personal exposure mixed effects model

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

logOdd_n_alkanes ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
186.85	201.35	-84.425	168.85

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	11.324	1.5219	7.4404	30	2.7208e-08	8.2156	14.432
'StoveGroup_C'	-3.0521	1.3185	-2.3148	30	0.027649	-5.7449	-0.35936
'StoveGroup_B'	-0.13073	0.941	-0.13893	30	0.89044	-2.0525	1.7911
'StoveGroup_A'	-1.8253	1.2602	-1.4484	30	0.15788	-4.3991	0.74842
'familymembers'	-0.20634	0.12595	-1.6382	30	0.11182	-0.46357	0.05089
'primarycook_1'	0.99952	1.0575	0.94517	30	0.35212	-1.1602	3.1592
'AGECAT_1'	-1.4924	1.1608	-1.2857	30	0.20839	-3.863	0.87822

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	1.6757	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.6757	NaN	NaN

A2.7.7. 'Light SVOC' factor personal exposure mixed effects model

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

logLight_SVOCs ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
177.61	192.11	-79.806	159.61

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	8.2098	1.3433	6.1115	30	1.0215e-06	5.4663	10.953
'StoveGroup_C'	0.13395	1.1638	0.1151	30	0.90914	-2.2428	2.5107
'StoveGroup_B'	-0.70906	0.83059	-0.85368	30	0.40005	-2.4053	0.98723
'StoveGroup_A'	-0.85567	1.1124	-0.76923	30	0.44777	-3.1274	1.4161
'familymembers'	-0.018466	0.11117	-0.1661	30	0.8692	-0.24551	0.20858
'primarycook_1'	-0.87379	0.93342	-0.93612	30	0.35669	-2.7801	1.0325
'AGECAT_1'	-0.0027845	1.0246	-0.0027177	30	0.99785	-2.0952	2.0897

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	1.4791	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.4791	NaN	NaN

A2.7.8. ‘Hopane and n-alkane’ factor personal exposure mixed effects model

Linear mixed-effects model fit by ML

Model information:

Number of observations	37
Fixed effects coefficients	7
Random effects coefficients	37
Covariance parameters	2

Formula:

logHopanes_and_n_alkanes ~ 1 + StoveGroup + familymembers + primarycook + AGECAT + (1 | INDIVID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
179.8	194.3	-80.901	161.8

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	8.615	1.3837	6.2261	30	7.423e-07	5.7891	11.441
'StoveGroup_C'	0.78099	1.1987	0.65151	30	0.51968	-1.6672	3.2292
'StoveGroup_B'	1.783	0.85553	2.0841	30	0.045769	0.035766	3.5302
'StoveGroup_A'	0.0077733	1.1458	0.0067843	30	0.99463	-2.3322	2.3477
'familymembers'	-0.0077907	0.11451	-0.068033	30	0.94621	-0.24166	0.22608
'primarycook_1'	-1.5495	0.96145	-1.6116	30	0.11752	-3.513	0.41408
'AGECAT_1'	-0.53513	1.0553	-0.50707	30	0.61581	-2.6904	1.6202

Random effects covariance parameters (95% CIs):

Group: INDIVID (37 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	1.5235	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.5235	NaN	NaN

A2.8. EC, OC, and TC median mass apportionment from the different PMF solutions presented.

Table A2-1 EC, OC, and EC+OC mass apportionment

Ambient	Light n-alkanes	Odd n-alkanes	Light SVOCs	PAHs	Hopanes and n-alkanes	Methoxyphenols
OC (ng/m3)	224.0	88.8	18.4	1014.7	230.1	284.8
OC (%)	12.0	4.8	1.0	54.5	12.4	15.3
EC (ng/m3)	7.8	2.1	14.4	158.4	12.7	0.4
EC (%)	4.0	1.1	7.4	80.9	6.5	0.2
TC (ng/m3)	231.7	90.9	32.8	1173.1	242.8	285.2
TC (%)	11.3	4.4	1.6	57.0	11.8	13.9
Cooking Area	Light n-alkanes	Odd n-alkanes	Light SVOCs	PAHs	Hopanes and n-alkanes	Methoxyphenols
OC (ng/m3)	4830.0	9296.4	1788.2	1933.7	3659.4	2516.2
OC (%)	20.1	38.7	7.4	8.0	15.2	10.5
EC (ng/m3)	158.1	819.3	0.0	493.2	119.7	27.1
EC (%)	9.8	50.7	0.0	30.5	7.4	1.7
TC (ng/m3)	4988.1	10115.7	1788.2	2426.9	3779.1	2543.3

TC (%)	19.5	39.5	7.0	9.5	14.7	9.9
Personal	Light n-alkanes	Odd n-alkanes	Light SVOCs	PAHs	Hopanes and n-alkanes	Methoxyphenols
OC (ng/m3)	3552.2	8716.6	1798.5	553.8	5907.5	1325.3
OC (%)	16.3	39.9	8.2	2.5	27.0	6.1
EC (ng/m3)	116.3	768.2	0.0	141.3	193.2	14.3
EC (%)	9.4	62.3	0.0	11.5	15.7	1.2
TC (ng/m3)	3668.4	9484.8	1798.5	695.1	6100.7	1339.5
TC (%)	15.9	41.1	7.8	3.0	26.4	5.8
Personal and Cooking Area	Light n-alkanes	Odd n-alkanes	Light SVOCs	PAHs	Hopanes and n-alkanes	Methoxyphenols
OC (ng/m3)	4645.1	8716.6	1798.5	721.9	5452.6	1357.1
OC (%)	20.5	38.4	7.9	3.2	24.0	6.0
EC (ng/m3)	152.1	768.2	0.0	184.1	178.3	14.6
EC (%)	11.7	59.2	0.0	14.2	13.7	1.1
TC (ng/m3)	4797.2	9484.8	1798.5	906.0	5631.0	1371.7
TC (%)	20.0	39.5	7.5	3.8	23.5	5.7

APPENDIX 3

Exposures to carbon monoxide in a cookstove intervention in Northern Ghana

A3.1. CO exposure study summary statistics

Summary statistics are presented in Table 1 grouped by the same variable categories as used in the exposure model presented in Section 3.1.

Table A3-1 . Descriptive CO exposure statistics

	Mean (ppm)	Median (ppm)	Std dev (ppm)
Control group	0.98	0.52	1.37
Gyapa/Philips	1.09	0.54	2.12
Philips/Philips	0.94	0.37	1.60
Gyapa/Gyapa	1.10	0.50	1.64
Primary cook females >5y	1.17	0.59	1.93
Non-primary cook females >5y	0.83	0.45	1.09
Non-primary cook males >5y	0.91	0.42	1.52
Children <5y	0.84	0.34	1.40
Poorest	1.10	0.65	1.69
Poorer	1.09	0.52	1.53
Poor	1.10	0.49	2.31
Less poor	0.86	0.44	1.24
Least poor	0.94	0.34	1.52
Harmattan bush burning	0.88	0.44	1.41
Hot dry	0.71	0.26	1.04
Light Rainy	1.21	0.62	1.38
Heavy Rainy	1.49	0.64	2.59
Transition	1.06	0.43	1.60

Exceedances of WHO Tier-1 standards from this study were calculated using the calibrated minute-data from the Lascar USB-CO monitors, and required 75% data completion for each time scale to be included. The fraction of exposure exceeding WHO tier-1 standards was low compared to most previous cookstove studies.

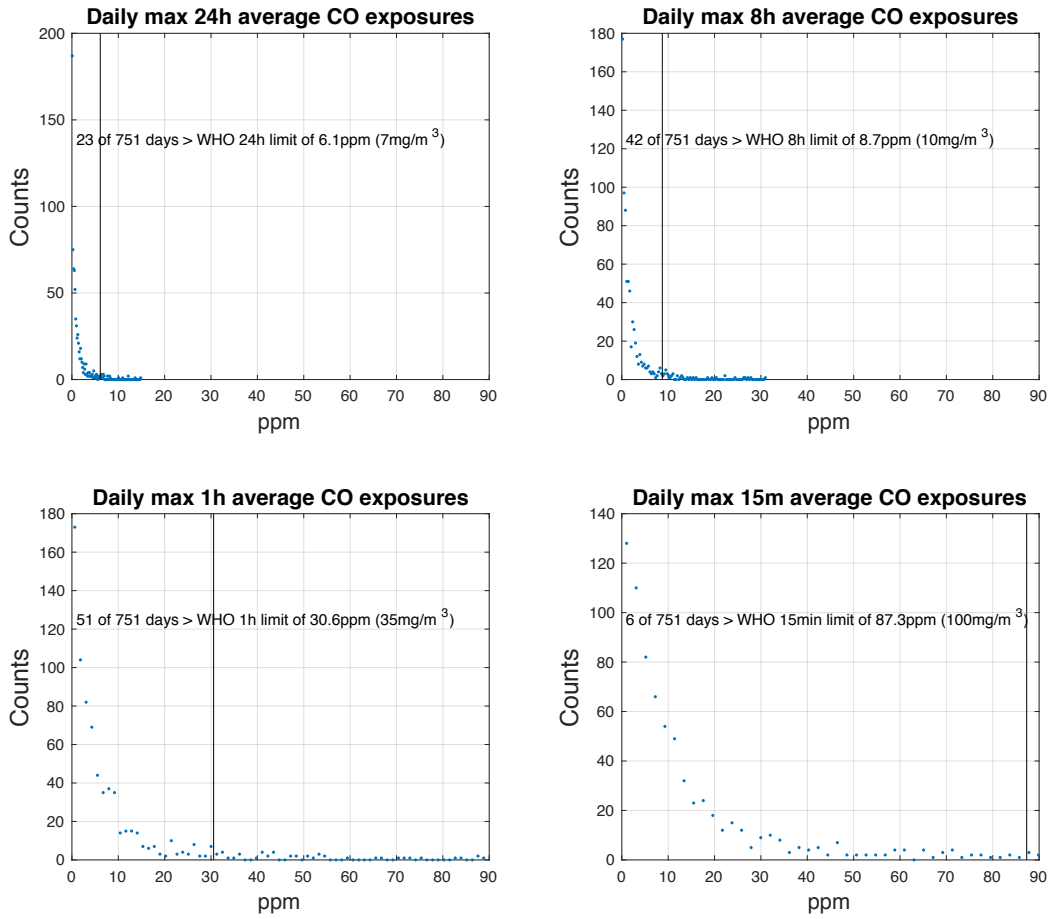


Figure A 3-1 Distributions of average CO exposure by time periods relevant to WHO tier-1 standards

A3.2. Lascar USB-CO calibration and quality assurance

Lascar monitors were calibrated with certified CO standards at the Hannigan Lab at CU Boulder. Typically, three or more calibrations points were used (Figure 2), but in some cases two-point span checks were employed. In the field, balanced sampling was performed from high and low concentration time points, representative of the concentrations the monitors

were exposed to in the field.

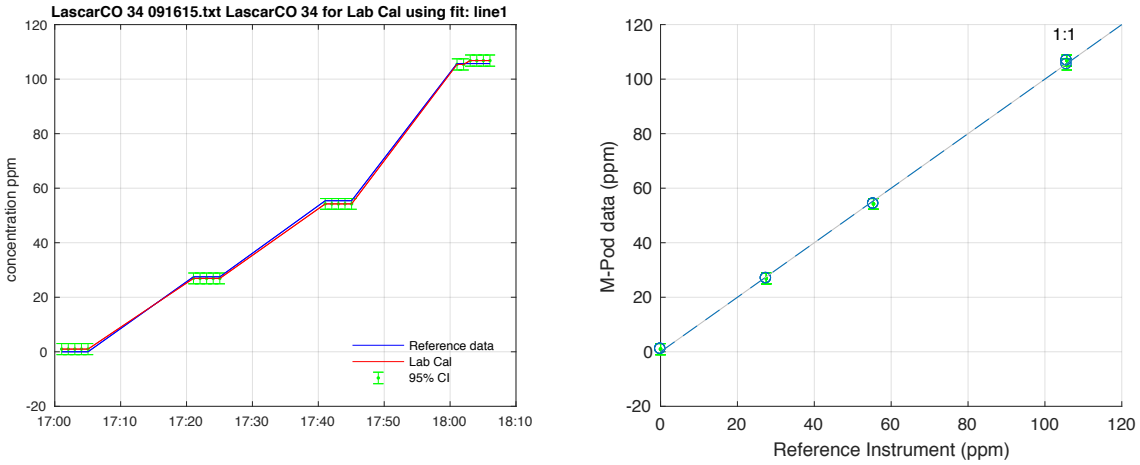


Figure A 3-2 Example calibration of a Lascar that returned from the field after deployment to Navrongo.

Data filtering for quality assurance was manually performed consistently and blind to the study group. There were various types of error observed with the monitors over time, and the data checker relied on consistency of issues, duplicate measures, and calibration quality to remove suspect data. A time series of calibration data deployments, both successful and flagged, is shown in **Error! Reference source not found.**.

Figure A 3-3 Lascar USB-CO calibration and data quality time line. Some Lascar monitors like #1 and #3 never operated correctly and were returned to the manufacturer. In most cases, the monitors were non-operational upon their return to the CU Boulder Hannigan Lab, so a post-calibration could not be performed.

A3.3. Lascar USB-CO duplicate analysis

The duplicate Lascar CO monitors were primarily deployed in the latter half of the study. In Figure 4 we present the comparison among duplicate measures for both the uncalibrated and calibrated data.

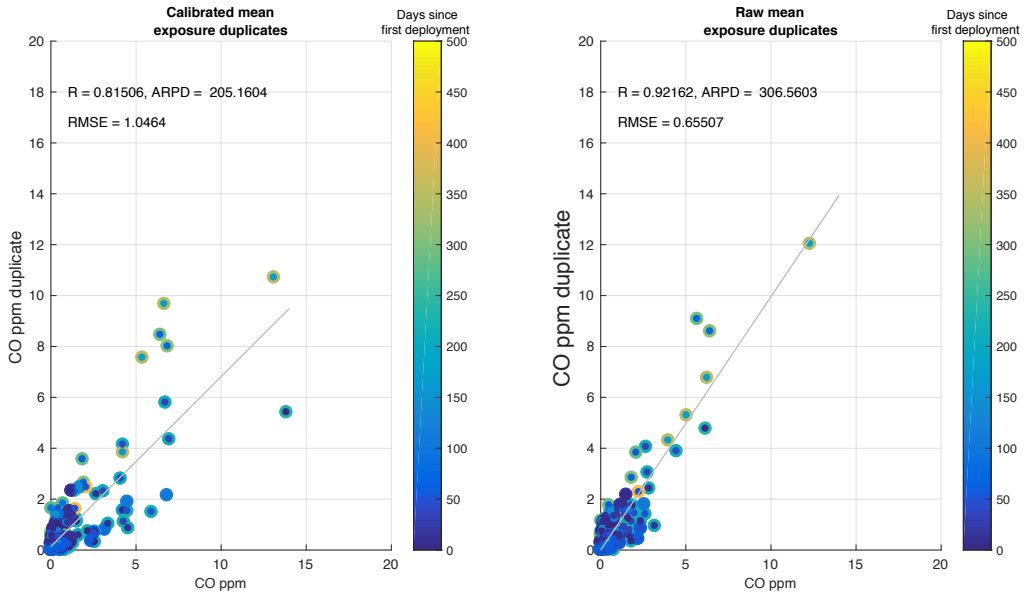


Figure A 3-4 Agreement of daily average Lascar USB-CO duplicates for both calibrated and raw values.

A3.4. Complete CO exposure mixed effects model results

The mixed effects model results presented in section 3.1 are presented in detail here with complete model output. The calibrated and uncalibrated model results are shown for comparison purposes.

Linear mixed-effects model fit by ML

Model information:

Number of observations	751
Fixed effects coefficients	15
Random effects coefficients	268
Covariance parameters	2

Formula:

LogCalibratedMeans ~ 1 + SES + season + StoveGroup + primarycookbygender_age5 + (1 | UserID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
2781.4	2860	-1373.7	2747.4

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	-0.43276	0.23368	-1.852	736	0.064429	-0.89152	0.025988
'SES_Poorer'	-0.13301	0.21612	-0.61543	736	0.53846	-0.5573	0.29128
'SES_Poor'	-0.25135	0.22349	-1.1247	736	0.2611	-0.69009	0.1874
'SES_Less_poor'	-0.45478	0.23658	-1.9223	736	0.054957	-0.91924	0.0096803
'SES_Least_poor'	-0.50334	0.23012	-2.1873	736	0.029034	-0.95511	-0.051572
'season_Heavy_Rainy'	0.32407	0.15867	2.0424	736	0.041472	0.012562	0.63557
'season_Light_Rainy'	0.38067	0.18368	2.0725	736	0.038566	0.020078	0.74127
'season_Transition'	0.030675	0.32699	0.093811	736	0.92528	-0.61127	0.67262
'season_Hot_dry'	-0.33017	0.16591	-1.9901	736	0.046948	-0.65588	-0.0044663
'StoveGroup_C'	0.011454	0.19579	0.058504	736	0.95336	-0.37292	0.39583
'StoveGroup_B'	-0.14274	0.19469	-0.73316	736	0.4637	-0.52495	0.23947
'StoveGroup_A'	-0.01935	0.20565	-0.094091	736	0.92506	-0.42308	0.38438
'primarycookbygender_age5_0F'	-0.43779	0.23105	-1.8948	736	0.058515	-0.89138	0.015809
'primarycookbygender_age5_0M'	-0.22312	0.23619	-0.94464	736	0.34515	-0.6868	0.24057
'primarycookbygender_age5_<5y'	-0.50652	0.16852	-3.0057	736	0.0027397	-0.83735	-0.17568

Random effects covariance parameters (95% CIs):

Group: UserID (268 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.68991	0.54502	0.87331

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.377	1.2937	1.4656

A3.5. Un-calibrated CO exposure mixed effects model results

Linear mixed-effects model fit by ML

Model information:

Number of observations	751
Fixed effects coefficients	15
Random effects coefficients	268
Covariance parameters	2

Formula:

LogCalibratedMeans ~ 1 + SES + season + StoveGroup + primarycookbygender_age5 + (1 | UserID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
2736.7	2815.3	-1351.4	2702.7

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	-0.46921	0.22609	-2.0753	736	0.038301	-0.91306	-0.025357
'SES_Poorer'	-0.087792	0.20908	-0.4199	736	0.67468	-0.49825	0.32267
'SES_Poor'	-0.17955	0.21619	-0.83053	736	0.40651	-0.60397	0.24487
'SES_Less_poor'	-0.39359	0.22888	-1.7196	736	0.08592	-0.84293	0.055748
'SES_Least_poor'	-0.46576	0.22258	-2.0925	736	0.036733	-0.90273	-0.028789
'season_Heavy_Rainy'	0.13046	0.15402	0.84703	736	0.39725	-0.17191	0.43283
'season_Light_Rainy'	0.093011	0.17827	0.52174	736	0.60201	-0.25697	0.44299
'season_Transition'	-0.0023797	0.31724	-0.0075012	736	0.99402	-0.62518	0.62042
'season_Hot_dry'	-0.46855	0.16096	-2.9111	736	0.0037108	-0.78454	-0.15257
'StoveGroup_C'	-0.028456	0.18937	-0.15026	736	0.8806	-0.40023	0.34332
'StoveGroup_B'	-0.16357	0.18829	-0.86875	736	0.38526	-0.53322	0.20607
'StoveGroup_A'	-0.076173	0.19892	-0.38294	736	0.70187	-0.46668	0.31434
'primarycookbygender_age5_0F'	-0.35722	0.22357	-1.5978	736	0.11052	-0.79613	0.081699
'primarycookbygender_age5_0M'	-0.15818	0.22855	-0.69207	736	0.48911	-0.60687	0.29052
'primarycookbygender_age5_<5y'	-0.47882	0.16308	-2.936	736	0.0034283	-0.79898	-0.15865

Random effects covariance parameters (95% CIs):

Group: UserID (268 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.66251	0.52274	0.83964

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.3389	1.2583	1.4247

A3.6. Time of day trends for cooking area CO and personal CO

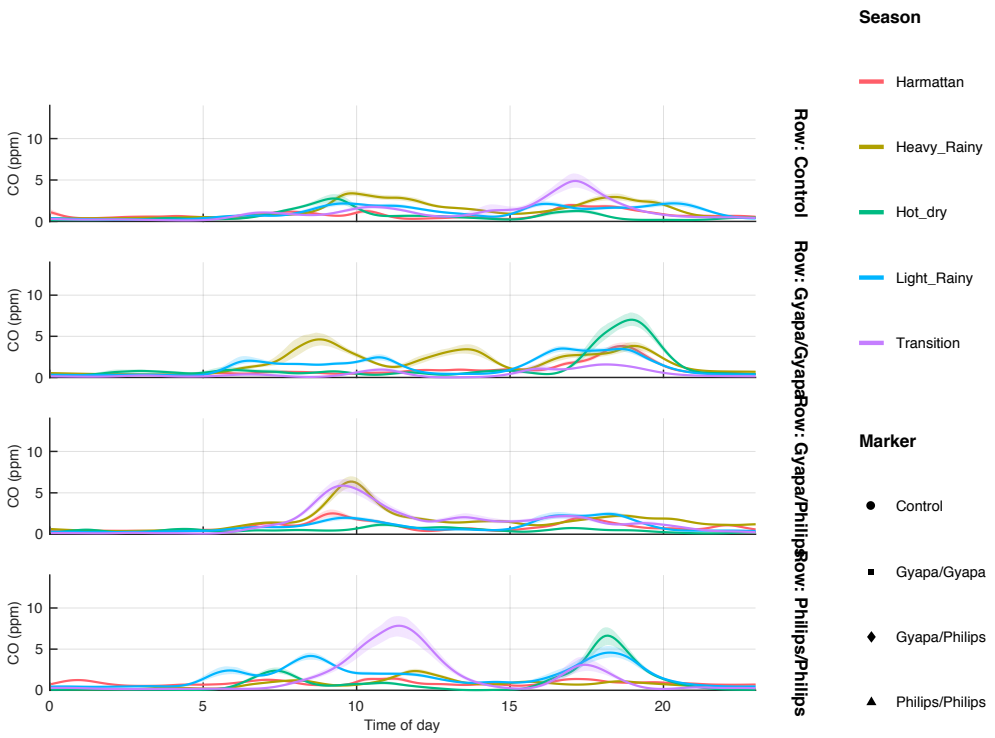


Figure A 3-5 Personal exposure by season and stove group, smoothed using b-splines

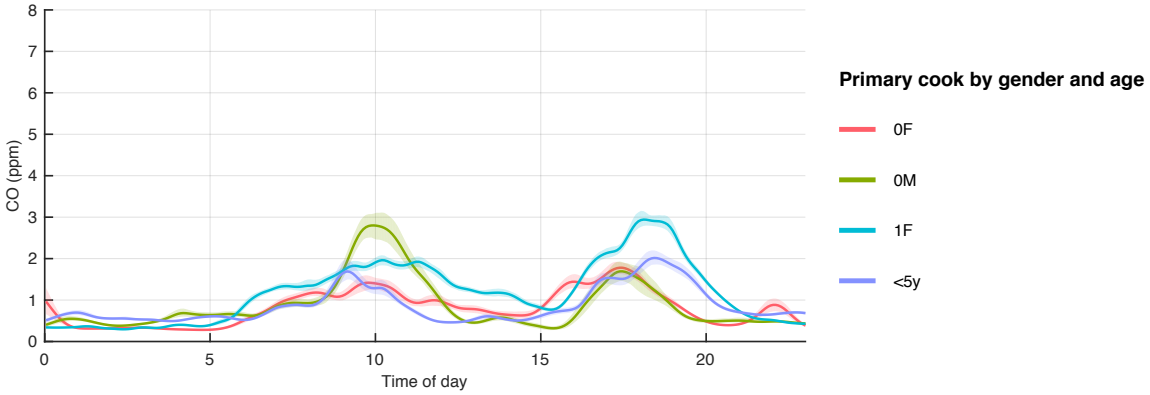


Figure A 3-6 B-spline smoothed personal CO exposure grouped by primary cook status gender group. '0' values are for non-primary cooks, and '1' is for the females listed as primary cooks. Children of both genders and under age five were grouped together for the '<5y' category. No males were listed as primary cooks.

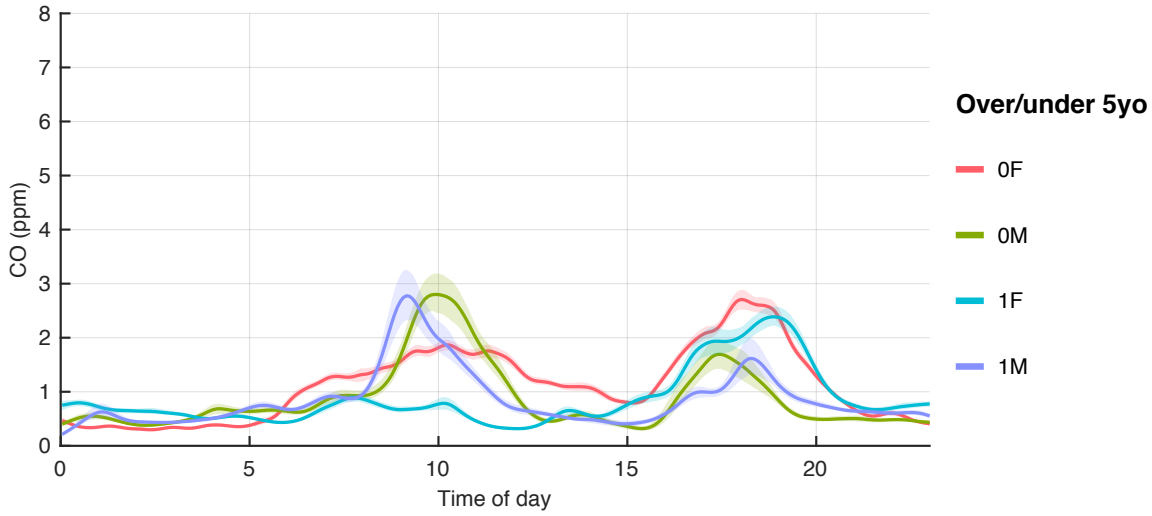
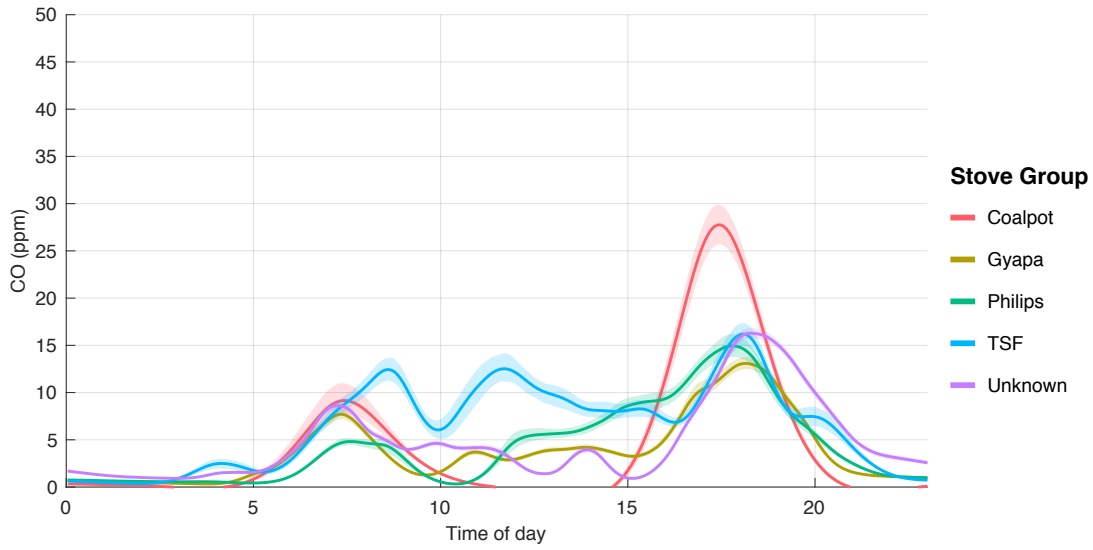


Figure A 3-7 B-spline smoothed personal CO exposure grouped by age and gender group. '0' values are under 5, and '1's are over 5 years of age.



A3.7. Modeling CO with carbonaceous $PM_{2.5}$

model_EC_simple =

Linear regression model:

LogECugm3 ~ 1 + LogCO

Estimated Coefficients:

	Estimate	SE	tStat	pValue
(Intercept)	0.13616	0.17358	0.78442	0.43454
LogCO	0.12822	0.11449	1.1199	0.26527

Number of observations: 108, Error degrees of freedom: 106

Root Mean Squared Error: 1.55

R-squared: 0.0117, Adjusted R-Squared 0.00237

F-statistic vs. constant model: 1.25, p-value = 0.265

model_OC_simple =

Linear regression model:

LogOCugm3 ~ 1 + LogCO

Estimated Coefficients:

	Estimate	SE	tStat	pValue
(Intercept)	3.3357	0.1399	23.844	2.132e-44
LogCO	0.21109	0.092272	2.2877	0.024141

Number of observations: 108, Error degrees of freedom: 106

Root Mean Squared Error: 1.25

R-squared: 0.047, Adjusted R-Squared 0.0381

F-statistic vs. constant model: 5.23, p-value = 0.0241

APPENDIX 4

Bluetooth Beacon proximity sensing to improve personal exposure assessment

A4.1. G-Pod cooking area micro-environment monitoring

The G-Pod air quality monitor sampling inlets were placed 1-meter away from the cookstove of interest, at 1-meter height, with BLE Beacons bolted to the outside of the cases. CO was measured with Alphasense CO-B4 electrochemical sensors. CO_2 was measured with NDIR sensors (S200, ELT Corp.). Temperature, humidity, and barometric pressure were also measured in the G-Pod, and on a subset of samples, total VOCs were measured with PID sensors (pID-Tech Plus Silver, Baseline-Mocon Inc.). Integrated $PM_{2.5}$ was collected and analyzed as described in Piedrahita et al. (2017). From December 2013 through November 2014, only the most-used cooking area was monitored, but from November 2014 – January 2016, the two most-used cooking areas were monitored.

A4.2. Cooking area microenvironment measurements calibration and data processing

We employed a multi-step protocol to ensure data quality over the duration of the study. CO and CO_2 sensors underwent lab calibrations at the University of Colorado Hannigan Lab before and after each sampling period (November 2013, October 2014, May 2015, October 2015, and February 2016). An exponential calibration model controlling for temperature was used for the Alphasense CO sensor (Masson et al., 2015), while a first order linear model was used for the ELT CO_2 sensor (Piedrahita et al., 2013). Span checks were performed at the NHRC in March 2015 after receiving cylinders of span gases. Calibrations were very consistent over time for these sensors, as has been previously shown (Masson et al., 2015).

The G-Pods were configured to sample at 15-second intervals, and 1-minute medians were used for further analysis. CO and CO_2 data were baseline-adjusted to the 5^{th} percentile of the ambient background concentration, to mitigate baseline sensor drift over time. We found evidence of uniform background levels of these pollutants (data not yet published), and since ventilation rates are very high in the measured microenvironments due to building styles, we considered this to be a reasonable approach since we could not perform full calibrations as often as desired.

A4.3. Beacon distance calibration

The iBeacon protocol includes a calibration constant to normalize the RSSI-to-distance conversion but Android devices do not use this method, which made it necessary to perform a calibration with our specific hardware (**Error! Reference source not found.**). Calibration was performed on an open sports field free of extraneous objects using an equation of the form distance $=10^{(p1 * RSSI)/p2)}$, as has been used commonly in beacon work (Anagnostopoulos and Deriaz, 2014). Stationary data was collected with two phones and two beacons at distances of 2, 5, 10, 20, and 40m, for durations of three minutes at each distance.

The data from each phone/beacon combination was fit individually to but in the end a single calibration using all the data was kept for further analysis because we determined that the inter-phone and inter-beacon differences were due to random experimental error, like orientation, rather than systemic differences in hardware. Additionally, not all phones were available at the time of calibration, so a bulk approach was deemed prudent. An r-squared value of 0.72 was obtained using all the data, with evenly distributed residuals (**Error! Reference source not found.**).

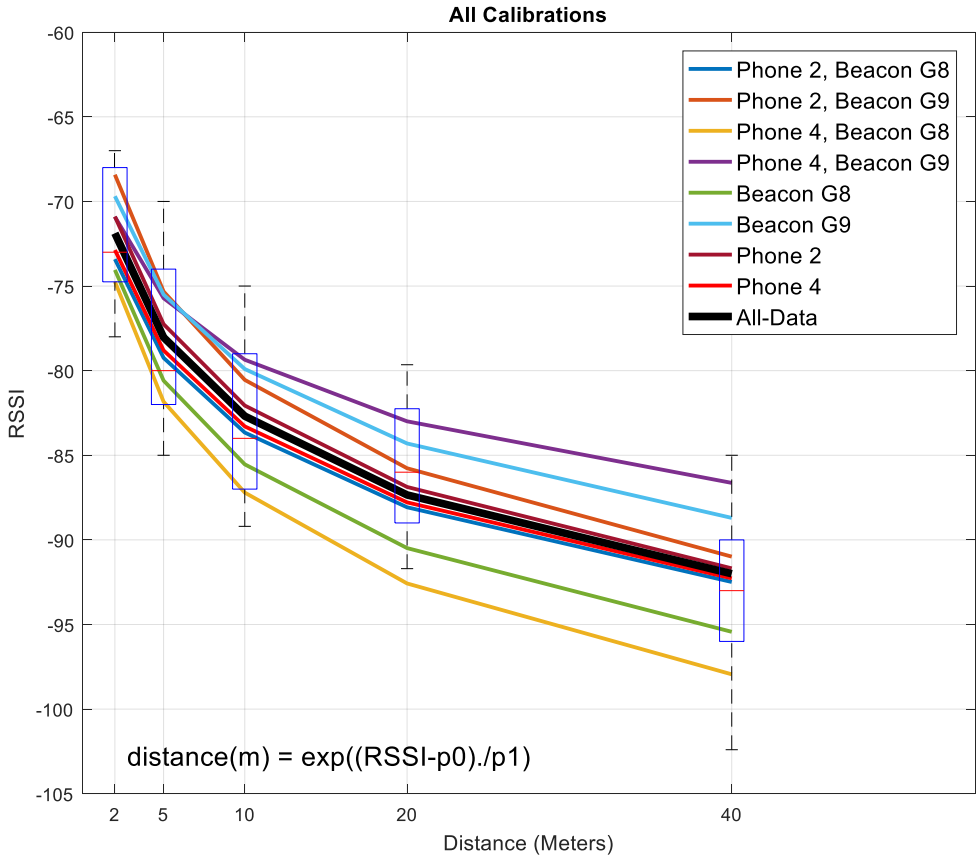


Figure A 4-1. RSSI-to-distance calibrations for various calibration models. The bold black line shows a fit using aggregate data from both phones, and both beacons, while the thin lines are phone/beacon specific. Box and whisker plots show the distributions of the all the raw data, with whiskers representing 5th and 95th percentiles. Note that the outlying curves on the top and bottom of the plot are from phone 4, suggesting a performance issue with that phone.

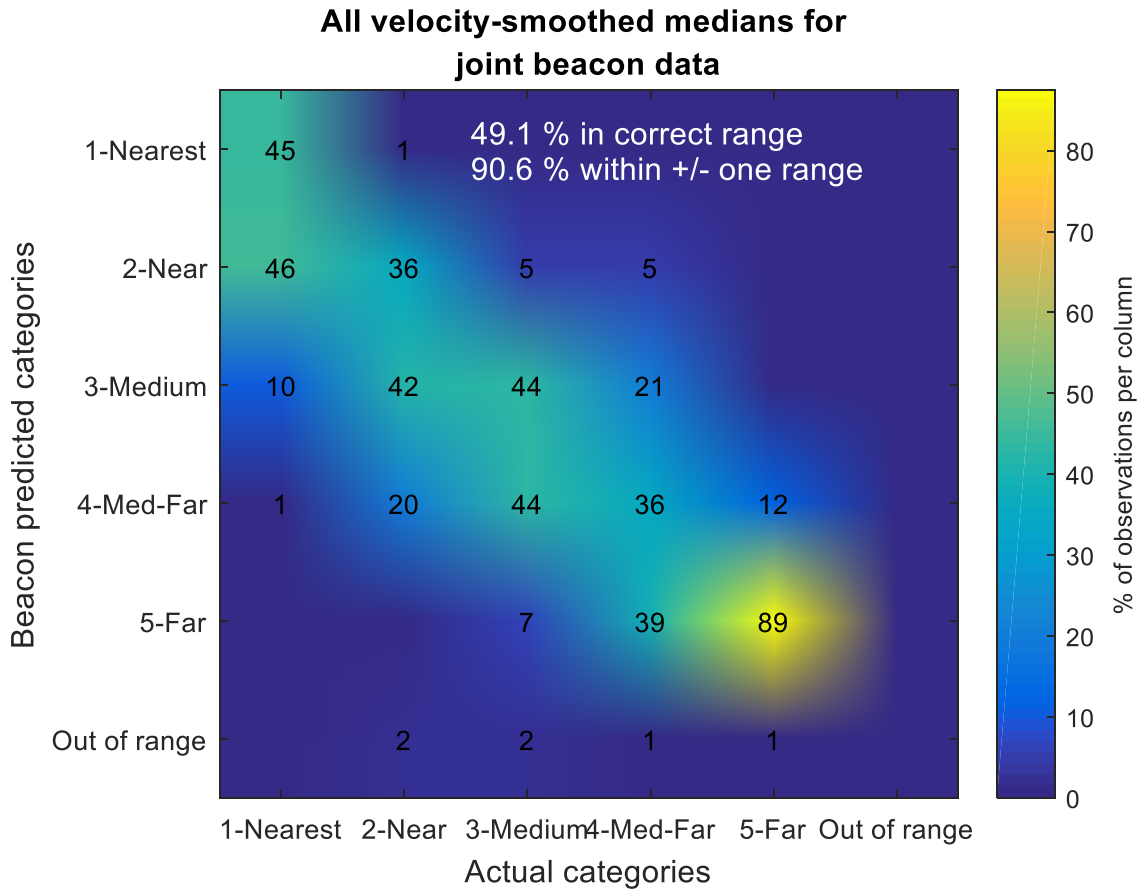


Figure 4-1. Modeled categories vs. known categories for all merged beacon signal data. Percentages add up to 100 by column, as the x-axis represent the known category values

A4.4. Participant protocol compliance

Compliance was calculated using the rolling standard deviation of one-hour segments of the minute beacon data and flagging hours in which the standard deviation of any available beacon signal in units of RSSI was greater than 2, excluding nighttime between 21:00 and 7:00. Standard deviation of 2 was selected based on noise variability during calibration periods and is in units of RSSI because the residuals are normally and evenly distributed throughout the distance categories, but residuals are not evenly distributed after passing through the exponential calibration curve in conversion to distance (m). Using this approach, average compliance was measured at 81.9%.

A4.5. Beacon system validation

A validation test was performed as part of an outdoors cookstove test. The same distance ranges were prepared, and a user walked throughout each range for 20 minutes. Three phones were placed at the epicenter of the region arcs, along with the stove. There were more obstructions in this test than the open field, making it a more realistic scenario. There are still limitations with this approach, and it is not meant to represent all indoor use, which could be highly variable due to placement of equipment, home layouts, building materials, and user behaviors. Dedicated indoor testing in a variety of environments would provide a better understanding of the expected performance.

Validation testing for both deployments and all combinations of phones and beacons showed correct classification of distance categories on 30.9% of observations on average, and 67.4% of observations were within one distance zone of the correct zone. System performance was not significantly different between the initial validation test and the outdoor cooking test. However, classification errors were not evenly distributed among distance categories, with lower matching success rates for the more distant ranges (Figure A4-3). This was expected, since the relative signal drop-off due to bodily interference is higher for closer ranges. Calibration showed inter-phone variability of 4.4m (RMS error) (**Error! Reference source not found.**), suggesting that each of our phones would have benefitted from individual calibrations, though such variability is model specific for the phone and may not be the case with other phone models.

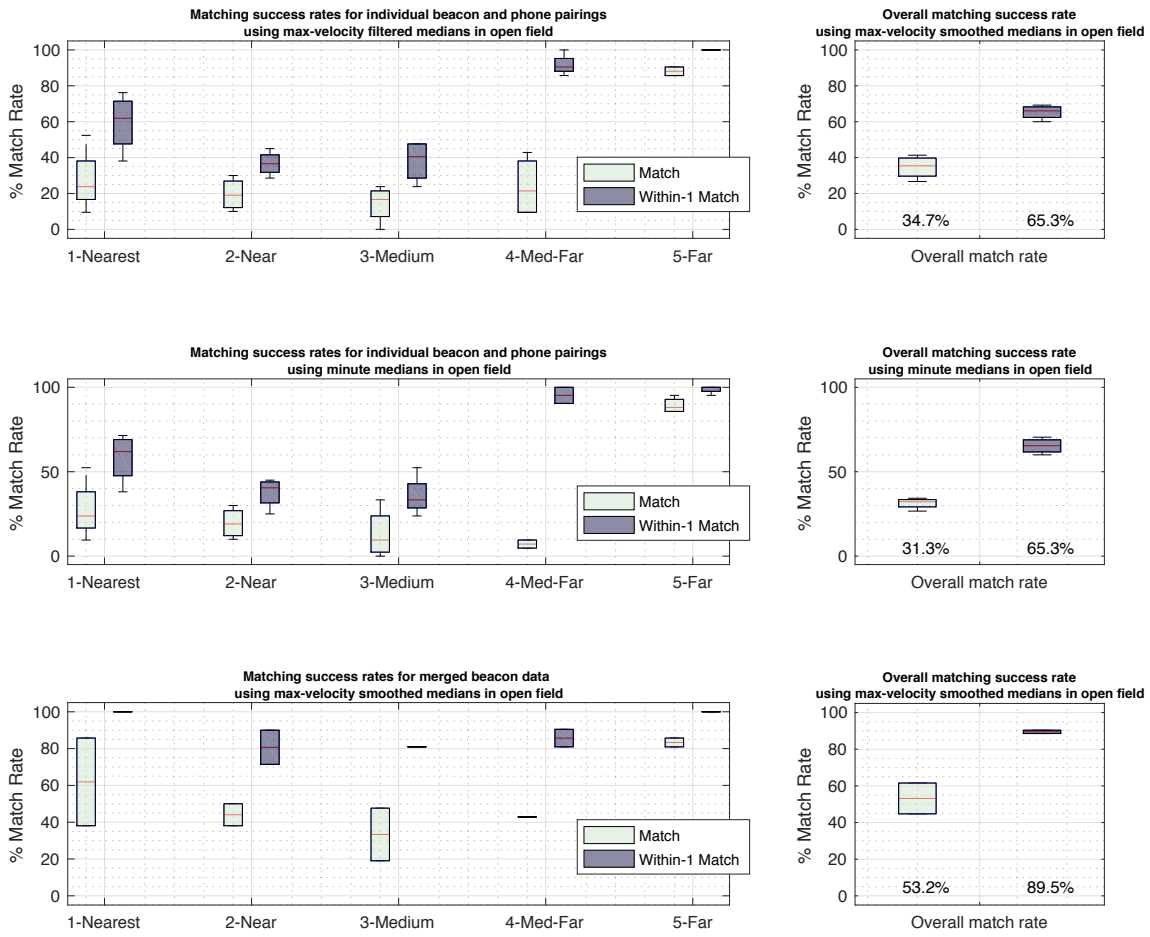


Figure A4-2. Performance from the validation deployment in an open field. Light colored boxes show the match rate, and dark boxes show the rate at which the algorithm predicted within one zone of the correct zone. Left frames show performance by distance zone, while right frames show overall performance. Top frames show match rates using the MV algorithm, the middle frames show rates using minute medians, and the bottom frames show match rates using the merged beacon data along with the MV algorithm.

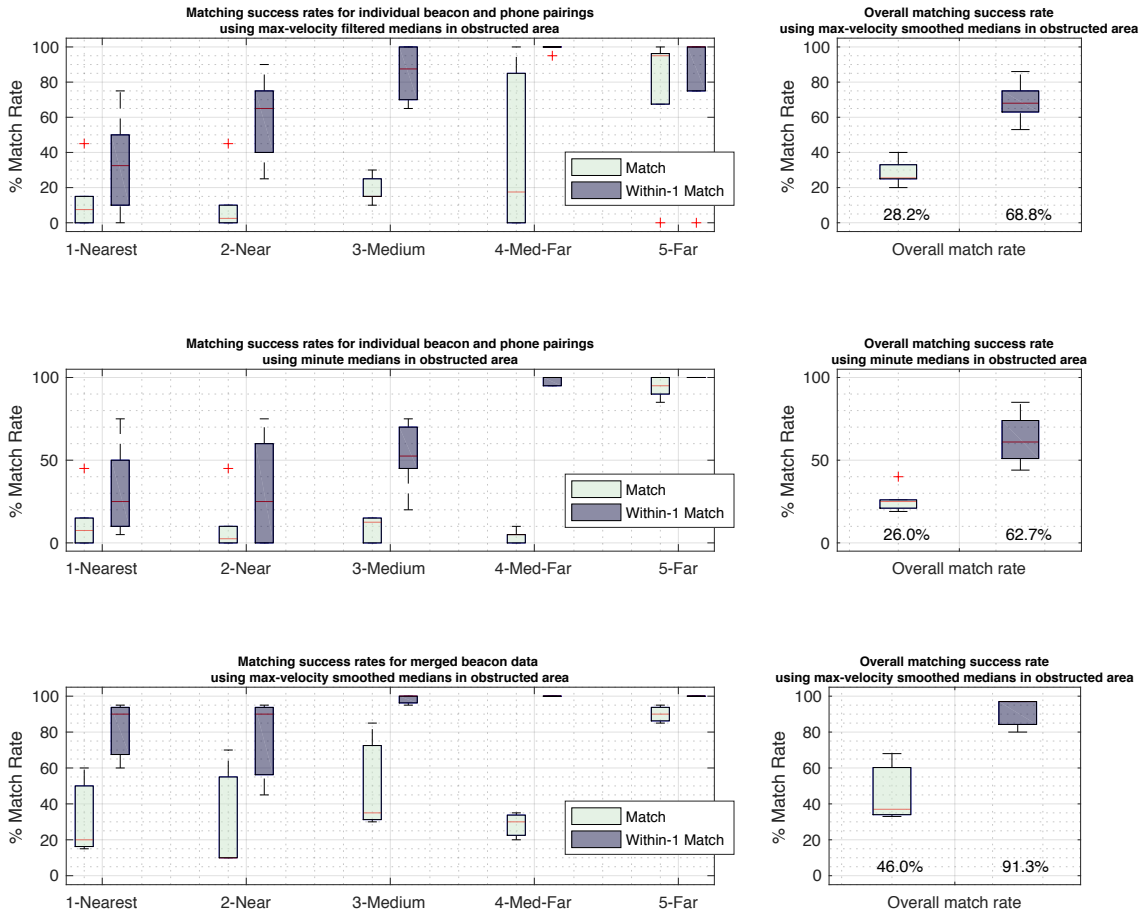


Figure A4-3. Performance from the test deployment with additional obstructions. Light colored boxes show the match rate, and dark boxes show the rate at which the algorithm predicted within one zone of the correct zone. Left frames show performance by distance zone, while right frames show overall performance. Top frames show match rates using the MV algorithm, the middle frames show rates using minute medians, and the bottom frames show match rates using the merged beacon data along with the MV algorithm.

The MV filter provided a 2.2% and 3.4% improvement over the simple medians for the direct match rate of the 'open field' and 'obstructed' data sets, and 0% and 6.1% improvement for the within-one match rate of the 'open field' and 'obstructed' data sets (Figure A4-2, Figure A4-3). The physical reasoning behind this approach suggests that it would improve performance in more variable and dynamic environments, with only minor potential drawbacks in outlying use cases.

Merging the data from both beacons worn by the user resulted in substantially better performance, since the attenuation effects were much less pronounced due to the improved direct line-of-sight to the phones at nearly all times. 53.1% of observations were correctly

classified on average, while 89.5% of observations were classified within one zone for the 'open field' test, while in the obstructed data set the values were 46.0% and 91.3%. The merged data had errors that were more evenly distributed among categories.

A4.6. CO and beacon modeling results

Table A 4-1 Summary of results from modeling personal CO exposure by cooking area CO.

	Personal vs. cooking area CO by zones (Eq. 5-3)				Daily average personal vs. cooking area CO (Eq. 5-4)			
	Expected value ppm (95% CI)	**Coefficient (95% CI)**	**% change (95% CI)**	**P-value**	**Expected value ppm (95% CI)**	**Coefficient (95% CI)**	**% change (95% CI)**	**P-value**
Intercept	.1 (.07, .16)	-2.27 (-2.69, -1.85)	NA	0.00	.14 (.07, .25)	-2. (-2.62, -1.38)	NA	0.00
log(Weighted cooking area CO)	2.74 (2.25, 3.33)	1.01 (.81, 1.2)	173.54 (124.75, 232.92)	0.00	2.24 (1.5, 3.36)	.81 (.4, 1.21)	124.25 (49.87, 235.56)	0.00
Random effect by individual		.35 (.13, .94)				0.00		
Random error covariance		1. (.76, 1.31)				1.28 (.82, 2.01)		
Adjusted R-squared		0.63				0.28		
N		123				38		

A4.7. Daily average modeling by stove group using only the data available with Beacons

Model information:

Number of observations	71
Fixed effects coefficients	4
Random effects coefficients	31
Covariance parameters	2

Formula:

LogPersonalCOMeans ~ 1 + StoveGroup + (1 | UserID)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
274.86	288.44	-131.43	262.86

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	-0.38888	0.689	-0.56442	67	0.57435	-1.7641	0.98636

'StoveGroup_C'	-1.1923	0.84385	-1.4129	67	0.16232	-2.8766	0.49207
'StoveGroup_B'	-1.0066	0.73792	-1.3642	67	0.17708	-2.4795	0.46626
'StoveGroup_A'	-1.2481	0.76329	-1.6351	67	0.10671	-2.7716	0.27544

Random effects covariance parameters (95% CIs):

Group: UserID (31 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	6.4937e-07	NaN	NaN

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	1.5406	1.307	1.8161

APPENDIX 5

Laboratory assessment of electrochemical carbon monoxide monitors

Table A5.1 Sensitivity and relative error modeled with cumulative exposure. *Significant at p = 0.05.

	Equation 1 for slope change over time	Equation 2 for relative error (%) using best calibration	Equation 2 for relative error (%) using pre/post calibration	Equation 2 for relative error (%) using raw data
	Coefficients (CI)	Coefficients (CI)	Coefficients (CI)	Coefficients (CI)
Intercept ('fresh' category)	8.29E-1 (8.68E-1, 7.89E-1)*	5.83E+0 (1.15E+1, 1.77E-1) *	1.07E+1 (1.98E+1, 1.69E+0) *	-7.03E+0 (6.07E+0, -2.01E+1)
Cumulative exposure ppm-hr	-5.75E-6 (-2.65E-6, -8.84E-6) *	-5.84E-4 (1.09E-4, -1.28E-3)	-5.72E-4 (4.60E-4, -1.60E-3)	-1.59E-3 (-2.35E-4, -2.95E-3) *
Batch ('used' relative to 'fresh')	6.78E-2 (1.20E-1, 1.51E-2) *	-7.56E+0 (-1.02E+0, -1.41E+1)	-1.15E+1 (-5.65E-1, -2.23E+1)	-5.35E+0 (1.13E+1, -2.20E+1)
Cumulative exposure:Batch	4.74E-6 (8.00E-6, 1.48E-6) *	5.35E-4 (1.23E-3, -1.63E-4)	4.76E-4 (1.53E-3, -5.82E-4)	1.31E-3 (2.79E-3, -1.59E-4)
Random intercept variance	3.68E-2 (2.36E-2, 5.74E-2) *	2.13E+0 (4.05E-1, 1.12E+1) *	5.21E+0 (1.13E+0, 2.39E+1) *	1.02E+1 (5.36E+0, 1.94E+1) *
Random slope variance	1.13E-6 (5.14E-7, 2.47E-6) *	7.31E-5 (1.80E-5, 2.97E-4) *	2.47E-4 (4.93E-5, 1.24E-3) *	6.41E-4 (2.66E-4, 1.54E-3) *
Error variance	3.40E-2 (2.95E-2, 3.91E-2) *	8.10E+0 (7.08E+0, 9.26E+0) *	1.18E+1 (9.99E+0, 1.38E+1) *	1.43E+1 (1.23E+1, 1.65E+1) *

A5.1. Calibration slope error over time

Model information:

Number of observations	119
Fixed effects coefficients	4
Random effects coefficients	20
Covariance parameters	4

Formula:

betaslope ~ 1 + cumulativerawdoseppmhr*BatchCat + (1 + cumulativerawdoseppmhr | Lascar)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
-411.88	-389.65	213.94	-427.88

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	0.82871	0.020056	41.32	115	7.7289e-71	0.78898	0.86843
'cumulativerawdoseppmhr'	-5.7452e-06	1.5637e-06	-3.674	115	0.00036405	-8.8427e-06	-2.6477e-06
'BatchCat_Used'	0.067784	0.026586	2.5496	115	0.0121	0.015122	0.12045

'cumulativerawdoseppmhr:BatchCat_Used' 4.7403e-06 1.6469e-06 2.8784 115 0.0047667 1.4782e-06 8.0025e-06

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.036805	0.023613	0.057364
'cumulativerawdoseppmhr'	'(Intercept)'	'corr'	0.81064	0.12702	0.97215
'cumulativerawdoseppmhr'	'cumulativerawdoseppmhr'	'std'	1.1262e-06	5.1381e-07	2.4685e-06

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.034002	0.029543	0.039132

A5.2. Best calibration approach average error over time

Linear mixed-effects model fit by ML

Model information:

Number of observations	119
Fixed effects coefficients	4
Random effects coefficients	20
Covariance parameters	4

Formula:

BestPercentError ~ 1 + cumulativerawdoseppmhr*BatchCat + (1 + cumulativerawdoseppmhr | Lascar)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
865.36	887.59	-424.68	849.36

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	5.8301	2.8541	2.0427	115	0.043364	0.17677	11.483
'cumulativerawdoseppmhr'	-0.00058433	0.00035017	-1.6687	115	0.097893	-0.0012779	0.00010928
'BatchCat_Used'	-7.5622	3.3048	-2.2882	115	0.023952	-14.108	-1.0159
'cumulativerawdoseppmhr:BatchCat_Used'	0.00053541	0.00035262	1.5184	115	0.13166	-0.00016306	0.0012339

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	2.1325	0.40538	11.218
'cumulativerawdoseppmhr'	'(Intercept)'	'corr'	0.99866	-1	1
'cumulativerawdoseppmhr'	'cumulativerawdoseppmhr'	'std'	7.3124e-05	1.7978e-05	0.00029742

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	8.0969	7.0827	9.2563

A5.3. Pre/post calibration average error over time

Linear mixed-effects model fit by ML

Model information:

Number of observations	119
Fixed effects coefficients	4
Random effects coefficients	20
Covariance parameters	4

Formula:

PrePostPercentError ~ 1 + cumulativerawdoseppmhr*BatchCat + (1 + cumulativerawdoseppmhr | Lascar)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
963.83	986.07	-473.92	947.83

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	10.733	4.5658	2.3508	115	0.020435	1.6894	19.777
'cumulativerawdoseppmhr'	-0.00057236	0.00052107	-1.0984	115	0.27432	-0.0016045	0.00045979
'BatchCat_Used'	-11.456	5.4985	-2.0835	115	0.039424	-22.347	-0.56452
'cumulativerawdoseppmhr:BatchCat_Used'	0.00047613	0.00053402	0.8916	115	0.37447	-0.00058165	0.0015339

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1	Name2	Type	Estimate	Lower	Upper

'(Intercept)' '(Intercept)' 'std' 5.207 1.1322 23.947

'cumulativerawdoseppmhr' '(Intercept)' 'corr' 1 NaN NaN

'cumulativerawdoseppmhr' 'cumulativerawdoseppmhr' 'std' 0.00024709 4.9266e-05 0.0012393

Group: Error

Name Estimate Lower Upper

'Res Std' 11.755 9.9859 13.836

A5.4. Raw data average error over time

Linear mixed-effects model fit by ML

Model information:

Number of observations 119

Fixed effects coefficients 4

Random effects coefficients 20

Covariance parameters 4

Formula:

RawPercentError ~ 1 + cumulativerawdoseppmhr*BatchCat + (1 + cumulativerawdoseppmhr | Lascar)

Model fit statistics:

AIC BIC LogLikelihood Deviance

1021.5 1043.8 -502.76 1005.5

Fixed effects coefficients (95% CIs):

Name Estimate SE tStat DF pValue Lower Upper

'(Intercept)' -7.0258 6.6135 -1.0623 115 0.29031 -20.126 6.0743

'cumulativerawdoseppmhr' -0.0015921 0.00068517 -2.3237 115 0.0219 -0.0029493 -0.00023492

'BatchCat_Used' -5.3475 8.3906 -0.63732 115 0.52518 -21.968 11.273

'cumulativerawdoseppmhr:BatchCat_Used' 0.0013148 0.00074417 1.7668 115 0.079923 -0.00015929
0.0027888

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1 Name2 Type Estimate Lower Upper

'(Intercept)' '(Intercept)' 'std' 10.191 5.3569 19.388

'cumulativerawdoseppmhr' '(Intercept)' 'corr' 0.96774 -0.98766 1

'cumulativerawdoseppmhr' 'cumulativerawdoseppmhr' 'std' 0.00064071 0.00026619 0.0015422

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	14.278	12.341	16.519

Table A5.2 Mixed effects modeling results for relationships between dynamic sensor characteristics and cumulative sensor exposures. *Significant at p = 0.05.

	Equation 6-3 for log(rise time (s))	Equation 6-3 for log(decay time (s))	Equation 6-4 for log(maximum slope)
	Coefficients (CI)	Coefficients (CI)	Coefficients (CI)
Intercept	6.00E+0 (5.45E+0, 6.56E+0)*	4.27E+0 (3.72E+0, 4.82E+0) *	-3.17E+0 (-3.77E+0, -2.56E+0) *
Batch	-6.40E-1 (-1.31E+0, 3.30E-2)	-1.08E+0 (-1.80E+0, -3.58E-1) *	3.08E-1 (-4.24E-1, 1.04E+0)
log(peak concentration (ppm))	-1.48E-1 (-2.51E-1, -4.40E-2)*	1.05E-1 (-2.38E-3, 2.12E-1)	7.16E-1 (6.04E-1, 8.28E-1) *
Cumulative exposure (ppm-hr)	6.44E-6 (1.31E-7, 1.27E-5) *	3.78E-6 (-4.45E-7, 8.00E-6)	-5.48E-6 (-1.20E-5, 9.94E-7)
Batch_Used:log(peak concentration)	1.24E-1 (-2.57E-3, 2.50E-1)	1.83E-1 (4.13E-2, 3.24E-1) *	3.46E-3 (-1.32E-1, 1.39E-1)
log(peak concentration): cumulative exposure (ppm-hr)	-1.66E-6 (-2.83E-6, -4.88E-7*)	-1.48E-6 (-2.28E-6, -6.68E-7*)	1.12E-6 (-8.36E-8, 2.33E-6)
Random intercept variance	5.27E-4 (4.47E-4, 6.21E-4) *	1.81E-1 (4.54E-2, 7.21E-1) *	8.92E-3 (1.89E-3, 4.21E-2) *
Random slope variance	9.37E-5 (7.77E-6, 1.13E-3) *	7.47E-3 (1.91E-3, 2.92E-2) *	6.24E-5 (5.30E-5, 7.34E-5) *
Error variance	1.91E-1 (1.62E-1, 2.25E-1) *	1.41E-1 (1.21E-1, 1.64E-1) *	2.25E-1 (1.92E-1, 2.65E-1) *

A5.5. Maximum slope model

Linear mixed-effects model fit by ML

Model information:

Number of observations	299
Fixed effects coefficients	6
Random effects coefficients	20
Covariance parameters	4

Formula:

logMaxSlope ~ 1 + BatchCat*logPeak_ppm + logPeak_ppm*cumulativedose60 + (1 + logPeak_ppm | Lascar)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
425.92	462.92	-202.96	405.92

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	-3.1658	0.30806	-10.277	293	2.3366e-21	-3.772	-2.5595

'BatchCat_Used'	0.3077	0.372	0.82716	293	0.40882	-0.42443	1.0398
'logPeak_ppm'	0.7159	0.056885	12.585	293	2.501e-29	0.60394	0.82785
'cumulativedose60'	-5.481e-06	3.2898e-06	-1.6661	293	0.096767	-1.1956e-05	9.9358e-07
'BatchCat_Used:logPeak_ppm'	0.0034585	0.068699	0.050343	293	0.95988	-0.13175	0.13867
'logPeak_ppm:cumulativedose60'	1.1248e-06	6.1397e-07	1.832	293	0.067972	-8.3581e-08	2.3331e-06

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.09447	0.043477	0.20527
'logPeak_ppm'	'(Intercept)'	'corr'	-1	NaN	NaN
'logPeak_ppm'	'logPeak_ppm'	'std'	0.007899	0.0072833	0.0085667

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.47461	0.43769	0.51464

A5.6. Rise time model

Linear mixed-effects model fit by ML

Model information:

Number of observations	292
Fixed effects coefficients	6
Random effects coefficients	20
Covariance parameters	4

Formula:

logT90rise ~ 1 + BatchCat*logPeak_ppm + logPeak_ppm*cumulativedose60 + (1 + logPeak_ppm | Lascar)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
370.92	407.68	-175.46	350.92

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	6.0037	0.28163	21.318	286	5.0183e-61	5.4494	6.558
'BatchCat_Used'	-0.63975	0.34178	-1.8718	286	0.062253	-1.3125	0.032975
'logPeak_ppm'	-0.14766	0.052665	-2.8037	286	0.0053978	-0.25132	-0.043996

'cumulativedose60'	6.4396e-06	3.2053e-06	2.0091	286	0.045469	1.307e-07	1.2748e-05
'BatchCat_Used:logPeak_ppm'	0.12358	0.064089	1.9283	286	0.054813	-0.002566	0.24972
'logPeak_ppm:cumulativedose60'	-1.6572e-06	5.9416e-07	-2.7892	286	0.0056382	-2.8267e-06	-4.8776e-07

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.022953	0.02115	0.024911
'logPeak_ppm'	'(Intercept)'	'corr'	1	NaN	NaN
'logPeak_ppm'	'logPeak_ppm'	'std'	0.0096794	0.0027867	0.03362

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.43689	0.40247	0.47427

A5.7. Decay time model

Linear mixed-effects model fit by ML

Model information:

Number of observations	355
Fixed effects coefficients	6
Random effects coefficients	20
Covariance parameters	4

Formula:

logT90decay ~ 1 + BatchCat*logPeak_ppm + logPeak_ppm*cumulativedose60 + (1 + logPeak_ppm | Lascar)

Model fit statistics:

AIC	BIC	LogLikelihood	Deviance
352.47	391.19	-166.24	332.47

Fixed effects coefficients (95% CIs):

Name	Estimate	SE	tStat	DF	pValue	Lower	Upper
'(Intercept)'	4.2694	0.28187	15.147	349	3.4692e-40	3.715	4.8237
'BatchCat_Used'	-1.0782	0.36637	-2.9431	349	0.0034674	-1.7988	-0.35767
'logPeak_ppm'	0.10494	0.054563	1.9232	349	0.055264	-0.0023764	0.21225
'cumulativedose60'	3.7762e-06	2.1463e-06	1.7594	349	0.079389	-4.4515e-07	7.9975e-06
'BatchCat_Used:logPeak_ppm'	0.18268	0.071901	2.5408	349	0.011493	0.041271	0.3241

'logPeak_ppm:cumulativedose60'	-1.4764e-06	4.109e-07	-3.5931	349	0.00037367	-2.2846e-06	-6.6827e-07

Random effects covariance parameters (95% CIs):

Group: Lascar (10 Levels)

Name1	Name2	Type	Estimate	Lower	Upper
'(Intercept)'	'(Intercept)'	'std'	0.42539	0.21313	0.84904
'logPeak_ppm'	'(Intercept)'	'corr'	-0.98406	-0.99828	-0.86018
'logPeak_ppm'	'logPeak_ppm'	'std'	0.086449	0.043718	0.17095

Group: Error

Name	Estimate	Lower	Upper
'Res Std'	0.37518	0.34788	0.40462