DISSERTATION

COOKSTOVE STARTUP MATERIAL CHARACTERIZATION AND QUANTIFICATION AND ACUTE CARDIOPULMONARY EFFECTS FROM CONTROLLED EXPOSURE TO COOKSTOVE AIR POLLUTION

Submitted by

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ABSTRACT

COOKSTOVE STARTUP MATERIAL CHARACTERIZATION AND QUANTIFICATION AND ACUTE CARDIOPULMONARY EFFECTS FROM CONTROLLED EXPOSURE TO COOKSTOVE AIR POLLUTION

Approximately three billion people burn solid fuel as their source of domestic energy, primarily in inefficient, poorly ventilated cookstoves. Exposure to air pollution resulting from this practice is a leading environmental health hazard and one of the top risk factors for disease globally. To help solve this problem, research has been focused on designing and disseminating cookstove technologies intended to operate more efficiently and emit less pollution and assessing the impact of these technologies on emissions and health. The underlying hypothesis is that use of improved cookstoves will reduce exposures and subsequent health burden compared to existing cookstoves. However, research to date has not effectively tested this hypothesis. While some studies suggest reductions in cardiovascular and respiratory health endpoints with reduced exposure to cookstove emissions, other work has shown very little impact of improved stoves on health.

This dissertation addresses two gaps in cookstove exposure and health science. The first gap pertains to emissions from cookstoves. Cookstove emissions are poorly quantified and heterogeneous, yet these emissions are an important source of health-relevant air pollution. The different fuels, stove designs, and operating practices used across different regions of the world contribute to the heterogeneity in and variability of cookstove emissions. We identified stove startup practices as a potentially important contributor to cookstove emissions but an area where very little data existed. Limited laboratory data suggested that the cookstove ignition event (also referred to as startup) could contribute substantially to overall emissions, however it was unclear

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what the contribution of the startup fuel might be. Prior to this work, knowledge of the types of materials used for cookstove startup was largely anecdotal.

The second gap is our understanding of health responses following exposure to cookstove air pollution generated by different stove designs. While household air pollution is estimated to have a substantial burden on global respiratory and cardiovascular health, empirical evidence describing this relationship across a range of technology types and exposure levels is limited. Comparison of stove technologies in the laboratory has been largely limited to emissions. The relationships between air pollution exposures and health effects is typically assessed in the field using observational epidemiologic study designs. Observational field studies usually only evaluate a single stove type in a single population and are subject to confounding, which limits the generalizability of findings and yields questions regarding how incrementally improved stoves translate to specific health benefits.

To address the startup emissions gap, I designed and administered a survey to gather information about the types of materials used for cookstove startup and, based on the results of the survey, conducted laboratory emissions measurements of frequently identified startup fuels. The survey targeted cookstove experts in the academic and private sectors. Respondents provided information that covered 48 geographic locations across 22 countries. Results indicated that a variety of materials are used to start cookstoves, including many non-biomass materials that may have health-relevant combustion byproducts. Paper, plastic, agricultural wastes, kerosene and other petroleum-based accelerants, and rubber-like materials (e.g., tires, footwear) were the most frequently indicated startup materials. Additional materials mentioned included fabrics, plastic packaging, soda bottles, snack food wrappers, and trash.

Informed by the survey results, laboratory tests were conducted to measure emissions from the burning of kerosene, plastic bags, newspaper, fabric, food packaging, rubber tire tubes, kindling, footwear, and wood shims. Measured pollutants included fine particulate matter mass (PM_{2.5}), PM_{2.5} elemental and organic carbon, methane, carbon monoxide, carbon dioxide,

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benzene, and formaldehyde. Results demonstrated substantial variability in the measured emissions across materials on a per startup event basis. For example, kerosene emitted 496 mg PM_{2.5} and 999 mg CO per startup event, whereas plastic bags emitted 2 mg PM_{2.5} and 30 mg CO per startup event. When considering emissions on a per startup event versus per mass basis, the ordering of materials from highest-to-lowest emitter changes. This result emphasizes the importance of establishing how much material is used to start a stove in order to quantify startup emissions. Further, the proportional contribution of startups to overall emissions can vary substantially depending on startup material type and stove type. Comparing our results for startup materials can contribute substantially to a cookstove's overall emissions. Startup material choice may be especially important for cleaner stove-fuel combinations where the marginal benefits of reduced emissions are potentially greater.

To address the gap in our understanding of health response to different cookstoves, we conducted a controlled human exposure study to investigate acute responses in cardiovascular and respiratory health following exposure to cookstove-generated air pollution emissions. Forty-eight young, healthy subjects received six two-hour exposures: five cookstoves treatments (liquid petroleum gas [LPG], gasifier, fan rocket elbow, rocket elbow, and three stone fire) at PM_{2.5} concentrations proportional to the stove's relative emission levels (10 to 500 μ g/m³) and a filtered air control (0 μ g/m³). Health measurements were conducted immediately after exposure, three hours after exposure, and 24 hours after exposure.

Immediately post-exposure, systolic pressure was lower for the three stone fire (500 μ g/m³ PM_{2.5}) compared to the control (0 μ g/m³ PM_{2.5}) (-2.3 mmHg, 95% confidence interval [CI] -4.5, -0.1 mmHg). Systolic pressure was also lower for the other stove treatments but to a lesser extent. No changes in diastolic pressure were observed. Forced vital capacity (FVC) was lower (40 to 60 mL), forced expiratory volume in one second (FEV₁) was lower (24 to 117 mL), and mid-expiratory flow (FEF₂₅₋₇₅) was lower (68 to 116 mL/s) immediately after exposure for the fan rocket, rocket

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elbow, and three stone fire treatments (100, 250, and 500 μ g/m³ PM_{2.5}, respectively) compared to the control, though no changes in FEV₁/FVC were observed.

At three hours post-exposure, no differences in systolic or diastolic pressure were observed for stove treatments compared to the control. Decreases in FVC, FEV₁, and FEF₂₅₋₇₅, but not FEV₁/FVC, were observed for all five stove treatments compared to the control (FVC: 8 to 30 mL decreases; FEV₁: 39 to 68 mL decreases; FEF₂₅₋₇₅: 30 to 122 mL decreases) three hours after exposure.

Twenty-four hours post-exposure, systolic blood pressure was 2 to 3 mmHg higher for all treatments compared to the control except for the rocket elbow stove. The stove treatments had no effect on lung function 24 hours after exposure compared to the control.

Results suggest that both blood pressure and lung function are affected by exposure to cookstove emissions on acute timescales. For the most part, effects occurred at a similar magnitude across all treatments, suggesting no evidence of a PM_{2.5} exposure-response. Results for both blood pressure and spirometry support an inflammatory pathway mechanism through which PM_{2.5} impacts health in the short-term. The controlled exposure design allowed for comparisons to be made across a wider range of PM_{2.5} exposures and stove types than is feasible in most field settings.

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Finally, thank you to my husband Derek. This dissertation would not exist without you! You encouraged me to come back to school and pursue my PhD. You believed in me when I wasn't so sure myself and provided the encouragement and support I needed to accomplish my goals – as well as the laughter I often needed to balance the times when everything started to feel too stressful. I could not ask for a better partner in life.

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DEDICATION

I dedicate this dissertation to my daughter, Alivia River.

I know you won't remember all the hours you spent "helping" me write this dissertation in your first few months of life, but I hope you learn a lesson from knowing it happened:

It is possible to do it all,

if you have the right people by your side for support.

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Acronyms

Acronym	Definition
ANCOVA	analysis of covariance
ATS	American Thoracic Society
BC	black carbon
BTEX	benzene, toluene, ethylbenzene, and xylenes
CAP	concentrated ambient particles
CAPS	the "Cooking and Pneumonia Study," a cookstove intervention trial conducted in Malawi from 2013 to 2018
CH ₄	methane
CI	confidence interval (statistical); typically a 95% CI
CO	carbon monoxide
CO ₂	carbon dioxide
COPD	chronic obstructive pulmonary disease
COV	coefficient of variation
DALY	disability-adjusted life year
EC	elemental carbon
EPA	Environmental Protection Agency
ERS	European Respiratory Society
FEF ₂₅₋₇₅	mid-flow forced expiratory flow rate average from the 25^{th} to 75^{th} percentile of the FVC
FEV ₁	forced expiratory volume in the first second of expiration
FVC	forced vital capacity
HEPA	high-efficiency particulate air
ISO	International Standards Organization
LPG	liquefied petroleum gas

Acronyms, continued.

Acronym	Definition
MCE	modified combustion efficiency; the ratio of carbon dioxide to the sum of carbon dioxide and carbon monoxide in a combustion-emissions plume
NAAQS	National Ambient Air Quality Standards
NO	nitrogen oxide
NO ₂	nitrogen dioxide
NO _x	nitrogen oxide and nitrogen dioxide
O ₃	ozone
OC	organic carbon
OM	organic matter
PEF	peak expiratory flow
PM ₁₀	particulate matter less than 10 micrometers in aerodynamic diameter
PM _{2.5}	fine particulate matter less than 2.5 micrometers in aerodynamic diameter
RAAS	renin-angiotensin-aldosterone system; a hormone signaling pathway involved in blood pressure regulation
RESPIRE	the "Randomized Exposure Study of Pollution Indoors and Respiratory Effects" study, a cookstove intervention trial conducted in Guatemala from 1991 to 2011
SO ₂	sulfur dioxide
STOVES	the "Subclinical Tests on Volunteers Exposed to Smoke" Study, a controlled exposure study conducted in Fort Collins, Colorado from 2016 to 2018
UFP	ultrafine particles
VOC	volatile organic compounds
WHO	World Health Organization

Measurement Units

Unit	Definition
%	percent
cm	centimeter

Units of Measurement, continued.

g	grams
g/kg	grams per kilogram; emissions factor calculated as the mass of pollutant emitted divided by to the mass of fuel consumed
g/MJ	grams per megajoule; emissions factor calculated as the mass of pollutant emitted divided by the useful energy output
hr	hour
Hz	hertz; frequency unit of one cycle per second
in	inch
kg/m²	kilograms per square meter; the unit of measurement for body mass index
L	liters
L/min	liters per minute
m	meters
mg	milligrams
mg/m³	milligrams per cubic meter
mg/startup	milligrams per startup event; emissions factor calculated as the mass of pollutant emitted divided by the mass of startup fuel consumed
min	minutes
$MJ_{\text{delivered}}$	megajoules delivered; the amount of useful energy transferred to the cooking vessel by a cookstove
mL	milliliters
mL/s	milliliters per second
mmHg	millimeters mercury; the unit of measurement for blood pressure
µg/m³	micrograms per cubic meter
μm	micrometer
ppb	parts per billion
ppm	parts per million
SD	standard deviation

CHAPTER 1: INTRODUCTION

Summary and Significance

Approximately three billion people use solid fuel as their source of domestic energy (Bonjour et al. 2013). Most solid fuel users rely on inefficient and poorly ventilated cookstove systems, which contribute to elevated levels of household air pollution. This household air pollution is a leading environmental health hazard and one of the top risk factors for disease globally (Gakidou et al. 2017). These exposures contributed to approximately 77 million disability-adjusted life years in 2016, including over 2.5 million premature deaths, primarily through cardiovascular and pulmonary disease (Gakidou et al. 2017).

Given the size of this problem, much work has focused on designing and disseminating improved or clean cookstove technologies that are affordable, practical, burn more efficiently, and emit less pollution. The driving hypothesis is that use of clean cookstoves will reduce exposures and improve health compared to traditional cookstoves. However, research done to date has not widely tested this hypothesis. A variety of non-profit and government agencies are investing resources into clean cookstove adoption (Global Alliance for Clean Cookstoves 2016; Martin et al. 2011) without a clear understanding of whether existing clean cookstove technologies are truly clean enough to decrease the health burden if widely implemented.

Current guidelines for evaluating stove performance are based on emissions of fine particulate matter (PM_{2.5}) and carbon monoxide (CO), the two most characterized components of emissions (ISO 2012; Jetter et al. 2012). While PM_{2.5} and CO emissions are health-relevant, many other health-relevant pollutants are known or suspected to be emitted from cookstoves. Many factors play a role in determining the emissions from cookstoves (e.g., fuel type, stove type, operating procedure), but comprehensive emissions data that accounts for these factors is lacking.

Additionally, the relationships between household air pollution exposures and the range of possible cardiopulmonary health effects are not fully understood (Gordon et al. 2014; Smith and Peel 2010). Though some field-based studies suggest that improvements in cardiovascular endpoints (e.g., blood pressure, electrocardiogram features) and respiratory endpoints (e.g., pneumonia, lung function decline) occur with reduced exposure to cookstove emissions (e.g., Alexander et al. 2017; Clark et al. 2013a; McCracken et al. 2011; McCracken et al. 2007; Smith et al. 2011), other work has shown very little impact of improved stoves on health (e.g., Clark et al. 2009; Martin et al. 2011; Mortimer et al. 2017; Romieu et al. 2009; Smith et al. 2011). Field-based studies typically investigate only a single stove type in a single population and can be subject to considerable confounding. These issues limit the generalizability of findings and yield questions regarding how incrementally improved stoves translate to specific health benefits. A controlled exposure study conducted in a laboratory setting can provide information about between-stove differences in health endpoints that is not feasible to collect in the field. A controlled human exposure study has the advantage of reducing limitations of observational-based field studies such as confounding and other biases that impede the ability to establish an exposure-response relationship. While a controlled exposure study cannot capture chronic exposures and clinical health outcomes (e.g., heart attacks, lung disease development or exacerbation), measurement of translatable risk markers such as blood pressure and lung function allows for connection between the two types of data.

Aims

The specific aims of this dissertation were as follows:

Aim 1: Characterize cookstove startup material types and emissions

Anecdotal knowledge indicates that a variety of non-standard fuels such as plastic bags, agricultural waste, consumer products waste, and rubber tires are used to ignite cookstoves. These non-standard fuels may have different emissions than the standard fuels used during the main cooking period (e.g., charcoal, wood) and therefore could be important for health. Yet, data

on the materials used to start stoves and their emissions is lacking. Elimination or substitution of startup fuels may be easier to implement globally than conversion to clean stoves; characterizing the types of fuels used and quantifying the emissions from these fuels is a critical step towards evaluating practical, feasible intervention options.

Aim 1a. Survey cookstove experts to determine common startup material types

Data on the materials used to start stoves is lacking. I designed and implemented an expert elicitation style survey to collect anecdotal knowledge about fuel types used during startup of cookstoves. Responses were analyzed using qualitative techniques to understand what startup fuel materials are used most commonly around the world.

Aim 1b. Conduct laboratory-based tests to measure emissions from startup materials

Several studies have noted the importance of the startup period in determining overall emissions (e.g., Bhattacharya et al. 2002; Carter et al. 2014; Lask and Gadgil 2017). Yet, no studies to date had systematically isolated startup emissions as a function of startup material type. Informed by the results of Aim 1a, I led an effort to characterize emissions from common startup materials: kerosene, plastic (e.g., plastic bags, packaging), newspaper, kindling, high-resin wood (i.e., *ocote*), fabric scraps, and rubber (e.g., tires, footwear). The testing protocol intended to replicate real-world startup events while simultaneously isolating emissions from the startup material to measure emissions of PM_{2.5} mass, PM_{2.5} elemental and organic carbon, CO, carbon dioxide (CO₂), methane, carbonyls, benzene, toluene, ethylbenzene, and xylenes.

<u>Aim 2: Assess differences in health endpoints following controlled exposures to emissions from</u> <u>various cookstove technologies</u>

Based on the hypothesis that some clean cookstoves will not improve relevant indicators of cardiopulmonary health compared to traditional stoves, I led a study to evaluate differences in acute markers of cardiovascular and respiratory health in human volunteers following exposures to emissions from various stove technologies. From October 2016 through January 2018, 48 human volunteers underwent two-hour exposures to emissions from five different stove

technologies and a clean air control, utilizing a crossover design. Treatments spanned $PM_{2.5}$ levels from 10 μ g/m³ to 500 μ g/m³. Technologies ranged from liquid petroleum gas (LPG) to the traditional three stone fire. Health endpoints were measured immediately post exposure and at three and 24 hours post-exposure. I conducted analysis of blood pressure and lung function.

Aim 2a. Blood Pressure

Blood pressure is an established preclinical marker of cardiovascular disease risk that has been shown to change meaningfully in acute time frames (Lewington et al. 2002; Turnbull 2003; Vasan et al. 2001). Increases in blood pressure, even within a normal range, can lead to increased risk of stroke, coronary heart disease, and heart failure (Ettehad et al. 2016). On a population level, a small increase in blood pressure will push a segment of the population from a pre-hypertensive to hypertensive state. Both ambient air pollution and cookstove-generated air pollution have been shown to have a relationship with blood pressure (Brook et al. 2010; Clark et al. 2013a; Fuks et al. 2014; Langrish et al. 2012; McCracken et al. 2007). In this study, brachial blood pressure was measured on the left upper arm with participants in a supine position after a minimum 10-minute rest period using an automated monitor (SphygmoCor XCEL, AtCor Medical Pty Ltd., Australia).

Aim 2b. Lung Function

Spirometry is a technique for characterizing respiratory function that is often used as part of the diagnosis for respiratory diseases such as chronic obstructive pulmonary disease, asthma, and interstitial fibrosis (Barreiro and Perillo 2004; Miller et al. 2005; Ranu et al. 2011). Lung function has been shown to be impaired by chronic exposure to ambient air pollution (e.g., Köpf et al. 2017; Li et al. 2012; Steinvil et al. 2009) as well as cookstove-generated air pollution (e.g., da Silva et al. 2012; Fullerton et al. 2011; Revathi et al. 2012). In this study, forced vital capacity [FVC], forced expiratory volume in one second [FEV₁], the ratio of FEV₁/FVC, and mid-expiratory flow [FEF₂₅₋₇₅] were measured using an ultrasonic spirometer (Easy on-PC, ndd Medizintechnik AG, Zurich, Switzerland).

CHAPTER 2: LITERATURE REVIEW

Cookstoves, Household Air Pollution, and Health: The Big Picture

Global Disease Burden

Nearly 40% of the world's population cooks over open fires or with rudimentary biomass-burning stoves (Bonjour et al. 2013). Exposure to household air pollution generated by this practice is a top-ten risk factor for disease globally (Gakidou et al. 2017). These exposures contributed to approximately 77 million disability-adjusted life years (DALYs) in 2016, including over 2.5 million premature deaths (Gakidou et al. 2017). The burden is primarily in the form of respiratory and cardiovascular diseases. Ischemic heart disease, ischemic stroke, and hemorrhagic stroke combined account for approximately half of this burden (estimated 29.8 million DALYs / 1.2 million deaths in 2016) (Gakidou et al. 2017; Smith et al. 2014). Household air pollution is the leading environmental risk factor for ischemic heart disease, the number one cause of death worldwide (Lim et al. 2012). Respiratory diseases make up most of the other half: lower respiratory infections, respiratory cancers, and chronic obstructive pulmonary disease were estimated to account for over 46 million DALYs, including 1.36 million deaths, in 2016 (Gakidou et al. 2017).

<u>Data Gaps</u>

Efforts to reduce the harmful effects of cookstove use have achieved only modest success (e.g. Alexander et al. 2017; Clark et al. 2013a; McCracken et al. 2011; McCracken et al. 2007). One reason for a lack of impact is that many improved cookstove designs that perform well under controlled laboratory settings do not perform as well in the real world (Roden et al. 2009). This is in part because laboratory testing often fails to account for the varied operating conditions in the real world, which can impact combustion dynamics and therefore emissions. Additionally, social factors impact clean stove adoption and use patterns which results in exposures that are different than anticipated. Further, despite numerous epidemiological studies conducted in the past few

decades, questions still remain regarding the level of exposure reductions necessary to improve health. The relationships between household air pollution exposures and the range of possible cardiopulmonary health effects are not fully understood (Gordon et al. 2014; Smith and Peel 2010). The connections between household air pollution and disease risk is primarily extrapolated from research on other pollution sources (i.e., active cigarette smoking, secondhand smoke, and ambient air pollution) (Burnett et al. 2014; Smith et al. 2014). More research is needed that explores emissions across a wider variety of cookstove practices, exposure levels, and health responses.

Gap 1: Emissions

Combustion of cooking fuels results in emissions of numerous air pollutants, including gas-phase and particle-phase species. Particulate matter (PM) and carbon monoxide (CO) are two of the most frequently measured pollutants in cookstove health studies. Emissions factors for PM and CO define the International Standard Organization's (ISO) cookstove performance tiers, which are intended to be guidelines for improved cookstove design (ISO 2018). However, it is unclear if these two pollutants are sufficient markers of household air pollution and adequately capture the health-relevant aspects of this multi-pollutant mixture. Further work to characterize a larger suite of compounds is needed.

Air Pollutants

Particulate Matter

Particulate matter air pollution is a mixture of solid and/or liquid particles that are suspended in air. Sources of PM in the atmosphere can be anthropogenic (e.g., vehicle exhaust, industrial processes, power plants, residential solid fuel use) or natural (e.g., volcanic emissions, sea spray) (Brook et al. 2004). Particulate matter can be generated as a primary pollutant from these sources or as a secondary pollutant that forms following reactions of gaseous pollutants (Brook et al. 2004).

Particulate matter encompasses a large range of particle sizes and morphologies, which impacts deposition in the airways. Common size distinctions are coarse PM (between 2.5 and 10 μ m in aerodynamic diameter [PM₁₀]), fine PM (less than 2.5 μ m [PM_{2.5}]), and ultrafine particles (typically 0.01 to 0.1 μ m [UFP]). Fine PM is of particular interest from a health perspective as it can be inhaled deeply into the lungs reaching small airways and alveoli and absorbed into the blood stream (Brook et al. 2004; Naeher et al. 2007). Combustion of biomass results in generation of a large number of particles smaller than 1 μ m, which further justifies the importance of the fine PM size distinction from an emissions characterization standpoint. The cardiovascular and pulmonary impacts of PM_{2.5} are summarized in subsequent sections.

Measurements of PM_{10} and $PM_{2.5}$ are typically reported as mass per volume air (μ g/m³). The U.S. Environmental Protection Agency's (EPA) regulatory limit for ambient $PM_{2.5}$ under the National Ambient Air Quality Standards (NAAQS) is 12 μ g/m³ annual mean (averaged over three years) or 35 μ g/m³ as the 98th percentile of the 24-hour average (averaged over three years) (U.S. Environmental Protection Agency). The World Health Organization (WHO) also designates guidelines for PM which, while voluntary, are more stringent: $PM_{2.5}$ annual mean of 10 μ g/m³ and 25 μ g/m³ for the 24-hour average (World Health Organization 2005). Use of traditional wood stoves can generate indoor air concentrations from 10 to more than 300 times higher than these levels (Johnson et al. 2011).

While particle mass concentration is health-relevant and therefore used to regulate emissions and define exposures, other particle characteristics such as composition, particle number, or morphology may also have health relevance (Brook et al. 2010). PM from biomass combustion sources typically includes a mixture of organic carbonaceous compounds (e.g., polycyclic aromatic hydrocarbons, aldehydes), elemental carbon, and trace metals and other ions (Brook et al. 2010; Naeher et al. 2007). The composition of PM emissions is expected to vary based on the type of biomass (e.g., different wood species, charcoal), fuel conditions (e.g., size,

shape, moisture content), and burn conditions (e.g., smoldering vs. flaming combustion) (Naeher et al. 2007). Yet, these compositional aspects have not been a major focus of research to date. *Carbon Monoxide*

Carbon monoxide (CO) is one of most commonly measured gaseous components of household air pollution (Brook et al. 2004; Naeher et al. 2007). Major sources of CO, aside from household burning of solid fuels, are gasoline-powered engines (used in vehicles as well as boats, lawnmowers, chainsaws, etc.), stationary combustion sources (e.g. power plants), and diesel-powered engines (Brook et al. 2004). The U.S. EPA's regulatory limits for CO under the NAAQS are 35 ppm for a one-hour average and 9 ppm for an 8-hour average (U.S. Environmental Protection Agency). By comparison, indoor air concentrations from the use of traditional wood stoves have been estimated to reach 24-hour averages of 20 to 40 ppm (Johnson et al. 2011).

Carbon monoxide impacts human health by binding to hemoglobin in the bloodstream; the affinity of hemoglobin to bind CO is considerably larger than the affinity for oxygen (Brook et al. 2004). Short-term exposures to high levels of CO can lead to asphyxiation; dizziness, confusion, and death can result. At lower levels, CO can interfere with oxygen release in tissues, leading to cellular hypoxia (Brook et al. 2004).

Carbonyls

Carbonyls are gaseous compounds known to cause acute eye and lung irritation; some carbonyls, including formaldehyde and acetaldehyde, are carcinogenic (Cogliano et al. 2005; International Agency for Research on Cancer 1999, 2012; U.S. Environmental Protection Agency 2018a). A major source of these compounds in the atmosphere is fossil fuel combustion (Wang et al. 2010; Zhang et al. 2012). Recent research has focused on ambient carbonyl concentrations in regions with heavy vehicle traffic and industrial activity, such as China and Korea (e.g., Seo and Baek 2011; Wang et al. 2015; Xu et al. 2010; Zhang et al. 2012).

Only a few studies have measured emissions of carbonyl compounds from cookstoves. Early work by Zhang and Smith (1999) found that formaldehyde and acetaldehyde were emitted

from a variety of stove/fuel combinations at levels estimated to result in indoor concentrations high enough for acute health effects. The few recent studies that have measured formaldehyde or acetaldehyde emissions were limited to wood fuels across and a small selection of stove types (e.g., Akagi et al. 2011; Hall et al. 2012; McDonald et al. 2000).

Volatile Organic Compounds

Anthropogenic volatile organic compounds (VOCs) are a broad class of gas-phase compounds that includes health-relevant benzene, toluene, ethylbenzene, and xylenes (collectively referred to as BTEX), which are common pollutants emitted from combustion of fossil fuels and other biomass. The BTEX compounds have been associated with a wide range of adverse impacts on reproduction, development, immune function, metabolic function, respiratory function, and cardiovascular function (Bolden et al. 2015; Olsson and Kjällstrand 2006; Piccardo et al. 2014). Benzene is designated as an A1 carcinogen according to the WHO and U.S. EPA (Baan et al. 2009; International Agency for Research on Cancer 2012; U.S. Environmental Protection Agency).

Previous work has indicated that benzene emissions from biomass burning in general can be considerable and occur at health-relevant levels (e.g., Akagi et al. 2011; Hall et al. 2012). Only a few studies, however, have characterized benzene or other VOC emissions from residential cookstoves and these few are limited in their scope of stove types, fuel types, and presentation of emissions factors for specific VOCs as opposed to categories of VOCs (e.g., Evtyugina et al. 2014; McDonald et al. 2000).

Cookstove Emission Measurements

A better understanding of how different cookstove technologies, fuels, and operating conditions impact total emissions and emission rates is critical for designing stoves and interventions that can reduce emissions and subsequent exposures. Several stove testing protocols exist (Arora and Jain 2016), which allow for standardized comparisons of emissions across technologies. Most work has focused on emissions of PM_{2.5} and CO across different main

fuels and stove designs, though some studies have also considered other health and climate-relevant pollutants (e.g., Brandelet et al. 2018; Carter et al. 2014; Jetter et al. 2012; Jetter and Kariher 2009; Lask and Gadgil 2017; Stockwell et al. 2015; Wathore et al. 2017). Few studies have investigated the impact of unique cooking practices on emissions.

Startup Materials

Most laboratory studies report using kerosene or highly-processed wood shims to ignite stoves, but these materials represent only a fraction of real-world startup materials (Arora and Jain 2016). Only one study was identified that compared emissions from lighting stoves with different startup materials. Based on the hypothesis that different startup materials may affect emissions, Arora et al. (2014) conducted tests using kerosene, wood chips, and mustard stalks to ignite two different cookstoves (forced draft and natural draft) as a sub-aim in their evaluation and comparison of two cookstove testing protocols. Researchers found that ignition with mustard stalks resulted in higher CO emissions as compared to wood chips or kerosene; this difference was more pronounced with a forced draft stove than a natural draft stove. When ignition with these different materials was included along with the main water boiling task in the calculation of overall emissions rates for the stoves, CO emission factors were lowest for kerosene-based ignition (0.9 ± 0.1 g/MJ and 1.4 ± 0.4 g/MJ, depending on the stove) and PM emissions were lowest for wood chip ignition (71.3 ± 5.6 mg/MJ and 184.3 ± 8.1 mg/MJ). Emissions were highest for both PM and CO when stoves were ignited using mustard stalks (PM: approximately 300 mg/MJ; CO approximately 3 g/MJ).

Several laboratory studies have noted the contribution of stove ignition to overall cooking emissions and demonstrated that the contribution can depend on the startup fuel type, method of igniting, and main fuel (e.g., Arora et al. 2014; Bhattacharya et al. 2002; Carter et al. 2014; Lask and Gadgil 2017; Wathore et al. 2017). However, these studies do not isolate emissions of the startup fuel from those of the main fuel and the types of startup materials used is limited.

Carter et al. (2014) reported that lighting using dry red pine hardwood contributed to 34 to 45% of overall PM_{2.5} emissions among Chinese gasifier stoves burning pelletized biomass main fuel. The authors' conclusion from this result is that the startup (lighting) process is contributing substantially to the overall emissions from a cooking event. The "lighting phase" was defined as the time from when the startup fuel was added to the stove's main fuel bed and lit until the time when the stove's flame was considered stable. This limitation means that we cannot say if emissions from the startup material makes a substantial contribution to the total emissions during this phase.

Wathore et al. (2017) measured in-field emissions from traditional and improved forced-draft gasifier style cookstoves, natural draft clay cookstoves, and traditional three stone fires/mud stoves in Malawi. Researchers noted a high peak in particle light scattering that occurred at startup for all stoves and determined that PM emissions during startup likely were large contributors to overall PM emissions during cooking. For the forced draft gasifier stoves, the contribution of the startup to overall PM emissions was considerably greater than for other stove types. The authors did not report whether the startup process involved a fuel type different from the main fuel (wood).

Two studies were identified that focused on how the method of lighting can impact emissions. Bhattacharya et al. (2002) demonstrated that lighting a stove with kerosene-soaked kindling from the top of the fuel bed resulted in decreased emissions of CO and NO_x compared to when lighting from the bottom of the fuel bed. Lask and Gadgil (2017) showed that using a lighting cone with charcoal stoves reduced emissions of ultrafine particles and CO. These studies provide evidence that the startup process is an important consideration for cookstove emissions, however more work is needed to understand how different startup fuels specifically contribute to overall emissions.

Gap 2: Cardiovascular Health

Air Pollution and Cardiovascular Disease

Approximately half of the global disease burden from household air pollution is estimated to be in the form of ischemic heart disease, ischemic stroke, and hemorrhagic stroke (Gakidou et al. 2017; Smith et al. 2014). However, few studies have investigated a relationship between household air pollution and major cardiovascular events such as cardiovascular-related death, heart attack, and stroke; instead, the association is primarily extrapolated from studies of outdoor air pollution and cigarette smoke, with a focus on PM_{2.5}-linked effects (Burnett et al. 2014; McCracken et al. 2012; Smith and Peel 2010).

Large cohort and time-series studies conducted in the U.S. and Europe, such as the Harvard Six Cities and American Cancer Society (ACS) cohorts (Dockery et al. 1993; Pope et al. 1995), the National Morbidity, Mortality, and Air Pollution Study (NMMAPS) (Samet et al. 2000a; Samet et al. 2000b), and the Air Pollution and Health: A European Approach studies (APHEA and APHEA-2) (Analitis et al. 2006; Katsouyanni et al. 1997), have demonstrated increased relative risks for all-cause mortality, cardiovascular mortality, cardiopulmonary mortality, and deaths from ischemic heart disease associated with ambient PM_{2.5}. Nitrogen dioxide (NO₂), sulfur dioxide (SO₂), CO, and ozone (O₃) have also been shown in some studies to have independent relationships with cardiovascular mortality (Brook et al. 2010). Overall, the literature indicates approximately 10% increases in all-cause mortality risk and up to 76% increases in cardiovascular disease mortality per 10 µg/m³ increase in PM_{2.5} (Brook et al. 2010). The range of clinical cardiovascular events associated with ambient levels of air pollution across the epidemiologic literature include ischemic heart disease, arrhythmia, heart failure, cardiac arrest, ischemic stroke, and peripheral vascular disease (Brook et al. 2010). Epidemiologic studies have demonstrated associations between air pollution and subclinical cardiovascular endpoints, including changes in markers of systemic inflammation, oxidative stress, and atherosclerosis, vascular and endothelial dysfunction, changes to heart rate variability and blood pressure, and changes in markers of thrombosis and blood coagulation (Brook et al. 2010). Given the large known impacts of ambient air pollution, the extrapolated impact from household air pollution – which occurs at much higher exposure levels – warrants attention. However, given the expected nonlinear curve for most air pollution exposure-response functions (Burnett et al. 2014), work to characterize the relationships at levels relevant to cookstove use is needed.

Blood Pressure as a Metric for Cardiovascular Disease

Blood pressure is an established preclinical marker of cardiovascular disease risk (Lewington et al. 2002; Turnbull 2003; Vasan et al. 2001). Several controlled exposure studies of ambient air pollution sources have demonstrated that blood pressure can increase immediately (within minutes) by 2 to 6 mmHg during exposure to concentrated ambient particles in the 100 to 200 µg/m³ PM_{2.5} range (Brook et al. 2009; Byrd et al. 2016; Urch et al. 2005). Small increases in blood pressure, even within the normal range, can lead to increased risk of stroke, coronary heart disease, and heart failure. In a meta-analysis conducted by Lewington et al. (2002), a 2 mmHg decrease in blood pressure was estimated to reduce the risk of stroke mortality by 10% and the risk of ischemic heart disease mortality and other vascular disease mortality by 7%. Similarly, a meta-analysis by Ettehad et al. (2016) determined that a 10 mmHg reduction in systolic pressure resulted in a 10% reduced risk of cardiovascular disease events, 17% reduction in coronary heart disease risk, 27% reduction in stroke risk, and 28% reduction in heart failure risk. While cardiovascular risks are higher among populations with high blood pressure, risk reductions in major cardiovascular events, coronary artery disease, and all-cause mortality are demonstrated with reductions in blood pressure even in populations with low mean blood pressure (systolic less than 130 mmHg) (Ettehad et al. 2016).

Mechanisms for PM_{2.5}'s Impact on Cardiovascular Disease, Specifically Blood Pressure

Blood pressure is a measure of the force exerted on artery walls as blood is pumped through the vasculature. Blood pressure is maintained in the process of balancing cardiac output, which is determined by heart rate, stroke volume, and peripheral resistance. The heart reacts to demands and changes in cardiac output primarily through neural responses (autonomic nervous system, including sympathetic and parasympathetic pathways) and humoral responses (endocrine system) that encourage vasodilation (to decrease pressure) or vasoconstriction (to increase pressure) (Vila et al. 2015). Short term changes in blood pressure are regulated primarily by the sympathetic nervous system and the renin-angiotensin-aldosterone (RAAS) system (Chopra et al. 2011). Baroreceptors in blood vessels are mechanically activated by high pressure and inhibit sympathetic output, resulting in bradycardia (decreased heart rate) that reduces cardiac output and lowers pressure; conversely, baroreceptors' response to low pressure results in tachycardia (increased heart rate) that increases cardiac output and raises pressure (Chopra et al. 2011). RAAS activation mechanisms involve changes in the circulating levels of hormones that bind to receptors that have vasoconstrictor properties (Chopra et al. 2011). Long term, increases in blood pressure can also be due to factors such as increased arterial stiffness (a result of activation of the RAAS pathway or inflammation that impairs smooth muscle), which reduces vessels' ability to dilate during systole and constrict during diastole and thereby maintain blood flow and central pressure (Dumor et al. 2018).

There are three main biological pathways through which the fine particulate matter fraction of air pollution (PM_{2.5}) is proposed to trigger systemic vascular dysfunction and lead to increases in blood pressure that may lead to cardiovascular events as noted above (Brook et al. 2010). First, PM_{2.5} that enters lung tissues can trigger pro-inflammatory responses such as increased cytokine expression, which leads to systemic oxidative stress and inflammation. Second, particles can interact with receptors in the lungs that activate the autonomic nervous system to favor sympathetic rather than parasympathetic responses. Third, PM_{2.5} can enter into the blood stream and circulate throughout the body, interacting with other circulating molecules that regulate endothelial functioning and cell signaling. Each of these pathways can potential result in vasoconstriction and endothelial dysfunction, which can increase blood pressure.

Epidemiologic Studies on Household Air Pollution and Blood Pressure

Several observational studies have demonstrated associations between blood pressure (primarily systolic pressure) and cookstove-generated exposures (e.g., Alexander et al. 2015; Alexander et al. 2017; Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018; Burroughs Pena et al. 2015; Clark et al. 2011; Clark et al. 2013a; Dutta and Ray 2012; McCracken et al. 2007; Neupane et al. 2015; Norris et al. 2016). However, most of these studies were cross-sectional in design, only measured blood pressure on one or two occasions per participant, and only conducted 24- or 48-hour exposure sampling for a subset of participants, if any (e.g., Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018; Burroughs Pena et al. 2015; Clark et al. 2011; Clark et al. 2013a; Neupane et al. 2015). Those that have included exposure assessment have found mixed results concerning associations with PM_{2.5}, CO, and/or black carbon. Additionally, several of these studies found evidence of effect modification, with impacts of stronger magnitude for older populations (over 50 years) than younger (Alexander et al. 2015; Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018; Clark et al. 2013a). This is important because it suggests that health impacts may be different with chronic/continued exposure over a lifetime and emphasizes the importance of the study population when interpreting results.

Two randomized control intervention trials have been conducted. McCracken et al. (2007) found that use of an improved stove with a chimney, compared to an open wood fire, resulted in lower personal 24-hour average $PM_{2.5}$ exposures (264 µg/m³ versus 102 µg/m³) among 120 Guatemalan women. Use of an improved stove with a chimney versus open wood fire was associated with lower systolic (-3.7 mmHg, 95% CI -8.1, 0.60) and diastolic (-3.0 mmHg, 95% CI -5.7, -0.4) pressure one year post-intervention (average 293 days, range, 2 to 700 days). Alexander et al. (2017) found that diastolic pressure was lower among pregnant Nigerian women who switched from kerosene or wood to ethanol fuels (2.8 mmHg lower on average at the last follow-up visit, approximately 38 weeks gestational age) compared to controls who continued to

cook with either kerosene or wood. However, no differences were seen in systolic pressure between the intervention and control groups throughout pregnancy or at final follow-up.

Burroughs Pena et al. (2015) analyzed the baseline health data from a longitudinal cohort of 1004 Peruvian men and women over 35 years old, approximately half of whom reported daily use of wood or dung fuel for cooking or lighting purposes for more than six months at any point in their lifetime. Compared to non-users, biomass fuel users had a 7.0 mmHg increase in systolic pressure (95% CI 4.4, 9.6) and a 5.9 mmHg increase in diastolic pressure (95% CI 4.2, 7.6). Additionally, biomass fuel users had a 5.0 times higher risk of pre-hypertension (95% CI 2.6, 9.9) and 3.5 times higher risk of hypertension (95% CI 1.7, 7.0) (Burroughs Pena et al. 2015). Dutta and Ray (2012) similarly measured an increased prevalence of pre-hypertension and hypertension among rural Indian women who used biomass fuels (734 women) compared to those who used LPG (452 women).

Baumgartner et al. have reported associations between household air pollution exposures and blood pressure in cross-sectional analyses of two populations of rural Chinese women (Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018). In an analysis of 280 women from 235 households whose primary exposure source was biomass fuels, researchers observed an association between 24-hour personal PM_{2.5} measurements and systolic, but not diastolic, blood pressure (systolic: 2.2 mmHg increase per natural log unit increase in PM_{2.5} [95% CI 0.8, 3.7 mmHg]; diastolic: 0.5 mmHg increase per natural log unit increase in PM_{2.5} [95% CI -0.4, 1.3 mmHg]) (Baumgartner et al. 2011). Relationships were stronger for women over 50 years old (systolic: 4.1 mmHg, 95% CI 1.5, 6.6; diastolic: 1.8 mmHg, 95% CI 0.4, 3.2). Blood pressure measurements were conducted in both winter and summer to capture differences in exposure across the seasons; the PM_{2.5} average was 55 μ g/m³ (range 9 to 492, median 52, interquartile range 61) in summer and 117 μ g/m³ (range 22 to 634, median 105, interquartile range 120) in the winter. Stronger associations were observed for women who lived near highways, indicating co-exposure effects (Baumgartner et al. 2014). Additionally, relationships were stronger when exposure was defined by the black carbon fraction of $PM_{2.5}$ (systolic: 4.3 mmHg, 95% CI 2.3, 6.3; diastolic 1.3 mmHg, 95% CI 0.2, 2.4) (Baumgartner et al. 2014). In a similarly designed study of 205 women from 204 households, associations were observed between 48-hour personal air pollution exposures (ranged from 14 to 1405 μ g/m³) and systolic pressure (2.4 mmHg higher per natural log unit change in PM_{2.5}; 95% CI not reported) but not diastolic pressure measured in both summer and winter (Baumgartner et al. 2018). When stratified by age, the effect persisted in women over 50 years old only (3.5 mmHg, 95% CI 0.0, 7.1).

Building from the Baumgartner et al. (2014) study, Norris et al. (2016) measured personal black carbon exposure among 45 Indian women who cooked primarily with wood in traditional stoves or rocket stove designs. Blood pressure was measured throughout cooking sessions in both winter and summer; black carbon was averaged over two to 20 minutes prior to blood pressure measurement as well as over the full cooking session prior to blood pressure measurement. Results indicated that short-term black carbon exposures were associated with increases in systolic pressure (0.2 [95% CI -1.7, 2.2] to 1.9 [95% CI -0.8, 4.7] mmHg per interquartile range increase in black carbon over six to 20 minutes averaging periods). Across an entire cooking session, interquartile range increases in black carbon were associated with 2.6 mmHg increases in systolic pressure (95% CI -4.1, 9.3). Increases in black carbon exposure (both acute and over a full cooking period) were associated with small, non-significant decreases in diastolic pressure.

Neupane et al. (2015) considered the relationship between CO and blood pressure in a cross-sectional study of women in Nepal who had primarily used biogas fuels (219 women) or wood (300 women) for the past ten years. Twenty-four hour average kitchen CO was lower among biogas users than firewood users; use of biogas was associated with lower systolic pressure and reduced odds of hypertension among older women (more than 50 years old) only (9.8 mmHg, 95% CI -20.4, 0.8; odds ratio 0.32 95% CI 0.14, 0.71).

Clark et al. (2011) similarly observed increases in systolic pressure with increased 48-hour indoor CO (1.78 mmHg per 24 ppm increase in CO, 95% CI -1.25, 4.81) and 48-hour personal CO (1.89 mmHg per 2 ppm increase in CO, 95% CI -0.48, 4.26) in a cross-sectional analysis of one-time measurements for 124 Nicaraguan women using traditional open-combustion wood stoves. No relationship was observed between blood pressure and 48-hour indoor PM_{2.5}. One year after intervention with a chimney stove among 74 of these women, Clark et al. (2013a) did not observe changes in systolic or diastolic blood pressure (systolic: -1.5 mmHg 95% CI -4.9, 1.8; diastolic: 0 mmHg 95% CI -2.1, 2.1), despite marked reductions in 48-hour average kitchen PM_{2.5} and CO measured among a subset of the women at follow-up (PM_{2.5} average reduced from 1801 µg/m³ to 416 µg/m³ in sample of 25, CO reduced from 25.8 ppm to 7.2 ppm in sample of 32). Subgroup analysis restricted to women greater than 40 years old, however, resulted in a 5.9 mmHg reduction in systolic blood pressure (95% CI -11.3, -0.4). Conversely, Alexander et al. (2015) observed a 4.8% decrease in systolic blood pressure (5.5 mmHg, from 114.5 to 109.0 mmHg) measured either during or immediately after cooking among 28 women in Bolivia, along with a 24-hour average kitchen PM_{2.5} reduction from 240 to 48 µg/m³ in a subset of 15 women, one year after an intervention that involved changing from indoor open-pit fires to better-insulated wood-burning stoves with chimneys. No change in diastolic pressure was observed. Notably, Clark et al. (2013a) found that about half of the participants still used their traditional stove whereas adoption in Alexander et al. (2015) was around 90%. In both studies, researchers observed potential effect modification or interaction with age, with an effect observed among older women but not younger when the population was stratified. Both studies were limited by the lack of a control arm to account for any time-variant confounding and measurements (of both health and exposure levels) and measurements were conducted on just single occasions at the pre-intervention and post-intervention times.

While these observational studies provide general support for a relationship between exposure to household air pollution and blood pressure, they are still limited in their ability to

provide information across a broad range of exposure levels, stove types, and populations, which is important for designing public health interventions or policies aiming to reduce cookstove-associated exposures. Given the range of observed responses across populations within field studies, more work is needed to understand the responses expected across stove types and exposure levels.

Gap 3: Respiratory Health

Air Pollution and Respiratory Health

The respiratory effects from exposure to household air pollution, including lower respiratory infections, respiratory cancers, and chronic obstructive pulmonary disease (COPD), were estimated to account for over 46 million DALYs, including 1.36 million deaths, in 2016 (Gakidou et al. 2017). Gordon et al. (2014) review the evidence of household air pollution exposures and respiratory risks: increased risk of acute lower respiratory infections (e.g., pneumonia, bronchiolitis, and potentially tuberculosis) among children and possibly adults, as well as altered clinical course of respiratory infections; nearly doubling of the risk of COPD and increased COPD exacerbations; likely increased risk of asthma development and increased asthma exacerbations; and potentially associations with lung cancer. Unlike cardiovascular disease, a more substantial body of work exists specifically linking household air pollution to respiratory health, including studies on COPD, lung cancer, and acute lower respiratory infections (particularly among children). This means that exposure-response and health burden estimates do not rely as heavily on extrapolation of data from other air pollution types (Smith et al. 2014). Still, while a large body of literature supports an association between household air pollution and various respiratory health risks, evidence is lacking on the exposure-response relationship (Gordon et al. 2014). Evidence from ambient air pollution and smoking strongly support biological mechanisms for an effect, particularly for lung cancer and COPD, and have been used to develop integrated exposure-response functions spanning the exposure range of cookstoves (Burnett et
al. 2014; Gordon et al. 2014). More information is needed specific to household air pollution to validate these response functions.

Three intervention trials have been conducted on clinical respiratory outcomes and improved stove use. In the RESPIRE trial in Guatemala, researchers found a non-significant decrease in relative risk of physician-diagnosed pneumonia among children up to 18 months old who lived in intervention households that received chimney stoves compared to children in homes using traditional open fires (Smith et al. 2011). Relative risks for fieldworker-assessed pneumonia, physician-diagnosed severe pneumonia, and RSV-negative pneumonia were all significantly lower in the intervention group. Notably, exposure distributions between the control and intervention group overlapped, likely resulting in measurement error in the categorical analysis that would bias results towards null. The CAPS trial in Malawi found no differences in pneumonia risk among children less than five years old who lived in homes using the intervention gasifier stove compared to children in homes using open fires (Mortimer et al. 2017). However, overall adoption of the stove was low. In rural Mexico, women who received an improved stove intervention had lower risk of reporting respiratory symptoms (e.g., cough, wheeze, difficulty breathing) as well as decreased decline in spirometry values over time compared to women who continued to use open fires (Romieu et al. 2009); adherence to the intervention was low. These studies overall present evidence that reductions in respiratory disease are possible with improved stoves. However, these studies also demonstrate the difficulty in achieving the lower air pollution exposures with the interventions tested. More work is needed to understand the potential for respiratory health improvements with different stove technologies when not conflated by issues with adoption and overlapping exposures.

Spirometry as a Metric for Respiratory Health

Spirometry is a useful tool for understanding general pulmonary function and diagnosing respiratory diseases (Miller et al. 2005). Lung function metrics determined during a spirometry test can be used to help differentiate between restrictive and obstructive patterns of lung function

impairments (Dempsey and Scanlon 2018; Lange et al. 2009). Restrictive ventilatory patterns – defined as lowered forced vital capacity (FVC) and forced expiratory volume in the first second (FEV₁) without changes in the FEV₁/FVC ratio – occur in diseases like interstitial fibrosis and chest wall deformities (Barreiro and Perillo 2004; Ranu et al. 2011). Restrictive patterns have been observed in cigarette smokers and populations with high prevalence of biomass fuel use and have been associated with increased inflammatory markers, hypertension, and cardiovascular disease (Barreiro and Perillo 2004; Godfrey and Jankowich 2016; Jankowich et al. 2018). Obstructive ventilatory patterns – identified by lowered FEV₁/FVC ratios and reduced FEV₁ – are seen in diseases such as asthma and COPD (Averame et al. 2009; Barreiro and Perillo 2004; Mohamed Hoesein et al. 2011; Ranu et al. 2011). Reduced mid-expiratory flow rate (FEF₂₅₋₇₅) is also indicative of obstruction, though this metric is highly dependent on FVC (Barreiro and Perillo 2004). While spirometry is regularly used in individual clinical settings for disease diagnosis and management, it has also been employed in a range of air pollution epidemiological studies as a marker of respiratory status in both healthy and impaired populations (e.g., Forbes et al. 2009; Köpf et al. 2017; Li et al. 2012; Steinvil et al. 2009).

Mechanisms for PM_{2.5}'s impact on Respiratory Health, Specifically Spirometry

The primary mechanism responsible for determining static lung volume is the balance of elastic recoil between the lungs and the chest wall (Lumb 2017b). Breathing is controlled by a number of muscles, most notably the diaphragm (Lumb 2017a). During inspiration, the diaphragm contracts, increasing the rib cage and lung volume and decreasing intrapulmonary pressure (Lumb 2017a). Accessory muscles of the chest and intercostal muscles along the rib cage assist with chest expansion, particularly under increased respiratory loading and ventilation rates (e.g., exercise) (Lumb 2017a). Expiration occurs primarily due to elastic recoil of the lungs, however, the weight of the abdominal muscles also creates increased pressure which can displace the diaphragm; additional contraction of abdominal and other expiratory muscles can oppose the

force of the rib cage muscles to push air from the lungs (Lumb 2017a). Tension and contraction of respiratory muscles is controlled by neuronal activity (Lumb 2017a).

Physical impedance or hindrance of the respiratory system can result from elastic resistance (e.g., resistance from the lung tissue, or alveolar surfaces) or non-elastic resistance (e.g., frictional resistance along the small airways) (Lumb 2017b). A variety of disease states can reduce maximum inspiration volume and expiratory flow, by narrowing or obstructing airways as a result of inflammation or reduced elastic recoil, which increases resistance and changes pressure gradients (Lumb 2017c, d).

Mechanisms for the effect of PM on the respiratory system have been well studied and documented through toxicological laboratory studies: deposition of inhaled particles onto respiratory tract surfaces initiates a cascade of cellular injury and inflammation (U.S. EPA 2009). Tissue damage and irritation throughout the respiratory pathway as a result of this PM deposition can lead to development and exacerbation of obstructive lung diseases like COPD and asthma, as well as gene mutations and tumorogenesis responsible for development of lung/respiratory cancers (Gordon et al. 2014; Perez-Padilla et al. 2010). The mechanisms may be through the contribution of PM to reactive oxygen species (ROS) in the respiratory tract, either directly through the oxidative potential of PM components (e.g., metals, organic species) or indirectly through stimulation of pulmonary cells leading to ROS production (U.S. EPA 2009). Reactive oxygen species in the pulmonary tract activate cell signaling pathways that stimulate the release of inflammatory and immune response molecules (e.g., cytokines, chemokines, proteases), which in turn triggers inflammatory responses in the lung tissues (U.S. EPA 2009). Additionally, oxidative stress and inflammation in the pulmonary system can increase susceptibility to bacterial or viral infection and lead to altered infection responses (Gordon et al. 2014; Perez-Padilla et al. 2010). Further, fine particles can deposit deep into the airways and translocate systemically, which can lead to systemic inflammation and non-respiratory effects such as cardiovascular impacts (Brook et al. 2010; Gordon et al. 2014; Perez-Padilla et al. 2010).

Epidemiologic Studies on Household Air Pollution and Spirometry

Cross-sectional field studies generally support that cookstove-generated air pollution has the potential to reduce lung function, particularly FVC and FEV₁ (da Silva et al. 2012; Desalu et al. 2010; Fullerton et al. 2011; Ibhafidon et al. 2014; Regalado et al. 2006; Revathi et al. 2012), though evidence is inconsistent as other similar studies find no associations between lung function and cookstove exposures (Clark et al. 2009; Diaz et al. 2007; Rinne et al. 2006). Findings for FEV₁/FVC ratios across these studies are mixed. Longer-term intervention studies have been more consistent in demonstrating lessened decline in lung function following use of improved stoves (Romieu et al. 2009; Smith-Sivertsen et al. 2009; Zhou et al. 2014).

Cross-sectional studies comparing biomass (wood) users to LPG users generally show reduced FEV₁ and FVC among wood users; results for FEV₁/FVC ratios are inconsistent. Revathi et al. (2012) compared non-smoking women in India who used wood (n = 50) to age-matched controls who used LPG (n = 50) for at least five years. Wood users had lower percent predicted FVC (100.60% vs. 107.18%), FEV₁ (106.80% vs. 114.60%), and FEF₂₅₋₇₅ (70.1% vs. 78.8%), but not FEV₁/FVC ratios (86.27% vs. 87.49%), than the LPG users. The odds of having an obstructive lung function pattern compared to a normal pattern were 2.19 times higher among the wood users than the LPG users. In a similar design, da Silva et al. (2012) performed pulmonary function tests on 80 Brazilian adults (average 43 years; both male and female) who lived in homes that cooked with either LPG or biomass (primarily wood). In line with the results of Revathi et al. (2012), non-smokers who cooked with biomass had significantly lower percent predicted FEV₁ than non-smoking LPG users (biomass: 88.50%, 95% CI 84.73, 92.26 vs. LPG: 94.65%, 95% CI 93.03, 96.26). However in contrast to Revathi et al. (2012), FEV₁/FVC ratios were also lower among biomass users (biomass: 0.79, 95% CI 0.76, 0.82 vs. LPG: 0.85, 95% CI 0.82, 0.88). Researchers found a negative correlation between the duration of exposure (years) and FEV_1 (r = -0.46, p < 0.001) and the FEV₁/FVC ratio (r = -0.63). The decrease in pulmonary function among non-smokers who cooked with biomass compared to non-smokers using LPG was similar to the

decrease among smokers using LPG. Researchers also observed increased odds of reporting pulmonary symptoms including cough, dyspnea, wheezing, and eye itching/tears among indoor biomass users compared to LPG users. Regalado et al. (2006) also observed lower FEV₁/FVC ratios among Mexican women who used biomass stoves (n = 778; mean ratio 79.9%) compared to women who used LPG (n = 67; mean ratio 82.8%); multiple regression indicated a 2.8% reduction (95% CI -0.3, 5.3). However, no differences in FEV₁ or FVC were observed (mean FEV₁: 1.98 ± 0.53 L for biomass users vs. 2.08 ± 0.48 L for LPG users; mean FVC: 2.49 ± 0.64 L for biomass users vs. 2.52 ± 0.56 L for LPG users). Conversely, multiple regression analyses conducted among a subgroup who had PM₁₀ exposure measured during cooking (n = 410) indicated that high peaks of PM₁₀ (greater than 2.6 mg/m³) were associated with 81 mL lower FEV₁ (95% CI -0.5, -15) and 122 mL lower FVC (95% CI -24, -220), but no differences in FEV₁/FVC ratio (0.5%, 95% CI -1.7, 2.5) (Regalado et al. 2006).

Several studies have also compared wood to non-LPG alternative fuels and found similar lower lung function among wood users. A cross-sectional study by Fullerton et al. (2011) comparing 156 wood users to 141 charcoal users in Malawi (both male and female) found lower lung function among wood stove users. Mean FVC values were 3490 mL for charcoal users versus 3190 mL for wood users and mean FEV₁ values were 2780 mL and 2430 mL, respectively. Multivariate analysis of FEV₁ indicated a non-significant association between cooking with wood and FEV₁ (-120 mL, 95% CI -290, 50). Desalu et al. (2010) compared lung function among Nigerian women using exclusively biomass fuels, including wood, straw, dung, or crop residue (n = 161), to women using exclusively non-biomass fuels including gas, kerosene, or electricity (n = 108), and found that FEV₁ and FVC were lower among biomass users (FEV₁: 1.77 ± 0.49; FVC: 1.96 ± 0.52 L) than non-biomass users (FEV₁: 2.25 ± 0.69 L; FVC: 2.44 ± 0.75 L), but not the FEV₁/FVC ratio (90 ± 11.46% vs. 92 ± 10.89%). Another study in Nigeria, conducted by lbhafidon et al. (2014), measured lower FEV₁ and FVC among wood users (n = 35) compared to both kerosene users (n = 34) and LPG users (n = 21) (both men and women). Mean FEV₁ values

were 3.37 L among LPG users compared to 2.69 L among kerosene users and 2.12 L for LPG users; FVC values were 3.81 L for LPG users, 3.09 L for kerosene users, and 2.66 L for wood users (differences between groups all significant at p < 0.05 for both FVC and FEV₁). The percentage of individuals with normal lung function (defined as FEV₁/FVC greater than 0.7 and FEV₁ of at least 80% predicted) was lowest among wood users and highest among LPG users. Notably, 8-hour average PM₁₀ concentrations in homes using firewood (269 ± 93.7 µg/m³) were higher than in homes using LPG (80.8 ± 9.52 µg/m³). Concentrations in homes using kerosene (236.9 ± 26.5 µg/m³) were closer to that of firewood than LPG, though not significantly different than either.

Conversely, several cross-sectional studies failed to observe lung function changes. A cross-sectional study of 91 women in Ecuador found no differences in FVC, FEV₁, FEV₁/FVC ratios, or FEF₂₅₋₇₅ among women using biomass compared to women using LPG only (Rinne et al. 2006). However, children in households using biomass only compared to LPG had a 0.41 L decrease in FVC (95% CI -0.76, -0.07), 0.39 L decrease in FEV₁ (95% CI -0.78, -0.01), and 0.53 L/s decrease in FEF₂₅₋₇₅ (95% CI -1.35, 0.29). In Honduras, researchers found a 0.07 L (95% CI 0.01, 0.13) increase in FEV₁ per interquartile range increase in personal PM_{2.5} (106 μ g/m³) among wood stove users (n = 59 women, 38 using traditional stoves and 41 using improved Justa stove versus a traditional stove (as a categorical metric) was not associated with FEV₁, although the improved stoves were found to reduce 8-hour average personal PM_{2.5} by 73% and indoor CO by 87% compared to the traditional stoves (Clark et al. 2009).

Cross-sectional analysis of baseline measurements from the RESPIRE trial indicated no association between FVC or FEV₁ and exhaled breath CO among 319 Guatemalan women using open wood fires (Diaz et al. 2007). At follow-up over 6, 12, and 18 months post-intervention, no statistically significant associations were found between assignment of the plancha stove and FEV₁, FVC, or FEV₁/FVC (Smith-Sivertsen et al. 2009). However, when exposure was defined

continuously as exhaled CO in breath measured at the same time as spirometry was conducted during follow-ups, researchers found a 35 mL decrease in FEV₁ (95% CI -61, -9) and a non-significant 26 mL decrease in FVC (95% CI -57, 6) – yet no change in FEV₁/FVC – for each one unit-increase in natural log-transformed CO (Pope et al. 2015).

A nine-year prospective intervention in rural China found that women who used the biogas fuel intervention combined with improved ventilation (n = 287) saw a decline in FEV₁ of 18 mL/year, compared to 35 mL/year in those who continued to use biomass without ventilation (n = 160; 16 mL/year difference, 95% CI 9, 23) and 21 mL/year for those who used biomass but with improved ventilation (n = 81; 3 mL/year difference compared to biogas plus ventilation, 95% CI -6, 11) (Zhou et al. 2014). Results for FVC were similar: 32 mL/year declines for those who did not have either intervention, compared to 21 mL/year among those who used biogas without ventilation and 17 mL/year for those who used biogas and ventilation (16 mL difference, 95% CI 7, 25). FEV₁/FVC ratio was not significantly different between the two groups. In a study in Mexico, lung function decline over a year of follow-up was less among women who were given and used a Patsari stove intervention (n = 228) compared to those who continued to cook over an open wood fire (n = 198; adjusted difference in FEV₁: 31 mL, 95% CI 7, 55; FVC: 16 mL, 95% CI -21, 54; FEF₂₅₋₇₅: 67 mL/s, 95% CI -8, 141; FEV₁/FVC ratio: 0.5, -0.2, 1.1) (Romieu et al. 2009).

Controlled Exposure Studies

General Study Design

Controlled exposure studies are a type of experimental epidemiologic study that mimics clinical trials, in which researchers fix the exposure conditions of study participants under randomized intervention schemes (Rothman et al. 2008). Randomization serves to create groups of exposed and unexposed participants that differ only randomly in variable factors that might impact the study outcome (e.g., potential confounders), resulting ideally in an equal distribution of the variability in these factors across the exposed and unexposed populations. When successful randomization has occurred, assignment into the control or exposed group will be

independent of the outcome of interest, thereby allowing any variation seen in the outcome between the two groups to be attributable to the exposure (Rothman et al. 2008).

Crossover Designs

Crossover designs are a type of repeated measure study in which each participant receives each treatment in a randomized sequence order (Kenward 2015). The goal of a crossover, similar to the goal of a parallel study such as a randomized trial, is to compare the effect of the different treatments. As such, randomization of the treatment orders as opposed to treatments is important for allowing the effect of the sequence to be statistically washed out (Kenward 2015). Compared to other study designs, the crossover has the benefit of allowing comparisons across treatments within participants, as each participant undergoes each treatment. This allows for statistical efficiency and the elimination of time-invariant confounders. Limitations of crossover designs include most notably that the effects of the treatment must be transient and not persist from one study period to the next (i.e., the effect of the treatment given in the first period does not carry over into the second period).

Latin Square Randomization

Latin squares are useful designs for determining treatment orders in crossover studies with more than two treatment conditions. In a Latin square design, each participant receives each of the study treatments, so the number of treatments is equal to the number of periods (Kenward 2015). The period can also be thought of as the treatment number or visit number (e.g., first visit/treatment vs. second, third, etc.). When looking across the full set of sequences in a Latin square, each unique exposure/treatment level occurs only once in each period. This creates balance (when data is complete) that results in statistical efficiency (less observations are needed to achieve the desired power compared to a design that does not have this balance), as the effect of the treatment order can be encapsulated in sequence and period variables (Kenward 2015). The use of a Williams-style Latin square adds an additional component of balance in which every treatment is preceded in sequence by each other treatment an equal number of times when

considering all sequence groups (Williams 1949). This arrangement reduces the need to adjust for first-order carryover effects (which would reduce precision) when data is complete, since the carryover effect will cancel out when analysis is completed across all sequences, given an assumption of no carryover or treatment-by-period interactions (Kenward 2015). The Williams square is balanced such that period, sequence, and previous treatment will not be confounders (provided there is limited missing data). Additionally, time varying factors that may be different from one study session to the next (e.g., ambient pollution or meteorological conditions) will not occur with any propensity towards a specific treatment and therefore cannot be confounders, except by chance.

Analysis Methods for Crossover Studies

Mixed regression models are often used in repeated measures designs (including longitudinal studies and crossover design studies) where the repeated measurements within the same individual creates correlated data (Fitzmaurice and Laird 2015). Mixed models can also account for clustering that occurs when participants within trials are randomized in groups rather than individually (Fitzmaurice and Laird 2015). Clustering of data, either due to repeated measures within the same individual or grouping of individuals, results in correlation that violates most standard regression assumptions of independent data; random subject effects are incorporated into mixed models to account for this correlation (Fitzmaurice and Laird 2015). Linear mixed models, which are extensions of a standard linear model, are appropriate for use when the outcome data is continuous (Fitzmaurice and Laird 2015).

Wood Smoke Controlled Exposure Studies

Blood Pressure

Few controlled exposure studies exist on wood smoke exposures; investigation of blood pressure in these studies is limited. Unosson et al. (2013) found no changes in systolic or diastolic blood pressure over the course of one hour following a three-hour exposure to wood smoke at $300 \ \mu g/m^3 PM_{2.5}$ compared to a filtered air control exposure. However, they observed other acute

hemodynamic responses including changes to central arterial stiffness and heart rate variability, indicating vascular impairment and autonomic nervous system perturbation occurred as a result of wood smoke exposure. Evans et al. (2015) exposed participants to a series of aerosols including environmental tobacco smoke, cooking oil fumes, and wood smoke (peak concentration target 350 µg/m³) in 20-minute sessions. The authors reported no effect of the aerosols on mean systolic pressure compared to the water vapor sham control, but note a 4 mmHg increase across the exposure sequence for all aerosols. However, observed changes in spectral powers of heart rate and blood pressure suggested that the aerosols increased peripheral vasomotion and baroreflex activity. Results of these studies provide evidence that wood smoke exposures can impact various markers of cardiovascular function that relate to the pathways described above, in an acute time frame.

Conversely, Hunter et al. (2014) found no changes in systolic or diastolic pressure among firefighters during a one-hour controlled exposure to wood smoke at target concentration of 1,000 μ g/m³ PM_{2.5}, or at follow-up six and 24 hours after exposure ended. Similarly, no changes in augmentation index or pulse wave velocity (other markers of hemodynamic changes) were observed, leading authors to suggest that other air pollutants or non-pollutant stressors (e.g., physical exertion) might be responsible for the cardiovascular events seen in wildland firefighters, as opposed to a PM-induced response.

Lung Function

Few studies have investigated lung function following controlled wood smoke exposures. Several controlled exposure studies have shown changes in airway inflammatory markers, increased exhaled NO, and clinical irritation symptoms following wood smoke exposures, indicating that wood smoke air pollution can elicit acute respiratory tract inflammation and irritation (Barregard et al. 2008; Riddervold et al. 2012; Stockfelt et al. 2012). However, other controlled exposure studies did not observe these effects (Ghio et al. 2012; Sehlstedt et al. 2010). If wood

smoke causes acute inflammation and irritation, then reductions in lung function would be expected following short-term wood smoke exposures.

Three studies were identified that measured lung function by spirometry following controlled wood smoke exposure. Riddervold et al. (2012) studied 20 volunteers exposed to filtered air (less than 20 µg/m³ PM), low wood smoke treatment (approximately 200 µg/m³ PM), and high wood smoke treatment (approximately 400 µg/m³ PM) for three hours in a crossover design. The authors found no differences in the change in FVC, FEV₁, or peak expiratory flow (PEF) from pre-exposure to 3.5 or six hours after exposure between the filtered air and the PM treatments. Sehlstedt et al. (2010) similarly exposed 19 volunteers to three-hour treatments of filtered air and diluted wood smoke at concentrations of 5 to 15 ppm CO, 0.2 to 0.4 ppm NO_x, and 180 to 300 µg/m³ PM_{2.5}. No changes in FVC or FEV₁ were observed immediately after, four hours after, or 24 hours after exposure. Ghio et al. (2012) found no changes in FVC, FEV₁, or PEF among ten volunteers following two-hour exposures to filtered air or wood smoke particles (approximately 500 µg/m³ concentrations; health measurements conducted immediately after and 20 hours after exposure). These studies may have been limited by a low number of participants.

Other Air Pollution Controlled Exposure Studies

Blood Pressure

Several controlled exposure studies of ambient air pollution sources have demonstrated that blood pressure can change immediately (e.g., within minutes of an exposure to concentrated ambient particles (CAP)), however effects may subside within minutes to a few hours after exposure ends. Urch et al. (2005) saw rapid increases in diastolic pressure, but not systolic or strangely mean pressure, during exposures to combined CAP (range 102 to 214 μ g/m³ PM_{2.5}) and ozone (115 to 128 ppb), reaching a linear change of 6 mmHg over two hours. Authors found strong associations with blood pressure changes and the organic carbon content of the exposure. Similarly, a study by Brook et al. (2009) wherein participants were exposed to different combinations of CAP (150 μ g/m³ PM_{2.5}) and ozone (120 ppb) found that diastolic blood pressure

increased during two hour exposures, reaching a change of 2.9 mmHg for just CAP versus 2.5 to 4.0 mmHg for CAP and ozone combined. Systolic pressure also increased by 2.3 to 3.5 mmHg, though non-significantly, with CAP exposure. No significant differences in blood pressure were seen immediately after the exposure ended or 24-hours post-exposure. An earlier study by Brook et al. (2002) found no effect on blood pressure ten minutes after a similar two hour combined exposure to CAP (150 μ g/m³ PM_{2.5}) and ozone (120 ppb); blood pressure was not measured during the exposure window. Byrd et al. (2016) found that both systolic and diastolic blood pressure were approximately 1.9 mmHg higher during two-hour coarse PM (2.5 to 10 μ m diameter) exposures than during filtered air control exposures; no differences were observed two hours after the exposure had ended. However, Cosselman et al. (2012) found that systolic pressure (but not diastolic) increased during two-hour diesel exhaust exposures (200 μ g/m³ PM_{2.5}), with increases sustained (though reducing in magnitude) through 24 hours after exposure ended.

Lung Function

Many studies have been conducted that observe airway responses (e.g., changes airway hyperresponsiveness and markers of airway inflammation) following controlled exposure to diesel exhaust and concentrated ambient particles (e.g., Nordenhall et al. 2001; Samet et al. 2007; Stenfors et al. 2004; Zhang et al. 2016). Yet, several studies on spirometry-measured lung function report null results. For example, Samet et al. (2009) exposed 19 young, healthy volunteers to filtered air and concentrated ultrafine ambient particles at a mean of 121,000 particles/cm³ (mass averaged $50 \pm 20 \,\mu$ g/m³) for two hours. No statistically significant changes in FVC, FEV₁, or diffusion capacity were observed for the particle exposure compared to the control immediately after or 19 hours after exposure, though small (less than 2% per 100,000 particles/cm³) non-significant decreases were observed. Gong et al. (Gong et al. 2003a; Gong et al. 2003b) report no changes in spirometry among 12 healthy and 12 asthmatic adults who underwent two-hour exposures to concentrated ambient particles at 200 μ g/m³ PM_{2.5}.

Other studies have observed spirometry changes. For example, Zhang et al. (2016) found that two-hour exposure to diesel exhaust, followed by administration of a methacholine allergen, resulted in reductions in FEV₁ compared to filtered air control exposures two hours later only among participants with glutathione-S-transferase T1 genotypes. The authors conclude that the gene-environment interaction suggests certain subpopulations are more susceptible to respiratory effects of diesel exhaust air pollution exposures. Hazucha et al. (2013) exposed older cigarette smokers (35 to 74 years) to concentrated ambient fine particles at approximately 100 µg/m³ or filtered air for two hours; at 22 hours post exposure, FEV₁ was significantly reduced following the particle exposure compared to the clean air exposure and immediately and three hours post-exposure results were suggestive of a decrease in FVC and FEV₁, though not significantly so.

CHAPTER 3: AN EXPERT SURVEY ON THE MATERIAL TYPES USED TO START COOKSTOVES

Summary

Household air pollution generated using solid-fuel cookstoves is a leading risk factor for morbidity and mortality worldwide. Many studies have quantified cookstove emissions with respect to the main fuels used (e.g., wood, charcoal and other biomass fuels). Anecdotal evidence suggests a variety of fuels are used to ignite cookstoves, however quantitative information on startup fuel types is lacking. Emissions from these startup materials will contribute to household air pollution. As such, understanding startup practices and the choices of startup materials is integral to understanding the full burden of household air pollution from cookstoves on health and the environment. We conducted an expert elicitation style survey to gather information about startup practices worldwide. Twenty-three respondents from academic and private sectors responded to a survey instrument on cookstove startup, providing information that covered 48 geographic locations across 22 countries. Responses to the open-ended questions were analyzed to quantify how startup materials vary according to factors such as location, seasonality, and the main cooking fuel/stove type. A wide variety of materials were reportedly used to ignite cookstoves, including many non-biomass materials that may have health-relevant combustion byproducts. Paper, plastic, agricultural wastes, waxes and other petroleum fuels (e.g., kerosene), and rubber-like materials (e.g., tires, footwear) were the most frequently indicated startup materials. Additional materials mentioned included fabrics, plastic packaging, soda bottles, snack food wrappers, and trash. Results from this survey can be used to direct future research on the impacts of startup materials on health and the environment, such as justifying the choice of materials to test in the laboratory.

Introduction

Approximately 2.8 billion people – around 40% of the world's population – use solid fuel as their primary source of domestic energy (Bonjour et al. 2013). Solid fuels are often burned in inefficient and poorly ventilated cookstove systems (Bonjour et al. 2013). Cookstove emissions contribute to indoor and outdoor air pollution that impacts human health and the environment (Gakidou et al. 2017; IPCC 2014). However, the estimated health and environmental impacts of cookstove emissions are highly uncertain. Thus, work is needed to better understand the practices that lead to cookstove emissions and to quantify the complex mixture of emissions that results.

One potentially important factor for understanding cookstoves emissions and subsequent health impacts is the stove ignition or startup process. During cookstove startup, a fast-burning fuel is often used to ignite the main fuel. Startup could be particularly important for human health, as the cookstove user is often very close to the stove during the startup phase, which might result in high personal exposures. Several laboratory studies have demonstrated that the startup phase contributes substantially to overall emissions, though the contribution can depend on the startup fuel type, method of igniting, and main fuel (e.g. Arora et al. 2014; Bhattacharya et al. 2002; Carter et al. 2014; Lask and Gadgil 2017; Wathore et al. 2017). The pollutant profiles from burning startup materials may be different from that of the main cooking fuels as the composition of these fuels is often different; yet, studies have not isolated the startup fuel emissions from the main cooking fuel emissions. Further, laboratory cookstove studies that attempt to quantify emissions have generally been limited to the use of cleanly-milled wooden shims or kerosene for startup. Yet, anecdotal evidence suggests that a variety of fuels such as plastic bags, agricultural and consumer product waste, and rubber tires are often used to ignite stoves. Although the primary solid-fuel types used worldwide have been documented (OECD/IEA 2006), information on the types and prevalence of startup materials is currently lacking. It is difficult to understand how startup practices contribute to overall cookstove emissions, air pollution, and health - and

therefore how modifications to startup practices could reduce pollution and improve health – without first knowing what types of materials are used for cookstove startup. To systematically gather this information first-hand would be a major undertaking because cookstove use occurs across diverse, often rural regions in predominantly low and middle-income countries.

We designed and implemented a survey to collect information about fuel types used during startup of cookstoves globally. Our survey incorporated methods for respondent selection and question style used in expert elicitation. Expert elicitation is a structured method for gathering and synthesizing knowledge in the form of technical, yet subjective judgments from subject-matter experts on questions that have qualitative uncertainty (Cooke 1991; US EPA-SAB 2010). Expert elicitation is a useful starting point for gaining insight into areas with limited existing research (Knol et al. 2010). We targeted experts who work in cookstove development, dissemination, and evaluation, and had spent time working with cookstove users in various communities. We targeted experts rather than cookstove end-users because it allowed us to gather reputable information on a wide range of locations, communities, and cooking practices in quick and more cost- and resource-effective ways. We asked respondents a series of open-response questions to gather information on the types of cookstove startup materials used in areas of the world where they had expertise.

Methods

Survey Development and Content

The online survey was developed using an iterative process of question design, review, and testing. Five individuals currently involved in cookstove research beta-tested the survey to provide feedback on content and clarity; these individuals were not included as respondents in the final survey. The survey was determined to be exempt from the requirements of the human subjects research protections by the Colorado State University Institutional Review Board.

Respondents were asked to identify up to three geographic locations of expertise and provide information about the cookstove technologies they have encountered in those regions.

The number of locations was limited to three to control the length of the survey; respondents who had experience in more than three locations were instructed to choose the three where they felt they had the most experience. Additionally, "location" was loosely defined and the response was open-ended (i.e., respondents filled in a blank space rather than selecting from pre-determined options) to allow respondents to be as specific or broad as they felt was justified based on similarities in startup practices across the location. For example, respondents could choose to define a location as specifically as a single community or as broadly as an entire country or world region. For each location, respondents identified the following: the two most commonly used stove types in the area, the main cooking fuel types associated with these stoves, and the two materials most commonly used to start these specific stoves (limited to two for survey length purposes). Additionally, participants were asked to list all other materials they were aware are used as startup materials in the location, whether commonly or uncommonly used and regardless of the stove type or main cooking fuel type with which it is used. Responses for stove types and fuel types (main and startup) were open-ended. This approach allowed the respondent to describe the stoves and fuels using their own terms, rather than limiting them to a predetermined list provided by the researchers. Additionally, respondents were asked whether the use of different startup materials in this location varied depending on factors such as season, availability of fuel types, or other considerations (semi-structured response). Finally, respondents were asked if they had ever collected data on the use of startup materials in the location, and if so, whether they looked at that data to inform their responses or not (respondents were not instructed prior to the survey to look at data or not). After providing information specific to up to three locations, respondents were asked, "in all locations that you have lived or worked, what is the most unique or surprising material you've seen used as a startup fuel?" Lastly, respondents answered questions about their years of work experience, their work sector, and their current workplace affiliation. The survey was anonymous as respondents did not provide their name or contact information. The survey is provided in Appendix A.

<u>Respondent Identification</u>

While no guidelines currently exist, current recommendations suggest that more than six experts are needed for robust results in expert elicitation style surveys; after inclusion of about 12 experts, the benefit of each additional expert opinion begins to taper as each new respondent does little to change the overarching trends represented by previous respondents (Knol et al. 2010). We therefore aimed to include at least ten respondents in our survey. We identified potential respondents based on the authors' knowledge of their current involvement and work in cookstove research, development, and dissemination. The final list of targeted respondents was determined with input from professional colleagues who have extensive experience in the cookstoves field. Several targeted respondents were identified through their involvement with various cookstove-related committees (e.g., International Standard Organization Technical Committee 285: Clean Cookstoves and Clean Cooking Solutions), working groups (e.g., Global Alliance for Clean Cookstoves Working Group on Technology and Fuels), and meetings (e.g., annual Engineers in Technical and Humanitarian Opportunities of Service Cookstove Conference). We sought respondents who represented a geographical spread of expertise as well as a range of experience including policy, stove design/testing, exposure/emissions assessment, and health. We also targeted representation from individuals with location-specific knowledge (i.e., individuals with field projects in specific locations), and general knowledge (i.e., individuals who perform non-location specific laboratory work and/or are involved in global policy). We avoided contacting multiple respondents who worked for the same organization or on the same research projects; however, we allowed respondents to share the recruitment email and survey web-link with colleagues. Individuals were contacted between December 15, 2015 and January 15, 2016. The survey remained open for responses until February 15, 2016.

<u>Analysis</u>

Two individuals (KF, EW) independently coded the response data. The primary coder (KF) developed a coding structure that allowed responses to be categorized and grouped; the

secondary coder (EW) applied this structure to the data but also provided suggestions for improvements to the structure in an iterative process. Coding was compared and discrepancies were reconciled by the primary coder.

The primary goal of coding responses to the open-ended survey questions was to identify each time a startup material was mentioned, paired with details about the use of the startup material such as location, stove type, and main cooking fuel type (i.e., the fuel that is ignited with the startup material and used to sustain the cooking event). Each identification of a startup material was counted as a single startup mention. Each respondent had the ability to identify multiple materials used within each location for which they responded as they were asked for the most common and second most common materials associated with different stove types and fuel types as well as "other" materials. Material mentions were categorized and grouped as part of the coding process. When a material description crossed multiple grouping categories, it was counted as both categories (e.g., "wood chips soaked in kerosene" was counted as a mention of both kerosene and kindling material). We tallied all material mentions across all responses and separately tallied responses according to whether the material was mentioned as a "most commonly used," "second most commonly used," "other," or "most unique" startup material.

We categorized results according to location, stove type, and main fuel type to compare differences in the materials used across these variables. Locations were coded to the country level as well as to a world region. We defined five regions (Africa, Mexico/Central America, East/South East Asia, South Asia, and South America) that encompassed all reported locations with a reasonable spread of responses per region (i.e., each defined region had at least three responses and no region contained more than approximately one third of the total responses) using the World Health Organization member state regions (World Health Organization 2017) as a rough guide. Stove types were grouped and coded as open/three stone fires, traditional stoves (including ceramic, clay, mud, or non-insulated metal), improved stoves (insulated metal or ceramic designs, such as jikokoa, rocket elbow, and gyapa style stoves), built-in stoves (larger

stoves with chimneys, including griddle, plancha, chulha, and patsari styles), improved built-in stoves (including "ONIL", "lorena", or other built-in styles where respondent specified it was an improved rather than traditional type), or advanced stoves (any that specifically used an advanced fuel type such as kerosene, electric, or liquid petroleum gas [LPG] as the main fuel source). Main fuel types were coded as wood, charcoal, coal, dung and agricultural/crop residues, LPG, or kerosene. If a main fuel description crossed multiple grouping categories, it was counted as both categories (e.g., "wood and charcoal" was counted as both wood and charcoal). We calculated percentages across these various categories as the number of material mentions for a given material compared to the total number of material mentions in that category.

For questions about the respondents' expertise/focal areas, we coded respondents into one of three cookstove sectors according to their workplace affiliation and job titles: academia (university researchers), policy (non-academic institutions conducting advocacy and/or energy policy work), and other private sector (e.g., non-academic groups conducting stove testing, stove development/design, or other traditional-style stove research). Participants provided an open-ended response describing their involvement in the cookstove field; these responses were used to group participants into the following focal areas: emissions testing (conducting tests of stove performance with regards to pollution emissions), health (any mention of working with health impacts of cookstoves), design (directly designs cookstoves or works with stove manufacturers), adoption/social aspects (involved with the social aspects of cookstove use, mentions "interventions" generally, or describes work on clean cookstove adoption or cooking behaviors), and field studies (any mention of conducting in-country, hands-on work with stove users). Respondents could be categorized as working under multiple focal areas.

The survey was created and hosted using Google Forms (Google, 2016). Qualitative coding was conducted using Microsoft Excel (2013). Analysis of coded responses was conducted using R (version 3.3.1, The R Foundation for Statistical Computing).

Results

Respondent Characteristics, Experience Levels, and Locations

We directly contacted 41 individuals to participate in the survey. Several contacted individuals forwarded the recruitment message to colleagues; one individual posted the message to a cookstoves email listserv. As such, it is difficult to know how many total individuals saw the call to participate, but we estimated it to be 45 to 60. We received completed survey responses from 23 individuals. Twelve of the respondents (52%) were employed in private research/design, seven (30%) were in academia and the remaining four (~18%) were involved in policy. Experience in cookstoves work ranged from 1 to 30 years (mean: 9 [standard deviation 7], median: 8; see Table 3.1). Although responses were anonymous and workplace affiliation was not included, based on the combination of identified locations and employment sector/focal area, we do not believe there was considerable overlap of respondents from the same employers or who worked on the same projects. Respondents provided information on 49 locations in 22 countries (see Table 3.2). Most respondents chose to identify a country or specific region within a country. Aggregated by region, responses were not evenly distributed. At the country level, India was the most highly cited with 14 responses, other countries were cited between one and four times.

The majority of respondents reported that they had not formally collected data on the startup materials used in the location for which they were responding (29 of the 49 locations; 59%) and based their responses on memory. Of those who had collected data (35%), two-thirds stated that they looked at that data to inform their responses to the survey (11 of the 49 locations [22%]), while the remaining third reported that they had collected data but did not look at the data while responding to the survey (6 of the 49 locations; additionally, 6% did not respond to this question [three of the 49 locations]).

Table 3.1 Survey Respondent Expertise and Characteristics. Respondents were categorized into sectors according to their workplace affiliation and job titles. Respondents were categorized into focal areas based on their involvement in the cookstove field. Respondents could be categorized as working under multiple focal areas.

	Total	By Focal Area ^a								
	Sector	Stove Design	Emissions	Health	Adoption/ Social	Field Studies				
All Sectors										
Number of Respondents	23	9	9	7	12	10				
Years of Experience										
[average±standard	9.0±7.1	14.8±7.2	11.1±8.8	5.4±3.6	8.0±8.3	5.3±3.1				
deviation	(1, 30)	(8, 30)	(2, 30)	(1, 10)	(1, 30)	(1, 9)				
(min, max)]										
Academics										
Number of Respondents	7	1	1	4	4	6				
Years of Experience [average±standard deviation (min, max)]	5.9±3.3 (1, 10)	8	8	6.0±3.9 (1, 10)	4.0±3.1 (1, 8)	5.2±3.1 (1, 8)				
Policy										
Number of Respondents	4	1	0	2	2	2				
Years of Experience [average±standard deviation (min, max)]	6.8±5.85 (2, 14)	14	NA	5.5±4.0 (2, 9)	8.0±8.4 (2, 14)	5.5±4.9 (2, 9)				
Private Research/Stove Design										
Number of Respondents	12	7	8	1	6	2				
Years of Experience [average±standard deviation (min, max)]	11.5±8.4 (2, 30)	15.9±7.75 (8, 30)	11.5±9.3 (2, 30)	3	10.7±10.5 (3, 30)	5.5±3.5 (3, 8)				

^aRespondents were allowed to identify multiple focal areas.

Table 3.2 Locations Represented by Survey Responses. Locations were coded to the country level as well as to a world region. We defined five regions influenced by the World Health Organization Regions, to encompass all reported locations with a reasonable spread of responses per region.

Pagion/Country	Number of	% of	% of population using solid		
Region/Country	Responses	Responses	fuels, 2013ª		
South Asia	16	33	63 ^b		
Afghanistan	1	2	80		
India	14	29	64		
Nepal	1	2	80		
Africa	13	27	79		
Benin	1	2	94		
Burkina Faso	1	2	95		
Ethiopia	1	2	>95		
Ghana	1	2	83		
Kenya	4	8	84		
Madagascar	1	2	>95		
Malawi	1	2	>95		
Mali	1	2	>95		
Nigeria	1	2	75		
Zambia	1	2	82		
Mexico/Central	11	22	Qc		
America		22	3		
El Salvador	1	2	19		
Guatemala	2	4	65		
Honduras	4	8	50		
Mexico	2	4	15		
Nicaragua	1	2	53		
Unspecified	1	2			
East/Southeast Asia	4	8	40 ^d		
Cambodia	2	4	88		
China	2	4	45		
South America	3	6	9°		
Bolivia	1	2	23		
Peru	2	4	34		
Unspecified	2	4			
Urban	1	2			
Rural	1	2			

^aSource: World Health Organization, Global Health Observatory Data Repository (<u>http://apps.who.int/gho/data/node.main.135?lang=en</u>).

^bEstimate for WHO South-East Asia region only (includes India and Nepal, does not include Afghanistan)

[°]Estimate for WHO Americas region (includes North America, Central America/Latin America, and South America)

^dEstimate for WHO Western Pacific Region (includes China and Cambodia)

Main Stove and Fuel Types

The stove types identified as the primary/most common and secondary/second-most common included a roughly even spread of three stone fires (25 responses [27% of the 92 total responses]), traditional style rudimentary stoves (ceramic or clay/mud pots/rings or non-insulated metal; 21 responses [23%]), improved styles (insulated metal and ceramic stoves such as rocket elbow or gyapa stoves; 18 responses [20%]), and built-in biomass stoves (traditional and improved combined; 21 responses [23%]), advanced-fuel burning stoves like kerosene and LPG were less common (seven responses [8%]). Three stone fires were relatively common across all regions (25 to 50% of reported stove types per region) except East/Southeast Asia, where no respondent identified use of a three stone fire as the most common or second most common stove type. Similarly, traditional-style, non-built-in stoves were relatively common (17 to 26%) across all regions except Mexico/Central America (only two responses, representing 9% of the mentioned stoves for that region), though notably this region had high reported use of traditional built-in style stoves (six responses [27%]). Built-in stoves, including regular and improved designs, were more frequently reported in South Asia (eight responses [29% of total responses for that region]), Mexico/Central America (10 responses [45%]), and East/Southeast Asia (two responses [25%]) than in Africa (only one response [4%]) and South America (no responses).

Across all stove types, wood was the most frequently named main fuel source (63 responses [68% of the 92 total responses]). Wood was identified as a main fuel source in more than 50% of the responses (54 to 91%) in all regions except East/Southeast Asia, where total responses were low but wood was still highly represented (three responses out of only eight responses for this region). Charcoal was the second-most frequently identified main fuel source (15 responses [16%]); though this was driven primarily by a high number of charcoal responses in Africa (10 responses out of 24 total responses for Africa). Other noted main fuel sources were LPG (six responses [7%]), agricultural/crop residues and dung (three responses [3%]), coal

(one response [1%]), kerosene (one responses [1%]), and a combination of multiple fuels like wood and charcoal together (two responses [3%]).

Types of Startup Materials Used Globally

Named startup materials were first tallied across all responses irrespective of stove type, main cooking fuel, or question posed to understand the scope of startup materials used world-wide. A total of 292 startup material mentions occurred across all survey responses; the groups and total count of responses per group are provided in Table 3.3. More than 40 unique materials were mentioned. These materials were categorized into eight main groups: accelerants, plastics, paper, kindling biomass, agricultural wastes, high-resin wood, rubber, and fabric. Types of materials that could not be categorized into one of the main eight categories were grouped as "other"; only 12 mentions fell into this category. The category included animal dung (three responses, associated with India, Guatemala, and general Central America), butter/"ghee" (one response, India), "insulation" (two responses from same respondent, Guatemala), household trash (two responses, China and Honduras), and an unspecified other wood type (one response, Honduras). Most materials that were categorized as "other" were mentioned by respondents as unusual or uncommonly used.

Accelerants, paper, and plastic together represented nearly half (49%) of the materials mentioned across all respondents. Common responses for accelerant materials included kerosene (37 of the 51 total mentions), but also waxes/paraffin and oils; these materials are reportedly used alone (poured over the main fuel source in a combustion chamber and then lit with a match to help in ignition of the main fuel source) or sometimes in conjunction with other materials. For example, according to one respondent, sawdust soaked in kerosene is a "traditional starter available on the market" in Mali, whereas in Cambodia, the traditional market-available starter is a bundle of leaves with resin (Respondent 4). Newspaper was the only paper type

Table 3.3 Materials Used for Startup: Categories and Total Count of Responses Across All Locations. Each identification of a startup material in response to any question within the survey was counted as a single "mention." Questions asked about the most commonly and second most commonly used materials associated with the two most common stove types and fuel types as well as "other" materials and "unique" materials within a given location; respondents could provide information relevant to up to three locations. When a material description was compound and crossed multiple grouping categories, it was counted as both categories.

Category	Definition	Any mention	Most commonly used	2 nd most commonly used	Other	Most unique/ surprising
Total responses	All startup materials, including a response of "None"	292	104	97	73	18
Accelerant	Kerosene, paraffin/wax, or burning charcoal material transferred from a previously lit stove	51	18	19	12	2
Plastic	Plastic bags, bottles, packaging material, polythene, and unspecified types	49	15	18	10	6
Paper	Including newspaper	42	11	21	10	0
Kindling	Small branches, leaves, twigs, sawdust, grass clippings, or other biomass based "tinder"; sometimes combined with resin or kerosene <i>(if so, "accelerant" also selected)</i>	40	13	11	12	4
Agricultural waste	Rice straw, dry maize/corn stalks and husks, crop/garden residues such as weeds and grasses	37	16	7	13	1
High resin wood	Wood types that burn easily, mainly <i>ocote</i> or other pinewoods	25	14	7	4	0
Rubber	Bike tires/tubes, flip flops, shoe soles, and other assorted rubber scraps	13	6	0	4	3
Other	Mostly animal dung; some mention of food scraps, general rubbish, insulation materials, and other infrequently mentioned materials	12	2	2	2	1
Fabric	Strips of fabric, including weaved fabrics	7	3	2	6	1
None	Respondent specifically stated that no starter is used	16	6	10	0	0

identified specifically. Similarly, plastic bags were the most common plastic type identified. Many individuals noted plastic materials as the most unique or surprising material type they have seen used as a fire starter, such as "broken basins [and] jerrycans" (Respondent 19, seen in high poverty rural area), "Coca Cola bottles and fried chip envelopes" (Respondent 8, seen in "the most poor villages" in rural Mexico), and "expanded polystyrene" packaging from electronic items (Respondent 9, seen in India).

Biomass-based materials (kindling, agricultural waste, and high-resin wood such as *ocote* or other pine species) represented about one third (36%) of the total startup material mentions. Kindling types were highly varied, including materials like "wood sticks with dried leaves attached" (Respondent 6, China), "small pieces firewood- peeled off the [main] firewood to be used" (Respondent 16, Kenya), "twigs & other small biomass" (Respondent 11, Guatemala), and "dry grass", "dry leaves", or "straw" (Respondent 16, Kenya, and Respondent 18, Malawi). Agricultural residues or wastes also varied by location, often indicative of the local crops – for example, corn cobs/maize stalks in Honduras, Guatemala, Kenya, and southern China, and rice straw in India and Nepal. Other unique agricultural waste mentioned included "bamboo, palm fronds, coconut husks" in Central America (Respondent 15), mustard stalks in India (Respondent 12), and "palm oil press residues" in Togo and Ghana (Respondent 19).

Rubber materials included "assorted [types] from insulation to tire tubes to flip flops" (Respondent 1, Cambodia), though the most common responses included tire tubes/scraps and flip flops. Rubber materials were often noted as a low use material (i.e., not the most commonly used material, but still a notable material) or as a material that the respondent felt was unique or surprising (e.g., "rubber nipples soaked in gasoline" seen in Tibet by Respondent 2, or an "old tennis shoe" seen in rural Guatemala by Respondent 15).

Variations by Region

The breakdown of responses according to region is shown in Figure 3.1. For most regions, responses indicated that a wide range of materials are used for stove startup. Responses from

Africa and South Asia covered all eight of the categories used to group materials; East/Southeast Asia had responses for all categories except paper, and Mexico/Central America had responses for all categories except rubber and fabric. Accelerants and kindling were the most frequently mentioned materials for Africa, with 18 mentions each out of 82 for this region, followed by plastics, which were mentioned 17 times and paper, which was mentioned 11 times. High-resin woods, fabric, rubber, and agricultural waste were all noted infrequently. Accelerants were also the most frequently mentioned material in South Asia (20 out of 79), though paper received a similar percentage of responses (19 out of 79). Agricultural waste was more frequently mentioned in South Asia than in other regions, with 17 out of 79 mentions. Resinous pinewood species were the most frequently noted startup material in Central America with 17 out of 72 mentions; this category was rarely mentioned across all other regions.

Variation in Startup Material Choice across Different Stove Types and Main Fuels

Reported use of startup materials varied with the main fuel used as well as by stove type, as seen in, Figure 3.2. Wood was the most frequently reported main fuel type (138 total responses out of 201 total); the most frequently mentioned startup materials used with wood-burning stoves were paper (28 responses) and plastic (23 responses), but agricultural waste, kindling, accelerant, and high-resin woods like pine and *ocote* were also frequently mentioned (20, 19, 18, and 17 responses, respectively). Rubber and fabric were rarely mentioned for igniting wood (three and two responses, respectively). Responses from other main fuel types were considerably fewer (only 37 responses relevant to charcoal stoves and nine for liquid fuel stoves); however, charcoal appears to follow a similar trend as wood, with more frequently reported use of accelerants, plastic, paper, and kindling materials than rubber, fabric, or other types of materials. Responses for liquid fuels (LPG, kerosene) indicate that no startup material is used with these stoves, or if one is, it is a small amount of the same material as the main fuel (e.g., kerosene). Similarly, reported use of startup materials varied with stove type (see Figure 3.3). Paper, plastic, kindling,



Figure 3.1 Materials Used for Startup: Responses by Region. Bar charts show the number of responses for each startup material category. Each unique mention of a startup material (associated with a primary or secondary stove, primary or secondary main fuel, or generally mentioned as an additional material) was included. Material categories are defined according to Table 3.3. Bars are colored by material type and arranged in the same left-to-right order for each plot. "Ocote" group includes all high-resin woods; "agwaste" is agricultural waste. The left panel shows response counts for all regions combined; right panel contains separate charts for the startup materials mentioned for each region. See Table 3.2 for list of countries represented within each region.



Figure 3.2 Materials Used for Startup: Responses by Main Fuel Type. Bar charts show the number of responses for each startup material category. Material categories are defined according to Table 3.3. Bars are colored by material type and arranged in the same left-to-right order for each plot. "Ocote" group includes all high-resin woods; "agwaste" is agricultural waste. Separate charts are provided for each main fuel source. Individual plots for coal and "wood and charcoal" combined were excluded because of the limited number of responses (1 and 3, respectively).

agricultural waste, and accelerant were frequently reported materials across three stone fires, traditional designs, and improved designs.

Other Reasons for Variation in Startup Material Choice

Nearly half of the respondents (22/46, 48%) indicated that choice of startup material varied with season, availability of main fuel, or other factors. Variation in startup material choice was reported more frequently in South America and South Asia (67% and 62%, respectively, answered "yes" to the question of whether startup material choice varied). In Africa, East/Southeast Asia, and Mexico/Central America the majority of respondents indicated no variation in startup choice (62%, 75%, and 64% respectively). Of those who did say that the startup materials varied, reasons provided for that variation included availability of different materials, the cost of materials and/or affluence of families to afford certain fuel types, level of education of the cooks, seasonality (e.g., dry vs. rainy seasons), and differing practices in urban versus rural locations.

Availability of certain materials was the most frequently cited reason that startup choice might vary (12 mentions); seasonality was the second most frequently cited reason (7 mentions). Respondents generally indicated that individuals will burn "whatever burns quickly and is available" (Respondent 4, Mali and Cambodia). Concepts of availability also intersected with location and season. For example, one respondent stated that in rural Honduras, "there are different wood species across the study villages based on location near certain forests, so different people will use whatever is the most easily/cheaply available" (Respondent 5). The agricultural seasons were a major cited influence on availability of different startup materials. "Crop residues are seasonal" (Respondent 12, India) and materials like maize stalks, corn husks, and corn cobs are "plentiful after [the] harvest" (Respondent 16, Kenya). Similar to preferentially burning agricultural wastes when available, individuals might preferentially burn household waste materials like plastic and paper when these materials are available as this doubles as a waste



Figure 3.3 Materials Used for Startup: Responses by Stove Type. Bar charts show the number of responses for each startup material category. Material categories are defined according to Table 3.3. Bars are colored by material type and arranged in the same left-to-right order for each plot. "Ocote" group includes all high-resin woods; "agwaste" is agricultural waste. Separate plots are provided for the startup materials mentioned across each stove type.

management solution (Respondent 9, India). Several respondents also noted that "wet or dry season makes a difference" (Respondent 14, Bolivia), because "the moisture content present in the [main] fuel" impacts how an individual starts their stove/fire (Respondent 23, India), and a "lack of dry materials" during the rainy season may drive individuals to purchase kerosene or other commercially-available starter materials instead (Respondent 18, Madagascar).

Two respondents indicated that education and economic situation explained choices in startup materials. One respondent stated that "Justa [improved] stove owners receive training to not burn trash in their new stove, so they might be less likely to startup their fires with things like newspaper, plastic, etc." (Respondent 5, Honduras). Similarly, another respondent stated that startup material choice "relies on economic and educational level of the ladies," as "in Villages where government programs of health care are implemented, they are aware of health risks associated with the use of plastic or other smoky [materials]" (Respondent 8, Mexico). This same respondent also noted that startup material choice "depends on family size and...life stage," explaining that "there are a lot of rural villages in Mexico where the men are working in the US and the lady is not able to go out ... far [from] her house and has small children", so these women would "use whatever they have available in their houses" (Respondent 8, Mexico).

Discussion

Nearly 40% of the world's population currently cooks over open fires or rudimentary cookstoves, which is a large source of air pollution with local, regional, and global implications. Documenting the range of cooking practices that occur around the world is important in order to design behavioral change interventions that reduce emissions. Almost no published documentation exists regarding stove startup practices. Indeed, the majority of the cookstove experts interviewed for this research reported that they had never specifically collected data about cookstove startup practices. The startup process could be important for overall cookstove emissions and air pollution exposures; further, the types of materials burned during startup may be an important determinant of emissions. Understanding what materials are commonly used

during startup is a necessary first step towards understanding the impact of the startup process on overall emissions and exposures and subsequently how changes to this process (such as encouraging the use of certain startup materials over others) might result in decreased emissions and/or lower exposures. This expert elicitation was an efficient and effective first step to gather information on startup materials relevant to a range of locations.

Our survey results indicate that a variety of materials are used to ignite cookstoves, but there are also commonalities across these materials. Three of the material groups identified - accelerants, paper, and plastic - together represented nearly half of the mentioned materials. Biomass-based materials (kindling, agricultural waste, and high resin wood) represented about one third of the responses. Understanding startup material choice is an important step towards understanding the contribution of startup materials to overall cookstove emissions and their subsequent health and environmental impacts.

The startup materials reportedly used with improved stove types have a similar pattern as those used with three stone fires and traditional stove types. A wide variety of materials are used across all stove types, with similar frequency between several major groups (accelerants, plastics, paper, kindling, and agricultural waste). More advanced stoves burning liquid fuels almost exclusively do not require a startup material, and if one is used, it is a similar advanced liquid fuel type (accelerants). Although it is difficult to draw conclusions on the comparison of startup practices across different fuel types due to the limited number of responses for some fuels, responses similarly indicated that a difference in startup practices is seen only when comparing kerosene or LPG fuels to other types of biomass fuels (e.g., wood, charcoal).

Our survey respondents may have overrepresented users of improved or advanced stoves and therefore over-represented the associated startup materials. The Global Alliance for Clean Cookstoves estimates that over 60 million households have transitioned from open fires and traditional-style stoves to more efficient improved or advanced stoves (Global Alliance for Clean Cookstoves 2015). This statistic indicates that a considerable number of improved or advanced

stoves are in use globally; however, it also implies that the vast majority of the 2.8 billion cookstove users worldwide are still using rudimentary or traditional style stoves. In our survey, about 30% of responses were related to improved or advanced stoves, which is likely greater than the percentage of cookstove users who use improved/advanced stoves compared to traditional or open fires. This likely occurred because we sought respondents who had expertise in the cookstove sector and therefore work with communities on research and projects aimed at understanding clean cookstove adoption. However, reported startup practices were similar across three stone fires/traditional stoves and improved designs, so this overrepresentation is unlikely to be a limitation of our conclusions.

Our survey provided a reasonable representation of the main fuel types used globally, which gives us confidence that the global startup material breakdown is representative. Our survey responses indicated 50 to 90 percent wood use depending on the region (average 68%) and 8% use of LPG and kerosene. Charcoal was a frequently reported main fuel in Africa but not elsewhere. These results align with estimates from The World Bank that 60 to 80 percent of households in developing countries use wood as their primary energy source and charcoal use is limited except in Africa (Malla and Timilsina 2014). Advanced fuel use is estimated to be higher than indicated by our survey, around 20% of energy consumed in developing countries (Malla and Timilsina 2014). However, this estimate includes use for heating, lighting, and other energy requirements, so is an overestimate of residential cookstove consumption.

Total responses for our survey were dominated by input relevant to Africa and South Asia (primarily India). Although these regions represent a large percentage of global cookstove users, more representation for East/Southeast Asia and South America is needed to confidently draw conclusions that the relative representation of different materials by survey respondents matches true global startup material use patterns. It should be noted that our survey results are not weighted to the population of cookstove users within a given country. For example, approximately 64% of the population of India (roughly 832 million of the 1.3 billion people) use solid fuel

cookstoves, whereas only 9% of the Americas region (approximately 90 million of the one billion people) uses solid fuels for cooking (see Table 3.2). Yet, our survey contained 14 responses from India and comparatively 11 responses relative to Mexico and Central America. Furthermore, region-to-region comparisons may be problematic using our results, due to the limited number of responses in some regions compared to others. In regions with fewer responses (for example, South America), the patterns of responses may not be representative but rather indicative of limited and specific experiences of the small number of respondents. When enough experts are included, additional expert respondents do not result in new information not mentioned by other respondents, or the ratio of responses indicating one answer versus another remains the same with additional responses. Reaching this critical level of responses provides a sense of confidence that the responses are representative of the "truth," as a consensus can be seen across the expert respondents. This consensus was reached across the total global perspective in this survey, as evident by the repetition of named startup materials with less than 5% (12/292) of responses not fitting into one of eight main groups. However, when broken into regional subsections, it is less clear whether this type of consensus was reached.

While this survey provides new information about the types of materials that are used during cookstove startup, there are still many gaps related to understanding startup practices. The use of respondents with experience in the cookstove field allowed us to gather information from relatively few individuals in a resource-effective way, while still representing a wide variety of regions and cooking practices. However, further work to survey actual cookstove users would be beneficial to obtain a more comprehensive understanding of practices across different communities and to ensure that the experts' responses are representative of the prevailing real-world conditions. Most respondents indicated that they had not ever formally collected data on cookstove startup practices, which means their responses are subjective. While the repetition and overlap of responses across experts provides confidence in their responses, more empirical evidence would be beneficial. All respondents indicated on-the-ground field study experience;
researchers currently involved in cookstove studies could add some survey questions on startup practices to their existing studies. For example, respondents indicated that there may be some variability in startup practices according to location, primary stove type, main cooking fuel type, seasonality, or other factors – yet, conclusions about how these factors play into a cookstove user's choice in startup practice are limited by the small size and representation from our survey. Future surveys could be conducted to better understand what determines startup material choice, which would be critical to any intervention work aimed at changing startup practice behaviors for purposes of reducing exposures and improving health.

More empirical evidence would help support the information provided by the experts in this survey. Field studies conducted around the world could incorporate systematic observations or simple surveys about the startup practices used in the studied communities. Additionally, work is needed to understand how various startup materials contribute to the health impacts of cookstove use. Since individuals may experience high exposures from being in close proximity to the stove during the startup process, these emissions may be of particular importance when considering the overall global impact of cookstoves on human health. The pollutant mixture emitted from burning startup materials is presumably different than the mixture emitted from the main cooking process, particularly for rubber and plastic materials, and therefore the health and climate impacts may vary. However, such emissions remain poorly documented. Further, information is lacking on how emissions differ across different types of startup materials. The results presented here can serve as a foundation for further work, such as justification for the choice of startup materials to test in the laboratory. The reported commonality of a small number of startup material types is useful as it can provide confidence that results from emission campaigns that are only able to test a small number of materials could still be generalizable to a wide population.

CHAPTER 4: CHEMICAL COMPOSITION AND EMISSIONS FACTORS FOR COOKSTOVE STARTUP (IGNITION) MATERIALS

Summary

Air pollution from cookstoves creates a substantial human and environmental health burden. A disproportionate fraction of emissions can occur during stove ignition (startup) compared to main cooking, yet startup material emissions are poorly quantified. Laboratory tests were conducted to measure emissions from startups using kerosene, plastic bags, newspaper, fabric, food packaging, rubber tire tubes, kindling, footwear, and wood shims. Measured pollutants included: fine particulate matter mass (PM_{2.5}), PM_{2.5} elemental and organic carbon, methane, carbon monoxide, carbon dioxide, benzene, and formaldehyde. Results demonstrate substantial variability in the measured emissions across materials on a per-startup basis. For example, kerosene emitted 496 mg PM_{2.5} and 999 mg CO per startup, whereas plastic bags emitted 2 mg PM_{2.5} and 30 mg CO. When considering emissions on a per-mass basis, the ordering of materials from highest-to-lowest emissions changes, emphasizing the importance of establishing how much material is needed to start a stove. The proportional contribution of startups to overall emissions varies depending on startup material type, stove type, and cooking event length; however, results demonstrate that startup materials can contribute substantially to a cookstove's emissions. Startup material choice is especially important for cleaner stove-fuel combinations where the marginal benefits of reduced emissions are potentially greater.

Introduction

Exposure to household air pollution generated by residential burning of solid fuel is a top-ten risk factor for disease globally (Gakidou et al. 2017). These exposures contributed to approximately 77 million disability-adjusted life years in 2016, including over 2.5 million premature deaths (Gakidou et al. 2017). Efforts to reduce the harmful effects of cookstoves have achieved only modest impact (Alexander et al. 2017; Clark et al. 2013a; McCracken et al. 2011; McCracken

et al. 2007), in part because emissions reductions demonstrated in the laboratory have not translated to the real world (Roden et al. 2009). One important source of uncertainty in cookstove emissions/exposures is the contribution from cookstove startup (i.e., the act of igniting a stove).

A range of materials, such as newspaper, plastics, and agricultural wastes, are used to ignite cookstoves (see Chapter 3). The composition of these materials differs from main cooking fuels (e.g., wood, charcoal); thus, the emission profiles (and potential health impacts) from these materials may also differ from cooking fuels. Yet, only one study was identified that compared emissions from lighting stoves with different startup materials; this study compared only PM_{2.5} emissions from kerosene and plant stalks (Arora et al. 2014). Several studies have noted the contribution of stove ignition to overall cooking emissions. For example, Carter et al. (2014) reported that lighting contributed to 34 to 45% of overall PM_{2.5} emissions among Chinese gasifier stoves. Bhattacharya et al. (2002) demonstrated that lighting a stove with kerosene-soaked kindling from the top of the fuel bed (versus the bottom) resulted in decreased emissions of carbon monoxide and nitric oxides. Similarly, Lask and Gadgil (2017) showed that using a 'lighting cone' with charcoal stoves reduced emissions of ultrafine particles and carbon monoxide. Yet, no studies to date have systematically isolated startup emissions as a function of startup material type (Arora and Jain 2016). Most laboratory studies report using kerosene or highly-processed wood shims to ignite stoves, but these materials represent only a fraction of real-world startup material types (Arora and Jain 2016).

Previously, we surveyed experts about materials used to start cookstoves (see Chapter 3). The survey identified eight common materials: accelerants (e.g., kerosene, waxes), plastic (e.g., plastic bags, packaging), paper, kindling, agricultural wastes, high-resin woods (e.g., *ocote*, pine), fabric scraps, and rubber (e.g., tires, footwear). We characterized emissions from these materials in a laboratory-based study. We developed a protocol to replicate real-world startup events while simultaneously isolating emissions from the startup material. We measured emissions of particulate matter mass less than 2.5 microns in aerodynamic diameter (PM_{2.5}),

PM_{2.5} elemental and organic carbon (EC, OC), carbon monoxide (CO), carbon dioxide (CO₂), methane (CH₄), various carbonyls (including formaldehyde and acetaldehyde), and benzene, toluene, ethylbenzene, and xylenes (collectively, BTEX).

Methods

Experimental Setup

Tests were performed within a total-capture, constant-volume dilution hood. Particulate and gas sampling probes operated at isokinetic flowrates were located at the duct centerline, from 1 to 3 m downstream of the hood plenum. The experimental setup is shown in Figure B.1.

Emissions Measurements

Carbon dioxide, CO, and CH₄ were measured at 1 Hz using nondispersive infrared detection (Siemens Ultramat 6E, Siemens AG, Germany). PM_{2.5} was measured gravimetrically. Elemental and organic carbon were measured offline using an OCEC Analyzer (Sunset Laboratory, OR, USA) (Maimone et al. 2011; Subramanian et al. 2004). Select carbonyl compounds were collected on 2,4-dinitrophenylhydrazine cartridges and analyzed offline using high-performance liquid chromatography with ultraviolet detection. Whole air samples were analyzed offline for BTEX using gas chromatography/mass spectrometry. More details are provided in Appendix B.

Material Types and Preparation

Materials tested included: laboratory-grade kerosene, plastic grocery bags, foil-lined food packaging, newspaper, dry kindling material, bicycle inner tubes, synthetic foam-based footwear, and fabric (Table 4.1). Additionally, we tested wood shims because they are often used in laboratory tests to ignite stoves (as is kerosene). Materials were prepared into pieces of roughly uniform size, shape, and composition. Materials were sized to represent how they would be used in a real-world situation. For example, rather than cutting the bags or packaging into small pieces, we kept the materials in their natural units (e.g., whole bags or wrappers).

<u>Test Protocol</u>

Materials were burned in a metal, non-insulated stove with an L-shaped combustion chamber ($9.5 \times 17 \times 21$ cm), chosen to mimic the combustion chamber of a simple rocket-elbow stove. Pilot tests were conducted to determine the quantity of each startup material that fit in the stove opening and the number of replicate startup events needed to achieve quantification limits for gravimetric PM_{2.5} analysis. The test protocol involved conducting multiple startup events over a 20 to 30 minute period. Pre-weighed bundles of startup material were ignited in the stove using a single match and burned until no longer visibly smoldering or until CO levels were below 4 ppm (whichever occurred first). Remaining material and ash were removed from the stove and weighed. This process was repeated two to seven times per test, dependent on the material type (Table B.1). No main fuel was present in the stove, to allow for the isolation of emissions from the startup material only. Samplers for offline analysis were run continuously throughout the test. Background tests were conducted by running all sampling instruments for 20 minutes without burning a startup material, randomly in the test sequence once per test day. Additionally, CO₂, CO, and CH₄ were measured 10-minutes pre- and post-test.

We conducted the tests in random order. Each material was assigned to be tested three times. Additional tests were conducted following deviations from the test protocol, instrument errors, or suspected higher-than-normal background levels (based on concurrent activity in the laboratory).

<u>Analysis</u>

Data processing and analysis were conducted using R (version 3.3.1, The R Foundation for Statistical Computing). Offline measurements were background corrected by subtracting the study-average background values. Time-resolved data (CO₂, CO, and CH₄) were background corrected using the 10-minute pre-/post-test average mixing ratio. Per-startup emissions factors (mg pollutant/startup) were determined by dividing the total emissions per test (which contained multiple startup events) by the number of startup events for that test. The number of startup events

per test was calculated on a mass basis (total mass of material burned within test divided by the average mass of a startup bundle for that material type [Table 3.1]). We then averaged per-startup emissions factors across multiple test replicates. The resulting unit of mean emissions per startup provides an estimate of how much emissions would be expected from a single cookstove startup event. Coefficients of variation (COVs) were calculated as the standard deviation divided by the mean (presented as a percentage) for each material type. We also calculated the mass of pollutant emitted per unit mass of material burned (calculated for each test, which contained multiple startup events, then averaged across test replicates). Modified combustion efficiency (MCE), a metric that indicates the completeness of combustion, was calculated for each test as the ratio of emitted CO_2 divided by the sum of emitted CO and CO_2 .

We estimated the contribution of startup materials to the overall emissions of several representative stove technologies that operate across International Organization for Standardization (ISO) Performance Tiers (ISO 2018). ISO Tier limits are defined by the mass of CO and PM_{2.5} emitted per unit energy delivered to the cooking vessel (mg/MJ_{delivered}). We defined the emissions for each stove as reported by Jetter et al. (2012) and the Global Alliance for Clean Cooking (2018). We defined the energy needs of a cooking event as 1.67 MJ_{delivered} (approximately equivalent to bringing five liters of water from 20°C to 100°C). This was chosen because five liters can be accommodated by most cookstove styles and represents a cooking activity that is commonplace around the world. This procedure also mimics the process used for many laboratory stove testing protocols, including the Water Boiling Test, which are the sources of data that underpin the ISO Tiers and are how the emissions factors were derived in Jetter et al. (2012). We added the emissions per startup to each stove's emissions per cooking event, assuming one startup occurs per cooking event. We assumed the startup material does not contribute to the energy delivered and that the stove type does not change combustion efficiency of the startup.

Results

Test characteristics

At least three tests were completed per material, with two to seven startup events per test (Tables 3.1 and B.1). One additional test was conducted for kerosene, fabric, and food packaging and two additional tests were conducted for newspaper (resulting in 32 tests across the nine materials). Reasons for additional tests included missing carbonyl data for one kerosene and one newspaper test, missing BTEX data for one fabric test, and concerns over potentially higher background levels in one newspaper and one food packaging test due to concurrent activity in the laboratory. However, background CO and CO₂ levels for these tests were consistent with the expected background levels. As a result, no tests or specific pollutant data were removed from the dataset prior to analysis.

Emissions per Startup Event

Mean emissions per startup (average value across all tests) are shown in Table 4.2 (additional carbonyl compounds are provided in Table B.2). Emissions by test replicate are shown in Figure 4.1 for several key pollutants (PM_{2.5}, CO, benzene, toluene, formaldehyde, acetaldehyde, EC, and OC). Emissions were consistent across replicates for most materials and pollutants. Coefficients of variation were less than 25% for 36 of the 54 key pollutant/material combinations. The average COV across materials was 31% for acetaldehyde and benzene, 29% for toluene, 19% for CO, and 12% for formaldehyde. PM_{2.5} mass had a COV of 56% across materials but was less than 35% for all materials except footwear, food packaging, and plastic bags.

PM_{2.5} emissions varied from an average of 2 mg/startup for plastic bags to 496 mg/startup average for kerosene (see Figure 4.1, Table 4.2). Kindling (355 mg/startup), footwear (224 mg/startup), inner tubes (151 mg/startup), and newspaper (127 mg/startup) also had relatively high PM_{2.5} emissions. Emissions of PM_{2.5} were lower for wood shims (45 mg/startup), fabric (14 mg/startup), food packaging (8 mg/startup), and plastic bags (2 mg/startup).

Fuel Type	Fuel description	Number of tests	Average fuel mass (g) per startup event <i>(range)</i>	Average burn time (min) per startup event <i>(range)</i>
kerosene	Laboratory grade kerosene (approximately 15 grams/startup)	4	15.7 (14.5, 17.6)	4.0 (2.1, 6.6)
kindling	Dried twigs and leaves from willow oak trees cut into pieces smaller than 8 in long x 2 in wide, mixed with small amounts of dried grasses, weeds, and wood shavings (two handful/startup)	3	19.9 (16.2, 25.2)	3.7 (2.9, 4.3)
footwear	Foam flip-flop/sandals (ethylene-vinyl acetate [EVA]/rubber composite ^a), cut into pieces approximately 4 in long x 1 in wide (two strips/startup)	3	6.9 (5.0, 8.8)	8.8 (6.5, 10.8)
inner tubes	Used bicycle inner tubes (butyl rubber/latex ^a) cut into pieces approximately 4 in long x 1 in wide (two strips/startup)	3	4.4 (3.3, 5.2)	5.0 (3.6, 6.6)
newspaper	Printed newspaper sheet (appx. 15x22 in), crumpled into a ball (two sheets/startup)	5	18.7 (13.6, 24.5)	2.9 (1.9, 4.8)
wood shims	Two wooden shims stapled together with a small wooden separator (one pair/startup)	3	17.6 (11.8, 21.5)	10.2 (7.3, 13.3)
fabric	Plain colored cotton t-shirts (no ink/graphics), cut into pieces approximately 6 in long x 1 in wide (two pieces/startup)	4	2.7 (0.9, 3.5)	2.8 (2.0,4.7)
food packaging	Wrappers (foil-lined polypropylene ^a) from individual-sized snack foods (1-3 wrappers/startup, aim for 10 grams total/test)	4	2.5 (1.1, 4.2)	4.1 (1.8, 6.8)
plastic bags	Brown and white plastic grocery bags (polyethylene ^a) with some ink design, tied in a lose knot (two bags/startup)	3	1.8 (0.7, 2.8)	3.4 (2.2, 4.5)

Table 4.1 Experimental Matrix. A total of 39 tests were conducted across nine startup fuel types (minimum of three tests per fuel).

^aMaterial composition is presumed based on online search of material makeup.

A summary of fine particle emissions is provided in Figure 4.2. The highest EC and OC emissions were for kerosene (181 mg EC/startup and 238 mg OC/startup). Elemental carbon emissions from other materials were below 80 mg/startup (footwear: 77 mg/startup; inner tubes: 69 mg/startup; kindling, newspaper, and wood shims: 8 mg/startup; food packaging: 5 mg/startup; fabric: 2 mg/startup; plastic bags: below background). Similarly, OC values were high for kindling (212 mg/startup) in addition to kerosene, but lower for other materials (newspaper: 73 mg/startup; inner tubes: 60 mg/startup; wood shims: 28 mg/startup; fabric and plastic bags: 5 mg/startup, footwear: 4 mg/startup; food packaging: 1 mg/startup). As seen in Figure 4.2, the relative contribution of EC and OC to overall emissions varied considerably; for kerosene the ratio of EC to OC was 0.76 and the ratio of EC to total PM_{2.5} mass was 0.36. Emissions of OC were more dominant for wood shims, kindling, newspaper, and fabric resulting in EC to OC ratios of less than 1 (0.29, 0.04, 0.10, and 0.40, respectively) and very low EC to total PM_{2.5} mass ratios (0.18, 0.02, 0.06, and 0.14, respectively). Conversely, EC fractions were greater than OC fractions for footwear, food packaging, and inner tubes (EC to OC ratios of 19.25, 5.0, and 1.15, respectively); EC to $PM_{2.5}$ mass ratios were also higher for these materials (0.34, 0.625, and 0.46, respectively). Assuming the organic mass to organic carbon ratio is in the 1.2 to 1.5 range (Reece et al. 2017; Turpin and Lim 2001), carbonaceous particles account for close to 100% of the PM_{2.5} mass emitted from all materials (Figures B.5 and B.6). Any non-speciated PM_{2.5} mass is likely comprised of ionic compounds (Reece et al. 2017; Turpin and Lim 2001).

Carbon monoxide emissions ranged from 30 mg/startup for plastic bags to 999 mg/startup for kerosene. Emissions of CO were more dichotomized across materials than PM_{2.5} emissions. Plastic bags, inner tubes, footwear, fabric, and food packaging emitted less than 150 mg/startup for any given test, whereas kerosene, kindling, newspaper, and wood shims emitted 319 to 1000 mg/startup. Average CO emissions were highest for kerosene and kindling (488 and 999 mg/startup, respectively) and lower for fabric, food packaging, and plastic bags (58, 46, and

30 mg/startup, respectively). However, CO emissions were also low for footwear (45 mg/startup) despite higher PM_{2.5} emissions for this material.

Total BTEX emissions followed a similar trend as CO in that they are highest for kerosene, kindling, and newspaper, whereas inner tubes, fabric, food packaging, footwear, and plastic bags have relatively lower BTEX emissions (Figure 4.3). Kerosene produced the highest total BTEX emissions (28.1 mg/startup, driven primarily by higher benzene at 18.5 mg/startup). Kindling had the second highest (15.9 mg/startup) followed by newspaper (10.2 mg/startup). Other materials emitted 4 mg/startup total BTEX or less. Benzene accounted for 50% or more of total BTEX emissions for all materials except footwear (newspaper: 76%, inner tubes: 72%, kerosene: 66%, fabric: 58%, kindling: 54%, food packaging: 51%, plastic bags: 50%, footwear: 23%). Toluene accounted for 13 to 26% of total BTEX emissions (kindling and wood shims: 26%, footwear: 23%, plastic bags: 20%, fabric: 19%, food packaging: 18%, newspaper: 16%, inner tubes: 14%, kerosene: 13%). Ethylbenzene was less than 10% of BTEX emissions for all materials. Though no pattern is seen for BTEX emissions across materials, similarities are apparent for wood shims and kindling (both wood-based materials): total BTEX emissions were around 54% benzene, 26% toluene, and 13 to 15% xylenes. Plastic bags and food packaging (both thin plastic materials) also had a similar compositional breakdown: approximately 50% benzene, 20% toluene, 25% xylene, and 5 to 6% ethylbenzene. Finally, newspaper and inner tubes were similar: approximately 75% benzene, 15% toluene, and 2 to 3% ethylbenzene, though with varying xylene (12% for inner tubes versus only 5% for newspaper). Differences in the BTEX compositions were mainly driven by changes in the benzene to toluene ratio, which ranged from 1.0 to 5.2 across the different material types (kerosene: 5.2, inner tubes: 5.0, newspaper: 4.7, fabric: 3.1, food packaging: 2.8, plastic bags 2.5, kindling: 2.1, footwear: 1.0).

Table 4.2 Mean Emissions per Startup Event. Values are background adjusted. n = number of tests, each test contained multiple startup events (2 to 7, see Table B.1) to achieve the total number of startups listed across all tests.

	Emissions (mg) Per Startup Event (Average [min, max])											
	kerosene (n=4 tests; 21 total startups) ^a	kindling (n=3 tests; 9 total startups)	footwear (n=3 tests; 7 total startups)	inner tubes (n=3; 12 total startups)	newspaper (n=5 tests; 33 startups) ^a	wood shims (n=3 tests; 6 startups)	fabric (n=4 tests; 24 startups)	food packaging (n=4 tests; 23 startups)	plastic bags (n=3 tests; 18 startups)			
	Carbon Gases											
carbon	45543	25029	9281	11359	29726	27803	4307	5277	2788			
dioxide	(43663, 47474)	(22622, 26252)	(6857, 13248)	(10875, 12227)	(28572, 30808)	(26723, 28733)	(4197, 4409)	(3706, 8155)	(2174, 3111)			
carbon monoxide	488 (477, 499)	999 (939, 1036)	45 (35, 58)	124 (108, 145)	844 (662, 986)	418 (319, 497)	58 (51, 64)	46 (31, 84)	30 (26, 38)			
methane	48 (31, 89)	62 (57, 69)	6 (<bg, 9)<="" td=""><td>9 (6, 12)</td><td>46 (29, 92)</td><td>27 (22, 33)</td><td>9 (<bg, 36)<="" td=""><td>27 (<0.5, 109)</td><td>1 (<0.5, 3)</td></bg,></td></bg,>	9 (6, 12)	46 (29, 92)	27 (22, 33)	9 (<bg, 36)<="" td=""><td>27 (<0.5, 109)</td><td>1 (<0.5, 3)</td></bg,>	27 (<0.5, 109)	1 (<0.5, 3)			
		-	-	Partic	ulate Matter							
PM _{2.5} mass	496 (371, 564)	355 (333, 397)	224 (73, 499 ^b)	151 (136, 174)	127 (91, 143)	45 (27, 57)	14 (11, 17)	8 (1, 13)	2 (<bg, 7)<="" td=""></bg,>			
PM _{2.5} EC	181 (80, 293)	8 (6, 12)	77 (58, 89)	69 (61, 83)	8 (4, 12)	8 (2, 12)	2 (<0.5, 3)	5 (<bg, 10)<="" td=""><td><bg (<bg,="" 1)<="" td=""></bg></td></bg,>	<bg (<bg,="" 1)<="" td=""></bg>			
PM _{2.5} OC	238 (136, 348)	212 (188, 257)	4 (1, 7)	60 (34, 87)	73 (58, 85)	28 (12, 37)	5 (4, 6)	1 (<bg, 3)<="" td=""><td>5 (3, 7)</td></bg,>	5 (3, 7)			
				Volatile Org	anic Compound	ls						
benzene	18.5 (16.9, 20)	8.5 (8, 9.3)	0.3 (0.3, 0.4)	1.8 (1, 3)	7.8 (4, 10.4)	2.2 (1.7, 2.6)	0.7 (0.4, 1.2)	0.7 (0.5, 0.9)	0.3 (0.2, 0.6)			
ethyl- benzene	1.4 (1.3, 1.5)	1.1 (1.1, 1.2)	0.1 (<0.1, 0.2)	0.1 (<0.05, 0.1)	0.3 (0.2, 0.5)	0.2 (0.2, 0.3)	0.05 (<0.05, 0.1)	0.1 (0.5, 0.1)	<0.1 (<0.05, 0.1)			
m+p xylenes	2.7 (2.5, 3.2)	1.5 (1.5, 1.6)	0.5 (0.3, 0.6)	0.2 (0.1, 0.4)	0.3 (0.2, 0.4)	0.4 (0.3, 0.5)	0.2 (0.1, 0.3)	0.2 (0.1, 0.4)	0.1 (<0.1, 0.1)			
o-xylene	1.9 (1.6, 2.2)	0.7 (0.7, 0.7)	0.2 (0.1, 0.3)	0.1 (<0.1, 0.2)	0.1 (0.1, 0.2)	0.2 (0.1, 0.2)	0.1 (0.1, 0.1)	0.1 (0, 0.3)	0.1 (0, 0.1)			
toluene	3.6 (3.1, 3.9)	4.1 (4, 4.1)	0.3 (0.2, 0.4)	0.4 (0.3, 0.5)	1.7 (0.9, 2.3)	1.1 (0.9, 1.2)	0.2 (0.1, 0.5)	0.3 (0.2, 0.3)	0.1 (0.1, 0.2)			
Carbonyls												
acet- aldehyde	2.2 (2, 2.4)	17.6 (5.2, 24.1)	0.6 (0.6, 0.6)	0.6 (0.4, 1)	3.8 (0.7, 7.1)	4.4 (3.3, 5.2)	1.1 (0.8, 1.3)	0.9 (0.7, 1.1)	0.6 (0.6, 0.7)			
acrolein	<0.05 (<0.1, 0.1)	1.2 (0.9, 1.6)	0.05 (<0.05, 0.1)	<0.05 (<0.05, <0.05)	0.6 (0.2, 0.8)	0.1 (0.1, 0.2)	<0.05 (<0.05, 0.1)	<0.05 (<0.05, <0.05)	<0.05 (<0.05, <0.05)			
form- aldehyde	4.8 (4.5, 5.3)	26.9 (20.4, 31.4)	1.3 (1.2, 1.4)	1.7 (1.5, 1.8)	17.1 (15.4, 19)	8.3 (6.4, 9.3)	1.7 (1.6, 1.7)	1.1 (1, 1.4)	1.1 (0.9, 1.2)			
croton- aldehyde	0.2 (0.2, 0.2)	1.5 (0.8, 1.9)	0.1 (<0.05, 0.2)	<0.05 (<0.05, 0.05)	0.4 (0.2, 0.5)	0.4 (0.3, 0.4)	<0.1 (0.05, 0.1)	0.05 (<0.05, 0.1)	0.1 (<0.05, 0.1)			

^aCarbonyls had one less n.

^bThe 499 mg/startup PM_{2.5} value is an unexplained outlier, when removed, the average is 86 mg/startup event.

EC = elemental carbon; OC = organic carbon; $PM_{2.5}$ = particulate matter mass less than 2.5 micrometers in diameter.

<BG = measured concentration was below mean background concentration; therefore, emissions factors could not be calculated.



Figure 4.1 Emissions (mg) per Startup Event for Select Pollutants. Each circle represents the calculated average emissions per startup event for an individual test. Materials are ordered from left to right by highest to lowest emissions of PM_{2.5} mass.



Figure 4.2 Emissions of Fine Particulate Mass, Elemental Carbon, and Organic Carbon for Different Startup Materials. Emissions in milligrams of pollutant per startup event. Inset highlights results for lower-emitting materials. The lighter grey bar on footwear emissions shows $PM_{2.5}$ emissions with the outlier included.



Figure 4.3 Comparison of the Relative Composition of BTEX Emitted Across Different Startup Materials. Emissions in milligrams of pollutant per startup event. Inset highlights results for lower-emitting materials. The stacked bar height represents the total BTEX emissions.

Acetaldehyde and formaldehyde emissions were higher for kindling than all other startup materials tested (acetaldehyde: 17.6 mg/startup, formaldehyde: 26.9 mg/startup). Other materials generally emitted less than a quarter of the amount of acetaldehyde and less than one third the amount of formaldehyde as kindling (with the exception of formaldehyde emissions from newspaper, which were about two-thirds the amount emitted from kindling). The next highest emitting materials included wood shims (acetaldehyde: 4.4 mg/startup, formaldehyde: 8.3 mg/startup), newspaper (acetaldehyde: 3.8 mg/startup, formaldehyde: 17.1 mg/startup), and kerosene (acetaldehyde: 2.2 mg/startup, formaldehyde: 4.8 mg/startup). Acetaldehyde emissions from all other materials were around 1 mg/startup or less (fabric: 1.1, food packaging: 0.9, plastic bags, footwear and inner tubes: 0.6). Formaldehyde emissions from all other materials were between 1 to 1.7 mg/startup (inner tubes and fabric: 1.7, footwear: 1.3, food packaging and plastic bags: 1.1).

Emissions on per-Mass Basis

Emissions on a per-mass basis are presented in Table B.3 and Figures B.2, B.3, and B.4. The ordering of materials from highest-to-lowest emissions changes when considering emissions in this unit compared to a per-startup basis (Figure 4.1 vs. Figure B.2), emphasizing the impact of the different mass of material needed for each startup event depending on the material type. An overall trend is maintained, however, wherein kerosene, kindling, and sometimes newspaper appear among the higher-emitting materials for non-CO₂ pollutants (e.g., PM_{2.5}, CO, BTEX, and carbonyls) while plastic bags, food packaging, and fabric emit less of the non-CO₂ pollutants.

Modified Combustion Efficiencies

Mean MCEs ranged from 0.93 (kindling) to greater than 0.99 (footwear, food packaging) (see Table B.4). MCE within a single startup event peaked quickly then tapered towards the end. The range of MCEs for a single test were low and within the range of predominantly flaming combustion. Mean 5th percentile values were lowest for newspaper and kindling (0.86 each) but

above 0.90 for all other materials. Mean 95th percentile values ranged from 0.98 to greater than 0.99 across all materials.

Impact of Startup Emissions on ISO Tiers

The implications of adding a startup event to a stove's overall emissions are illustrated in Figure 4.4. For each stove, addition of a startup using plastic bags (the lowest-emitting material studied) does not change the stove's Tier rating. However, a kerosene startup (the highest-emitting material studied) results in certain stoves shifting Tiers. For relatively "cleaner" cookstoves (Tiers 3 through 5), the contribution from a startup material to overall emissions is large proportional to the emissions from the cooking event. The limit values for Tiers 3 through 5 are narrow, so shifts from one tier to the next can happen with only a small change in emissions (owing to the nonlinear tier spacing). The addition of the kerosene startup therefore shifts these stoves into "dirtier" Tiers. For example, the Mimi Moto gasifier shifts from Tier 4 to Tier 1 when accounting for a kerosene startup. Within the higher-emissions Tiers (Tier 0, 1, and 2), the contribution of a startup, regardless of startup material choice, is smaller compared to the emissions from the stove and the tier limits are wider. As a result, the shifts are less drastic. For example, the Berkeley Darfur and Protos stoves each shift one tier, whereas the Envirofit G-3300 stays within Tier 1 (shifting from the lowest end of the Tier to the highest limit).



Figure 4.4 Influence of Startup Material on Cookstove Emissions. The ISO Tiers for cookstove performance are plotted as ranges of CO and PM_{2.5} emission factors (log scale). Emissions for select stove types (Global Alliance for Clean Cookstoves 2018; Jetter et al. 2012) are shown as solid-filled circles; open symbols depict the change in emissions after including the contribution from selected startup materials. For illustrative purposes, arrows demonstrate the magnitude of these effects for the Mimi Moto stove (a low-emissions, Tier 4 gasifier). Calculations assume one startup event for a single 1.67 MJ cooking event (i.e., bringing five liters of water to boil at normal temperature and pressure).

Discussion

Results demonstrate that emissions profiles vary across startup materials, likely due to a combination of factors. For example, different material shape and chemical composition likely effect both the rate and efficiency of combustion, leading to differential emissions. Further, the mass of a given material needed to complete a startup event also plays a role in overall emissions, regardless of combustion efficiency (i.e., 1.8 g of plastic bags vs. 17.6 g of wood shims). Kerosene and kindling demonstrated consistently high emissions across multiple pollutants on a per-startup basis – often an order of magnitude higher than other materials. Newspaper and wood shims were also consistently among the highest emitters of the pollutants measured. Plastic bags were the lowest-polluting choice of the materials studied across nearly every pollutant measured. Food packaging and fabric were also consistently lower emitters. Kerosene is a very common fire starter used globally and also used as a main fuel in some advanced stove designs. Our data indicates that, of the materials characterized, kerosene is the highest emitted of the pollutants measured on a per-startup basis.

Implications of Startup Material Emissions on Overall Cookstove Emissions and Exposures

The use of startup materials adds to the emissions for all cookstoves, shifting towards higher total emissions from a cooking event. However, the extent to which this shift matters depends on the base emissions from the main cooking event. For example, emissions from a main cooking event using wood fuels in a three stone fire are estimated to be on the order of 0.5 to 1 g PM_{2.5} per liter of water boiled per hour; emissions from a more advanced wood-burning gasifier are lower, around 0.03 g/liter-hour for PM_{2.5} (Jetter et al. 2012). For a simple cooking event such as an hour of simmering one liter volume of water or food, a kerosene startup (at approximately 0.5 g of PM_{2.5}) would contribute 33 to 50% of total PM_{2.5} emissions from a three stone fire, yet 94% of total PM_{2.5} emissions from a gasifier. Conversely, a low-emitting startup material such as plastic bags, which registered approximately 0.002 g PM_{2.5} per startup event, would only contribute to less than 1% of total PM_{2.5} emissions from a three stone fire and around

5% of PM_{2.5} emissions from a gasifier. Notably, some stoves are not ignited every time a cooking event occurs; instead, they may be ignited once and then remain lit throughout a full day or longer. Additionally, factors such as fuel type, fuel moisture content, cooking practices, and stove condition can affect the main stove emissions in real-world settings. Impact assessments should consider how the startup contributes under different stove use patterns for a more accurate comparison of total emissions across stoves.

An additional consideration, however, is that evidence suggests PM-associated health effects from cookstove air pollution exposures may be nonlinear: greater changes in relative health risks occur with increases in exposure at lower exposure levels than with the same increases at higher starting exposure levels (Burnett et al. 2014; Smith and Peel 2010). The 2018 ISO Cookstove Performance Tiers reflect this nonlinear health relationship, with lower-emission Tiers occupying a tighter distribution than the higher-emission Tiers (ISO 2018). Considering how a startup event impacts total emissions for a cooking event within this Tier system, our results illustrate that the differences in emissions across different startup materials may be most important from a health perspective when considering lower-emission/higher-performance Tier stoves. The emissions differences across startup materials become less important in the lower-performance Tiers, where the base emissions from the main cooking event are higher (so differences in emissions from one startup material to another are proportionally less meaningful to the overall emissions).

We measured emissions from the startup combustion, not exposure. To draw conclusions about the health impact of startups, further work is needed to translate emissions factors to estimated indoor air concentrations and subsequent exposure levels. While relative differences in emissions can be expected to roughly translate to relative differences in exposure, there are several factors that could result in a disconnect across different startup materials. For example, behavioral aspects such as how the cook interacts with the stove and startup material during the ignition process could change combustion dynamics or impact breathing.

Metrics for Emissions: Per Startup vs. Mass Burned

We standardized emissions to "per-startup" units to approximate real-world lighting emissions in a way that does not depend on differing burn rates and densities of startup materials. Relative differences in emissions on a per-startup basis can be reasonably expected to translate to relative differences in exposure and therefore health risks.

Emissions factors on a mass basis (grams pollutant emitted per kilogram material burned, as shown in Table B.3 and Figures B.2 and B.3) allow the emissions reported here to be related to other combustion applications, such as military waste burning or residential trash burning in backyard burn pits. When considering emissions in these units, the differences across materials changes drastically from what is seen on a per-startup basis. Emissions of the measured pollutants become more similar between different startup materials. Emissions from kerosene, wood shims, kindling, and newspaper (which were relatively high across multiple pollutants on a per-startup basis) appear within the same range as emissions from fabric, plastic bags, food packaging, inner tubes, and footwear (relatively low on a per-startup basis). This emphasizes that the choice to compare emissions across multiple materials on a per-startup basis drives conclusions that depend on comparisons across materials and is therefore integral to the extrapolation of these laboratory-based findings to real-world settings.

Few reports exist detailing emissions from startup materials that are comparable to this study. Our measured emissions for CO, CO₂, methane, formaldehyde, benzene, EC (black carbon), and OC are all within an order of magnitude of those reported for open waste burning by Wiedinmyer et al. (Wiedinmyer et al.). The composition of open waste is undefined but presumably mixed material types; our emission factors could help refine estimates for certain types of waste. Additionally, some comparisons can be drawn between emissions from startup materials and general cookstove emissions factors. Our emission estimates for wood shim startups (g/kg CO: 24; PM_{2.5}: 3; benzene: 0.12; toluene: 0.06; acetaldehyde: 0.25; formadehyde: 0.47) are in line with reported emissions from wood-burning cookstoves

(g/kg CO: 17 to 60; PM_{2.5}: <10; benzene: 0.1 to 2.5; toluene: <0.5; acetaldehyde: 0.1 to 0.8; formaldehyde: 0.2 to 1.7) (Chen et al. 2007; Christian et al. 2010; Stockwell et al. 2015; Word Health Organization 2014). Studies of kerosene wick stoves cover a wide range of PM_{2.5} emissions (from 0.29 to 124 g/kg) (Lam et al. 2012; Word Health Organization 2014), potentially due to differences in stove type and operating conditions (Lam et al. 2012). Our estimates (32 g/kg) are in line with higher estimates from other research. Similarly, CO emission estimates for kerosene-burning cookstoves vary from very low (2.8 g/kg) (Chen et al. 2007) to within the range of our kerosene startup estimates (31 g/kg, compared to 11 to 21 g/kg or 27 g/kg) (Lam et al. 2012; Word Health Organization 2014).

Few studies have reported on carcinogenic emissions (e.g., benzene, acetaldehyde, formaldehyde) from cookstoves or biomass fuels. Our results indicate that these carcinogens are emitted from startup materials at levels that are important to human health compared to the main cooking event. For example, Zhang and Smith (Zhang and Smith 1999) report emissions of carbonyls from cookstoves burning different main fuel types (wood, coal, kerosene, and LPG) where acetaldehyde emissions were 0.02 to 0.17 g/kg and formaldehyde emissions were 0.02 to 0.14 g/kg. Our emissions factors were higher (acetaldehyde: 0.09 to 0.88 g/kg; formaldehyde: 0.19 to 0.91 g/kg). On a per-startup basis, acetaldehyde emissions ranged from approximately 0.5 to 18 g/startup for acetaldehyde and 1 to 27 g/startup for formaldehyde. Various reports indicate emissions from biomass burning range from less than 0.1 to 2.5 g/kg for benzene and 0.02 to 0.18 g/kg for toluene (Akagi et al. 2011; Stockwell et al. 2015; Tawfiq et al. 2015; Yokelson et al. 2013). These values are aligned with emissions from our startups (0.05 to 1.18 g/kg benzene, 0.05 to 0.23 g/kg toluene); which translate to per-startup emissions that can reach several grams.

Other Strengths and Limitations

We replicated aspects of real-world startups by burning small bundles of material within a generic stove. We expect test-to-test variability in emissions due to the inherent variability in

cookstove combustion and the startup process that the experiments were designed to capture. The COVs showed 12 to 56% variability between tests, dependent on the pollutant. However, there may be other contributors to variability in real-world emissions that we did not capture in our protocol. Startup material composition and quantity may be more variable than we captured. For example, the kindling startup material we tested was comprised of twigs, grasses, and weeds sourced from northern Colorado; kindling materials used to start cookstoves around the world can be highly varied across different regions based on the local flora.

We chose to burn the startup material only, without a main fuel bed. This was beneficial for isolating the emissions from just that startup material and ensuring that our integrated measurements did not capture any emissions from the main fuel, which would be harder to control across test replicates. Additionally, not all startup materials are appropriate for use with all fuel types, making selection of a consistent main fuel difficult. However, the burning dynamics of the startup material could be different when integrated with a main fuel type (for example, if the heating of a fuel bed under the startup material impacts the burn temperatures or airflow dynamics across the startup material). We observed predominately flaming combustion (mean MCE: 93 to 99%), which is desired when attempting to ignite fuel; different combustion conditions could lead to different emissions. Further research is needed to quantify the impact of interactions between startup, main fuel, and stove type on emissions.

Our study is limited in that we only tested a subset of startup materials that are used globally. A recent survey conducted by our group indicated that a wide variety of different materials are used to start stoves around the world, however those materials could reasonably be categorized into eight main groups of commonly used material types (see Chapter 3). The emissions data reported here cover the most common categories (accelerants [kerosene], plastics [plastic bags], paper [newspaper], kindling, rubber/rubber-like materials [inner tubes, footwear], and fabric). We additionally tested food packaging/wrappers as these materials as well as household trash in general were noted, though less frequently, within the survey. While our

selection of materials covers a range of startup materials globally, there could still be additional variation in emissions expected due to inherent variability in material types within these groupings. For example, plastic bags may not be representative of other types of plastic-based materials used during startup, such as plastic packaging/wrappings or plastic bottles. We did not test agricultural waste and high-resin pine woods (the remaining two of the eight common material type groups that emerged from the survey results) due to cost constraints and logistical challenges in procuring these materials. Emissions from these materials may be important to consider from health and climate perspectives, similar to the materials tested in this study, and should be included in future work.

Our study was more comprehensive than many previous laboratory emissions studies in the pollutant types characterized, including particulate matter characteristics, carbon gases, volatile organic compounds, and carbonyls. However, combustion emissions are a complex mixture and we did not measure all classes of pollutants expected to be emitted. For example, we did not measure dioxin and dioxin-like compounds or polychlorinated biphenyls. These highly toxic compounds are known to be emitted from burning halogenated plastics and papers (Hu et al. 2009; Verma et al. 2016; Zhang et al. 2015), so may be relevant for some startup material types. Some classes of unmeasured pollutants may follow different trends across the material types than the trends seen in our measured pollutants, which would impact conclusions about the overall health risks associated with different material types.

Previous studies aimed at understanding cookstove emissions as a whole have characterized emissions from the stove startup phase (Bhattacharya et al. 2002; Carter et al. 2014; Lask and Gadgil 2017). These studies predominately use kerosene or wood shims to ignite the main fuel and focus on understanding emissions from the ignition phase – the first minutes of the main fuel burning after it has been ignited with aid from the startup material – as opposed to understanding the emissions coming from the startup material. Other studies have noted the use of different materials to start cookstoves in the field, such as newspaper, kindling, lighter fluid, or

cow dung (Dutta et al. 2007; Roden et al. 2009), yet have not characterized how these different materials relate to measured emissions. To our knowledge, this is the first study to isolate and characterize pollutants emitted from a variety of real-world relevant cookstove startup materials.

CHAPTER 5: ACUTE EFFECTS ON BLOOD PRESSURE FOLLOWING CONTROLLED EXPOSURE TO COOKSTOVE AIR POLLUTION IN THE SUBCLINICAL TESTS OF VOLUNTEERS EXPOSED TO SMOKE (STOVES) STUDY

Summary

Exposure to household air pollution from solid fuel combustion for heating and cooking is a top risk factor for disease globally; cardiovascular diseases make up about half of the burden worldwide. We conducted a controlled human exposure study to investigate acute responses in blood pressure following exposure to cookstove-generated air pollution emissions. Forty-eight young adult, healthy subjects received six, two-hour exposures spaced at least two weeks apart: five cookstoves treatments (liquid petroleum gas [LPG], gasifier, fan rocket elbow, rocket elbow, and three stone fire) at fine particulate matter (PM_{2.5}) concentrations representative of the stove's emissions (10 to 500 μ g/m³) and a filtered air control (0 μ /m³). Immediately post-exposure, systolic pressure was lower for the three stone fire (500 μ g/m³ PM_{2.5}) compared to the control (0 μ g/m³ PM_{2.5}) (-2.3 mmHg, 95% CI -4.5, -0.1 mmHg). Systolic pressure was also lower for the other stove treatments but to a lesser extent. No differences were observed at three hours post-exposure for stove treatments compared to the control. At 24 hours post-exposure, systolic blood pressure was 2 to 3 mmHg higher for all treatments compared to the control, except for the rocket elbow stove. No clear differences were observed in diastolic pressure for any time point or treatment. These results contrast with other wood smoke controlled exposure studies where no immediate blood pressure responses were observed, but are consistent with evidence from ambient air pollution studies and field household air pollution studies.

Introduction

Exposure to household air pollution generated by residential stove use is a leading global risk factor for disease, contributing to over 77 million disability-adjusted life years - including over 2.5 million deaths in 2016 (Gakidou et al. 2017). Cardiovascular disease accounts for approximately half of this burden (Smith et al. 2014). Given the scope of this problem, much work

has focused on designing and disseminating improved or clean cookstove technologies that burn more efficiently and emit less pollution. The driving hypothesis is that use of clean cookstoves will reduce exposures and improve health compared to traditional cookstoves. While some studies have shown exposure and health benefits of improved stoves (e.g., Alexander et al. 2017; Clark et al. 2009; Clark et al. 2013a; Martin et al. 2011; McCracken et al. 2011; McCracken et al. 2007; Thomas et al. 2015), several cross-sectional studies and large intervention trials have failed to demonstrate significant health benefits (e.g., Alexander et al. 2017; Mortimer et al. 2017; Romieu et al. 2009; Smith et al. 2011). Despite investment into clean cookstove adoption (Global Alliance for Clean Cookstoves 2015; Martin et al. 2011), scientific understanding of "how clean is clean enough" is still limited.

The connection between household air pollution and cardiovascular health risk is primarily extrapolated from research on other pollution sources at the upper and lower ends of the particulate matter exposure continuum (i.e., active cigarette smoking, secondhand smoke, and ambient air pollution) (Smith et al. 2014). While several studies have demonstrated associations between cardiovascular disease risk and cookstove exposures (e.g., Alexander et al. 2015; Alexander et al. 2017; Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018; Burroughs Pena et al. 2015; Burroughs Pena et al. 2017; Clark et al. 2011; Clark et al. 2013a; Dutta and Ray 2012; McCracken et al. 2011; McCracken et al. 2007; McCracken et al. 2012; Neupane et al. 2015; Norris et al. 2016), the evidence is still limited. Field studies typically investigate only a single stove technology in a single population and participant adherence to intervention protocols (i.e., adoption of the stove) is often mixed; further, observational designs are potentially subject to bias and confounding, and many studies lack sufficient exposure characterization. Controlled exposure studies can complement field-based studies by examining the acute effects of exposure under carefully controlled conditions. Controlled exposure studies are limited in their ability to examine long-term exposures and clinical cardiovascular outcomes; however, measurement of acute-phase health markers along an established cardiovascular disease pathway can provide insight into the potential for clinically-relevant differences that would manifest following long-term exposures (Brook et al. 2010; Langrish et al. 2012).

Blood pressure is an established preclinical marker of cardiovascular disease risk that has been shown to change meaningfully in acute time frames (Lewington et al. 2002; Turnbull 2003; Vasan et al. 2001). Small increases in blood pressure, even within a normal range, can lead to increased risk of stroke, coronary heart disease, and heart failure (Ettehad et al. 2016). Studies on both ambient air pollution and household air pollution have provided evidence of an association with blood pressure, on various timescales (e.g., Brook et al. 2010; Brook et al. 2011; Clark et al. 2013a; Fuks et al. 2014; Langrish et al. 2012; McCracken et al. 2007; Norris et al. 2016; Urch et al. 2005).

The purpose of this study was to determine if controlled human exposure to air pollution generated by various cookstove designs influences blood pressure. Specifically, we explored associations between blood pressure and two hour exposures to pollution that covered a range of targeted fine particulate matter mass ($PM_{2.5}$) levels, from 10 to 500 µg/m³, at various post-exposure time points over 24 hours.

Methods

Eligibility Criteria and Recruitment Methods

Forty-eight healthy, non-smoking, young adult volunteers were recruited for this study. Participants who were eligible according to a self-reported recruitment questionnaire completed an additional in-person screening exam with the study staff and physician (details provided in Appendix C). Eligibility criteria were: 18 to 35 years old at the time of recruitment; never smokers; body mass index between 19 to 28 kg/m² with body weight greater than 110 pounds; no history of heart disease, diabetes, kidney disease, systemic sclerosis, or any chronic inflammatory disease such as asthma, arthritis, or severe allergies; normal non-hypertensive blood pressure and normal electrocardiogram at the screening exam; normal blood test results, including no evidence of iron-deficient anemia (as determined by blood results from screening exam);

spirometry values greater than 70% of the predicted value for the age/gender during the screening exam; not currently taking statins, anti-inflammatory medication, or other medications unless cleared by the study physician during the screening exam (oral contraceptives and some daily anti-depressant/anti-anxiety medications were approved for continued use); no use of tetrahydrocannabinol (marijuana) or illicit drugs within the past three months; no ear or abdominal/thoracic surgery in the past month; no cancer (current or in remission for less than six months); do not have a central intravenous line or port; have never had a mastectomy; do not have a pacemaker; not currently pregnant, breastfeeding, or planning a pregnancy within six months; not regularly exposed to smoke, dust, fumes, or solvents (occupationally or recreationally/at home), or regularly burned candles or incense, within the last three months; no history of claustrophobia; no fear of needles; not planning to donate blood during the timeframe of participation; not allergic to latex; and live within 20 miles of the study facility and not planning to move more than 20 miles away within six months. All study protocols were approved by the Colorado State University Institutional Review Board. All participants provided written informed consent.

Overarching Design

Each participant received six exposure treatments (a filtered air control and five levels of air pollution; see "Exposure Treatments and Administration") with a washout period between treatments (maximum six weeks due to holidays though typically just two or three weeks; sequences ranged from 13 to 16 weeks long). Participants were blinded to which treatment they received on each study session. Our study followed a Williams design; a Latin square crossover design that is balanced across treatments and first-order carry-over effects (Williams 1949). This design is robust to time invariant person-level factors (i.e., subject effects), because each person receives each treatment (Jones and Kenward 2014). It also reduces the possibility of confounding due to time variant factors that might differ from one study session to the next and impacts of the treatment orders (e.g., treatment-by-period interactions) (Jones and Kenward 2014). Under this

design, the 48 participants were divided into groups of four who completed the study together. We specified six unique treatment orders ("sequences"). Each treatment occurred one time per sequence. Across the six sequences, each treatment occurred exactly once in each time slot (e.g., first treatment session vs. second, third, etc.) and was both proceeded and followed by each other treatment exactly once. We assigned two groups (8 people) to each sequence. The first group of the sequence started their study sessions on Mondays and the second group started their sessions on Wednesdays of the same weeks. We conducted the study in three rounds (October 2016 to February 2017; March to June 2017; August 2017 to January 2018); within each round, two sequence groups alternated weeks until completion of all six treatments.

Participants were scheduled each study day with 30-minute staggered start times; each participant maintained the same start time for their six study sessions. Assignment of participants into sequence groups, week days, and time slots was random, but with consideration of the participant's availability (e.g., participants were recruited on a rolling basis into the ongoing study round and were allowed to specify whether they were not available to be placed into certain dates/time slots; participants were placed into the available date/time slots randomly). Participants were blind to their assigned sequence and researchers were blind to the treatment orders within each sequence during the assignment process.

Participants who missed a scheduled study session due to illness or unforeseen conflict were allowed to make up the missed treatment at the end of the sequence; makeups were not necessarily completed on the same day of week or starting time as their regular schedule. Makeups were conducted ten days to 14 weeks after the last scheduled treatment. Participants remained blind to the treatment during makeups. More detail on the overarching design is provided in Appendix C.

Study Session Protocol

Participants were instructed to abstain from medications (e.g., over-the-counter allergy or pain medications), nutritional supplements, and vitamins starting 72 hours prior to study day and

from caffeine, alcohol, strenuous exercise, and smoke exposures (e.g., campfires/wood stoves, cigarette smoke, marijuana smoke) starting 24 hours prior to study day, continuing through the end of the 24-hour follow-up period. Participants were also asked to avoid high-fat and high-cholesterol foods on study days. Surveys were administered to determine compliance with these protocols (see "Health and Covariate Measurements").

Following a day-of clearance by the on-site study physician, baseline health measurements were taken. Participants then spent two hours in the exposure chamber receiving the treatment. Additional rounds of health measurements were conducted starting immediately post-exposure and at three hours post exposure. Participants remained on site until the three-hour post-exposure measurements was completed, after which participants returned to home and/or work. A low-fat, low-cholesterol lunch was provided between the immediate post-exposure and three-hour post-exposure measurements; participants received the same lunch each study session. Participants returned for a final round of health measurements approximately 24 hours after the end of the exposure treatment. This was repeated six times (once for each exposure treatment level).

Exposure Treatments and Administration

Treatments consisted of a filtered air control and air pollution generated from five different types of cookstoves, each with an intended target $PM_{2.5}$ level (see Table 5.1). Cookstoves included a liquefied petroleum gas (LPG) stove operated with commercially available propane, a wood-burning gasifier operated with Douglas fir wood chips, a forced-draft (i.e., fan-powered) rocket elbow, a standard rocket elbow, and a traditional three stone fire each operated with milled Douglas fir wood sticks (see Appendix C). Target $PM_{2.5}$ levels were 10, 35, 100, 250, and 500 µg/m³, respectively. The filtered air (target $PM_{2.5}$: 0 µg/m³) was generated by pulling conditioned laboratory air through a high-efficiency particulate air (HEPA) filter. Pollution was generated within a total-capture fume hood, diluted with HEPA-filtered laboratory air, and then drawn into the exposure chamber at a constant flow rate. A nephelometer with a $PM_{2.5}$

size-selective cyclone (DustTrak DRX 8533, TSI Incorporated, USA) was calibrated to the wood and LPG stoves separately (based on gravimetric filter data). The DustTrak and a gas analyzer (Siemens Ultramat 6E, Siemens AG, Germany) were used to monitor PM_{2.5}, carbon monoxide (CO), and oxygen levels in the chamber in real time; humidity and temperature were also monitored (Omega HX94BC transmitter and Type K thermocouple, OMEGA Engineering, U.S.A.). A dynamic control system (LabVIEW[™], v15.0 32-bit, National Instruments) was used to automate the process of real-time averaging and dilution of pollution levels.

The exposure chamber consisted of a main exposure room (internal volume: 2.7 m height x 3.5 m width x 2.8 m length) and an airlock/anteroom (2.7 x 3.5 x 1.2 m). Up to four participants could be in the chamber at the same time. Participant were asked to remain seated at assigned desks and avoid watching suspenseful videos, talking to each other, or talking on a cell phone for the duration of the exposure period; however, activities within the facility were not monitored or restricted (participants were allowed to use computers/internet, read books, listen to music, nap, etc.). Participants wore headphones while inside the exposure chamber, which reduced the noise generated by the exposure delivery system and allowed the study nurse to communicate with them from outside the chamber. Participants' blood pressure, heart rate, and oxygen saturation levels were measured and recorded by a registered nurse every 15 minutes during the exposure period for safety purposes.

Health and Covariable Measurements

A series of health measurements were initiated at four time points during each study session: baseline (pre-exposure), immediately post-exposure, three hours post-exposure, and 24 hours post-exposure (see Appendix C); only blood pressure measurements are described herein. Because participants arrived at the facility at the same time (between approximately 7:30AM and 9AM) and followed the same protocols each study session, health measurements occurred at approximately the same time of day across sessions. Blood pressure measurements

were the second health measurement in the series and occurred approximately 20 to 30 minutes after the start of the series (see Results).

Brachial blood pressure was measured on the left upper arm with participants in a supine position after a minimum 10-minute rest period using automated oscillatory monitor (SphygmoCor XCEL, AtCor Medical Pty Ltd., Australia). Three measurements were taken with one minute between each measurement; the last two measurements were averaged.

Questionnaires were administered at each time point to assess other covariables across study sessions, such as the participant's mode of commute to our facility, consumption of alcohol, caffeine, and medication, and exposures to smoke and/or fumes. Hourly ambient PM_{2.5} and CO data for the 24 hours prior to and throughout each study session were collected from a local county monitoring site via the U.S. EPA's Air Quality Data API (https://aqs.epa.gov/api). Temperature data was obtained from Colorado State University Atmospheric Science Department's Christman Field Weather Station (https://www.atmos.colostate.edu/fccwx/fccwx data form.php).

Statistical Analysis

Linear mixed-effect models were employed to estimate the differences between continuous blood pressure metrics at each post-exposure time point for the stove treatments compared to the control (filtered air) treatment. Separate models were run for each post-exposure time point (immediate, three-hour, 24-hour) and for each blood pressure metric (systolic, diastolic).

The primary models contained a fixed effect of categorical treatment type and a random person intercept to account for non-independence across repeated measures within our crossover design (i.e., each individual completing multiple treatments). We also included a random effect for day of year, to account for within-day correlation for individuals who received treatments on the same day. The baseline (pre-exposure) blood pressure value was included as a covariate in the model to account for differences in individuals' starting blood pressures across

treatment/study sessions (Vickers and Altman 2001). No other covariates were included in the model. The crossover design eliminates the need to control for individual-level confounders (e.g., age, sex, BMI) as each person participates in each treatment. The blinded, Williams-square random-order treatment assignments and restrictions on participants' behaviors makes the design robust to first-order carryover effects external confounders that might vary across study days (e.g., ambient PM_{2.5} conditions, caffeine/alcohol consumption), as the distribution of these variables is expected to be similar across all treatment groups, when data is balanced (Jones and Kenward 2014; Kenward 2015). Descriptive statistics and univariate analyses were conducted to confirm that associations between these covariates and the treatment groups did not occur by chance or due to imbalances caused by missing data.

Additional models were evaluated as alternatives to the main model. We developed a mixed-effect model that considered more structured study design parameters relevant to our Williams square, such as each individual's assigned sequence group and the day of week (Monday vs. Wednesday), and only included data that was collected within the intended sequence (i.e., not including makeup sessions). We also ran the same model as the primary model but (1) on a data set that excluded data collected outside of the intended treatment sequence, and (2) on a data set that excluded data from study sessions where the exposure mean was outside of a narrowed range around the target value (see Appendix C).

Descriptive statistics describing the distributions of the exposure levels for each treatment type were calculated using each individual's specific two-hour average. We calculated the mean of participants' averaged exposure concentrations by calculating a two-hour average for each individual and then averaging across all individuals. We calculated the mean 5th and 95th percentile of exposure by determining each individual's 5th and 95th percentile for a given treatment and then averaging across all individuals. Data processing and statistical analyses were performed in R (version 3.3.1, The R Foundation for Statistical Computing) using RStudio

(Version 1.0.136). We used the Ime4 package for mixed effect models (Bates et al. 2015). Model assumptions were evaluated.

Results

Exposures/Treatments

The mean of participants' averaged exposure concentrations for $PM_{2.5}$ mass were within 10% of the target concentrations for the fan rocket, rocket elbow, and three stone fire treatments, 20% for the LPG treatment (+2 µg/m³), and 30% for the gasifier treatment (+16 µg/m³); however, there was a range of individual two-hour averages within each treatment (see Table 5.1). Evaluation of the standard deviations for each participant's two-hour average and the 5th and 95th percentile of one-second data for each participant showed little overlap between treatments; though, short-duration deviations from the target value occurred throughout the exposure period (see Table 5.1).

The mean of participants' averaged CO exposures per treatment type generally increased with increasing PM_{2.5}, ranging from 2 ppm for the control filtered air exposure up to 9 ppm for the three stone fire.

Study Population

Our study population consisted of 48 participants, including 26 males and 22 females (Table 5.2). Participants ranged in age from 21 to 36 years (mean \pm SD, 27.5 \pm 3.6 years), were within the normal or low overweight BMI categories (mean \pm SD, 23.4 \pm 2.2 kg/m²), and had non-hypertensive baseline blood pressures. Values were comparable between men and women. Participants predominately identified as non-Hispanic white (42/48 participants; 88%).

The total missing data rate was 6% (see Appendix C). Of the 48 participants, 22 (46%) completed the study in the intended, assigned order with no missed study sessions (treatments). Using make-up dates at the end of a study round to complete missed treatments outside of the intended order, the majority of participants contributed data relevant to all six stove treatments (38/48 participants; 79%); 94% (45/48) had data for at least five treatments.

	Fuel	Participants	PM _{2.5} (μg/m ³)					CO (ppm)	
Treatment ¹		Who Completed Treatment (n)	Mean [SD] ²	Min, Max Individual Exposure ²	Standard Deviation Mean [SD] ³	5 th Percentile Mean [SD] ³	95 th Percentile Mean [SD] ³	Mean [SD]²	Min, Max Individual Exposure ²
Control	None	47	1 [2]	-1 ⁴ , 9	1 [1]	0 [2]	2 [3]	2 [2]	1, 10
LPG	Propane	44	8 [3]	3, 13	5 [3]	3 [3]	18 [8]	3 [1]	1, 6
Gasifier	Wood chips	44	46 [9]	30, 76	10 [10]	33 [12]	63 [24]	5 [3]	1, 14
Fan rocket	Wood sticks	44	95 [9]	77, 111	16 [6]	72 [11]	122 [22]	8 [2]	5, 12
Rocket elbow	Wood sticks	47	254 [9]	236, 276	28 [9]	209 [17]	297 [20]	6 [2]	3, 11
Three stone fire	Wood sticks	47	463 [41]	367, 531	63 [19]	349 [68]	554 [46]	9 [4]	4, 20

 Table 5.1 Distributions of the Individual Mean Two-Hour Pollutant Exposures Measured during Treatments among 48 participants.

¹Target PM_{2.5} levels for each treatment were: HEPA-filtered air (0 µg/m³ PM_{2.5} target level), LPG stove (10 µg/m³), gasifier stove (35 µg/m³), fan rocket stove (10 µg/m³), rocket elbow stove (250 µg/m³), and three stone fire (500 µg/m³). CO did not have a target level.

²Measured pollutant mean is of the participants' two-hour average values, calculated by determining the two-hour average of the one-second exposure data for each participant and then averaging across all participants for each treatment. Standard Deviation (SD) is the standard deviation for this mean. Min and max individual values are the lowest and highest two-hour average value measured for a single participant.

³Standard Deviation Mean is the average of the standard deviations calculated for each participant's two-hour average exposure window; Mean 5th and 95th percentiles are the average of the 5th and 95th percentile values of one-second data for each participant's two-hour average exposure window. ⁴Negative values are a result of a DustTrak calibration artifact.

SD = standard deviation; LPG = liquefied petroleum gas; PM_{2.5} = fine particulate matter mass less than 2.5 µm in diameter, CO = carbon monoxide

Variable (units)	Statistic	Total (n=48)	Female (n=22)	Male (n=26)	
BMI (kg/m²)	Mean [SD]⁵	23.4 [2.2]	23.5 [2.6]	23.3 [2.0]	
	min, max	19.4, 28.7	19.7, 28.7	19.4, 26.0	
Age (years)	Mean [SD]b27.5 [3.6]27.5 [3.4]min, max20.5, 36.122.8, 34.0		27.5 [3.4] 22.8, 34.0	27.4 [3.9] 20.5, 36.1	
Baseline SBP	Mean [SD]⁵	116 [9]	113 [9]	118 [8]	
(mmHg)	min, max	99, 135	100, 135	99, 135	
Baseline DBP	Mean [SD]⁵	69 [6]	69 [7]	69 [5]	
(mmHg)	min, max	59, 86	60, 86	59, 80	
Participants with data for all treatments ^a	Percent	79	82	77	

Table 5.2 Description of Study Participants.^a

^aParticipant was counted if they had data for baseline measurement and at least one post-exposure follow-up time point.

^bSD = standard deviation. Mean calculated as the population mean of each individuals' average baseline health measurement across their completed study sessions.

BMI = body mass index; SBP = systolic blood pressure; DBP = diastolic blood pressure

Differences in Health Outcomes for Stove Treatments Compared to Control

Due to the study day protocols and ordering of all health measurements conducted, the average timing of blood pressure measurements were 30 minutes post exposure for the immediate time point, 3 hours and 26 minutes for the three hour time point, and 24 hours and 13 minutes post-exposure for the 24 hour time point (see Appendix C). Mean systolic and diastolic blood pressures at each time point and treatment are presented in Table 5.3. Mean blood pressures were within normal, non-hypertensive ranges (less than 130 systolic and/or 80 diastolic) across all measurements (average across all measurements and time points: 115.7 mmHg systolic / 68.9 mmHg diastolic; percent of measurements with systolic pressure \geq 80 mmHg: 9%; percent of measurements with diastolic pressure \geq 80 mmHg: 8%).

The reported use of alcohol, caffeine, and medication were low throughout the study (see Appendix C). Univariate analyses indicated no associations between these and other covariates (e.g., ambient PM_{2.5} and CO, mode of commute to our facility) across the various treatments (see
Appendix C). As such, no additional covariates were included as potential confounders in models assessing the association of treatments with blood pressure outcomes.

Effect estimates and 95% confidence intervals for the difference in blood pressure post-exposure for each stove treatment compared to the filtered-air control treatment are presented in Table 5.3 and Figure 5.1. Results that compare to the three stone fire as the reference are provided in Appendix C. Secondary model results were consistent with the main model (see Appendix C).

At the immediate post-exposure measurement, systolic pressure was significantly lower compared to the filtered air control for the three stone fire treatment (500 μ g/m³ PM_{2.5}; -2.29 mmHg, 95% CI -4.48, -0.10) and suggestively lower for the gasifier (35 μ g/m³ PM_{2.5}; -1.81 mmHg, 95% CI -4.03, 0.41). There were no differences for all other treatments at this time point (Table 5.3; Figure 5.1).

No significant associations were found between systolic pressure and treatments at three hours post exposure (Table 5.3; Figure 5.1). However, effect estimates were suggestive of lower systolic pressure for the treatment than the filtered air control for the fan rocket (100 μ g/m³ PM_{2.5}; -1.76 mmHg, 95% CI -4.02, 0.50) and three stone fire (500 μ g/m³ PM_{2.5}; -2.05, 95% CI -4.25, 0.15) treatments. Effect estimates were suggestive of higher pressure for the treatment than the control for the LPG (10 μ g/m³ PM_{2.5}; 1.10 mmHg, 95% CI -1.13, 3.33) and gasifier (35 μ g/m³ PM_{2.5}; 0.99 mmHg, 95% CI -1.24, 3.23) treatments.

At 24-hours post-exposure, systolic blood pressure was significantly increased by 2 to 3 mmHg compared to the control for all treatments except the rocket elbow (LPG: 3.11 mmHg, 95% CI 0.65, 5.27; gasifier: 2.3 mmHg, 95% CI 0.11, 4.48; fan rocket, 2.54 mmHg, 95% CI 0.39, 4.70; three stone fire: 2.41, 95% CI 0.28, 4.53) (Table 5.3; Figure 5.1). No exposure-response with increasing target PM_{2.5} across cookstove treatments was apparent. Associations were consistent with the null for diastolic pressure at every time point for all stove treatments compared to the control.

 Table 5.3 Mean Difference in Blood Pressure for Stove Treatments Compared to Control at Each Measurement Time.

	Baseline ^a	Effect Estimate (95% CI) [mmHg difference compared to					
Treatment	value mmHg	control treatment]					
		Immediate ^a	3-hour ^a	24-hour ^a			
		post-exposure	ost-exposure post-exposure				
Systolic Pressure							
LPG	116.5 (10.7)	-0.22 (-2.45, 2.01)	1.10 (-1.13, 3.33)	3.11 (0.95, 5.27)			
gasifier	115.7 (10.8)	-1.81 (-4.03, 0.41)	0.99 (-1.24, 3.23)	2.30 (0.11, 4.48)			
fan rocket	115.0 (9.2)	-0.44 (-2.67, 1.78)	-1.76 (-4.02, 0.50)	2.54 (0.39, 4.70)			
rocket elbow	115.6 (9.7)	-0.58 (-2.78, 1.63)	-0.47 (-2.68, 1.73)	-0.07 (-2.21, 2.07)			
three stone fire	117.0 (11.3)	-2.29 (-4.48, -0.10)	-2.05 (-4.25, 0.15)	2.41 (0.28, 4.53)			
Diastolic Pressure							
LPG	69.2 (6.7)	-0.67 (-2.16, 0.83)	-0.01 (-1.74, 1.71)	0.32 (-1.55, 2.20)			
gasifier	69.1 (6.9)	-0.75 (-2.24, 0.74)	0.25 (-1.48, 1.99)	-0.37 (-2.27, 1.53)			
fan rocket	68.2 (7.3)	-0.13 (-1.63, 1.36)	-0.41 (-2.15, 1.34)	-0.06 (-1.94, 1.83)			
rocket elbow	69.1 (7.3)	0.35 (-1.13, 1.83)	0.23 (-1.48, 1.94)	-1.71 (-3.58, 0.15)			
three stone fire	70.2 (7.6)	-0.87 (-2.34, 0.60)	-0.77 (-2.48, 0.93)	0.81 (-1.04, 2.67)			

All estimates are adjusted for baseline (pre-exposure) blood pressure.

^aBaseline pre-exposure measurements occurred on average 25 minutes before entering the exposure facility (range 66 to 12 min). The immediate post-exposure measurements occurred on average 30 minutes (range 16 to 49 min) after exiting the facility. The average time of the three hour post-exposure measurements was 3 hr 26 min (range 3 hr 12 min to 3 hr 47 min) after exiting the exposure facility, and the 24 hour measurements were 24 hr 14 min (range 22 hr 13 min to 25 hr 44 min) after exiting the facility.

Control value at baseline: systolic: 115.2 (9.6) mmHg; diastolic: 68.6 (6.6).

SD = standard deviation; CI = confidence interval; LPG = liquid petroleum gas; mmHg = millimeters mercury



Figure 5.1 Effect Estimates and Confidence Intervals for Difference in Blood Pressure for Stove Treatment Compared to Control, by Stove Type and Post-Exposure Time Point. Top panel: systolic pressure; Bottom panel: diastolic pressure.

Discussion

Overall Summary

Household air pollution exposure from cooking and heating using solid fuels is a leading contributor to cardiovascular disease worldwide, yet there are many gaps in our understanding of how different stove types and exposure levels contribute to health effects and whether stoves with decreased emissions compared to traditional three stone fires may be beneficial for health. In the present study, we observed evidence that short-term (two hour) exposures to emissions from cookstoves resulted in a 2 to 3 mmHg increase in systolic blood pressure compared to filtered air control at 24 hours post-exposure. Conversely, immediately post-exposure, we observed a trend of small, non-significant decreases in systolic blood pressure as compared to control that seemed to return to null three hours post-exposure (and for some treatment types, moved towards small [1 mmHg] increases in blood pressure). These changes were seen across a range of stove types representing varying PM_{2.5} exposure levels from 10 to 500 μ g/m³ and did not appear to follow an exposure-response pattern that corresponded with the PM_{2.5} or CO concentrations. No differences in diastolic pressure for stove treatments compared to control filtered air were observed at any time point.

Comparison to Other Literature - Cookstove Studies

Several field studies have demonstrated associations between blood pressure (primarily systolic pressure) and cookstove-generated $PM_{2.5}$ exposures in women (Alexander et al. 2015; Alexander et al. 2017; Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018; Burroughs Pena et al. 2015; Clark et al. 2011; Clark et al. 2013a; Dutta and Ray 2012; McCracken et al. 2007; Neupane et al. 2015; Norris et al. 2016). McCracken et al. (2007) found that use of an improved stove with a chimney was associated with lower systolic (3.7 mmHg, CI -8.1, 0.60) and diastolic (3.0 mmHg, CI -5.7, -0.4) pressure post-intervention among older Guatemalan women (average age 53.3 ± 12.0 years). Alexander et al. (2017) found that diastolic pressure at 38 weeks gestation was 2.8 mmHg lower on average among pregnant Nigerian

women who switched to ethanol fuels at the beginning of their pregnancy compared to controls who continued to cook with either kerosene or wood; though, no differences were seen in systolic pressure between the intervention and control groups. Field studies may be limited in their interpretation in that most have been cross-sectional in design, only measured blood pressure on one or two occasions per participant, and defined exposure based on a categorical stove type or if exposure measurements were included, only conducted a limited number of short duration sampling periods (e.g., Baumgartner et al. 2011; Baumgartner et al. 2014; Baumgartner et al. 2018; Burroughs Pena et al. 2015; Clark et al. 2011; Clark et al. 2013a; Neupane et al. 2015). Thus, these studies may be subject to confounding and measurement error (in both health and exposure metrics) that can reduce precision and bias results. Further, while these studies provide support for an association between exposure to household air pollution and blood pressure under real-world conditions, they are limited in their ability to provide information across a broad range of exposure levels, stove types, and populations, which is important for designing public health interventions or policies aiming to reduce cookstove-associated exposures.

<u>Comparison to Other Literature – Controlled Air Pollution Exposures</u>

Few controlled exposure studies exist on wood smoke exposures; investigation of blood pressure in these studies is further limited (e.g., Bonlokke et al. 2014; Forchhammer et al. 2012; Ghio et al. 2012; Riddervold et al. 2011; Riddervold et al. 2012; Stockfelt et al. 2012; Stockfelt et al. 2013). In line with our findings, Unosson et al. (2013) found no changes in systolic or diastolic pressure during the one hour after a three-hour exposure to wood smoke (300 µg/m³ PM_{2.5}) compared to filtered air exposure; similarly Evans et al. (2015) reported no immediate effects on systolic pressure following 20-minute exposure sessions to environmental tobacco smoke, cooking oil fumes, and wood smoke (peak concentration target 350 µg/m³) compared to a water vapor sham control. However, both studies saw other acute hemodynamic responses (e.g., changes in central arterial stiffness, heart rate variability indexes) suggestive of vascular impairment, autonomic nervous system perturbations, and/or baroreflex activity occurring on an

acute time frame as a result of wood smoke exposure. Neither study included a follow-up measure at a later time point (e.g., 24 hours). Hunter et al. (2014) found no changes in systolic or diastolic pressure among firefighters during a one-hour controlled exposure to wood smoke (1,000 μ g/m³) or at follow-ups six and 24 hours after exposure ended.

Several controlled exposure studies of ambient air pollution sources have demonstrated that blood pressure can increase immediately (within minutes) by 2 mmHg or greater during exposure to concentrated ambient particles in the 100 to 200 μ g/m³ PM_{2.5} range, however these studies suggest that effects may subside within minutes to a few hours after exposure stops (Brook et al. 2002; Brook et al. 2009; Byrd et al. 2016; Urch et al. 2005). Only one of the identified ambient controlled exposure studies maintained follow-up through 24 hours, however researchers did not observe an effect at this time (Brook et al. 2009). It is possible that our study did not see any effects on blood pressure at the immediate or three hour post exposure time points because perturbations that may have occurred during or throughout the exposure window, which are likely to be through an immediate autonomic nervous system activation pathway, had subsided by the time these measurements were conducted. Unlike our findings, several of these studies saw greater effects on diastolic than systolic pressure (Brook et al. 2009; Urch et al. 2005). Numerous observational studies of ambient air pollution indicate that PM_{2.5} levels are associated with elevated blood pressure on lags of one to five days (Brook and Rajagopalan 2009). For example, Brook et al. (2011) found that 24 hour personal PM_{2.5} exposures among Michigan residents were associated with increased systolic blood pressure one day later (1.41 mmHg per 10 µg/m³ PM_{2.5} increase).

Biologic Pathways of Cardiovascular Responses from Air Pollution Exposure

Studying acute responses in health markers known to be part of the pathway of cardiovascular disease development (Brook et al. 2010) is an innovative approach to providing insight into the potential for clinically-relevant differences that would manifest following long-term exposures. Increases in blood pressure, even within a normal range, can lead to increased risk

of stroke, coronary heart disease, and heart failure (Ettehad et al. 2016); on a population level, even a small increase in blood pressure can push a segment of the population from a pre-hypertensive to hypertensive state and considerably increase risks of cardiovascular outcomes such as stroke and ischemic heart disease mortality (Lewington et al. 2002). General consensus indicates three main biological pathways through which air pollution - specifically PM - can trigger systemic vascular dysfunction and lead increases in blood pressure and subsequent cardiovascular events: pro-inflammatory responses leading to oxidative stress and inflammation, activation of the autonomic nervous system and increased parasympathetic responses, and direct interaction of particles with molecules in blood circulation that regulate endothelial function and cell signaling (Brook et al. 2010). Each of these pathways can potentially result in vasoconstriction and endothelial dysfunction, which can increase blood pressure. Our results, which indicate a 24-hour response of elevated blood pressure following PM_{2.5} exposure, are aligned with a pro-inflammatory/stress activation pathway leading to a delayed hypertensive response, as opposed to autonomic nervous system activation, which is more immediate (Langrish et al. 2012). Further work to investigate circulating inflammatory markers would help confirm this hypothesis.

Exposure Assessment: PM_{2.5} vs Other Pollutants

Our facility monitored PM_{2.5} concentrations in real time and maintained rolling averages across the two-hour exposure window by adjusting the ratio of filtered dilution air added. As a result, we achieved mean PM_{2.5} levels for each person-session that were close to the target values for each treatment, with little overlap between treatments. Exposures were not held constant at the target level throughout the duration of the two-hour exposures as cookstove combustion is variable. However, analyses of the 5th and 95th percentiles of the two-hour windows demonstrated that even short-duration peaks or dips in the exposure levels remained reasonably separable from one stove treatment to the next.

Air pollution generated from cookstoves is known to be a complex mixture, including particulate matter of varying size distributions (e.g., ultrafine number counts) and compositions (e.g., differing breakdowns of polycyclic aromatic hydrocarbon content, elemental vs organic ratios) and complex gas mixtures (e.g., volatile organic compounds like benzene, carbonyls like formaldehyde and acetaldehyde). However, we only monitored PM_{2.5} and CO levels within the exposure chamber and did not consider all health-relevant pollutants which could impact relationships seen across the various stove treatments. While previous studies indicate that PM_{2.5} is the likely driving agent behind blood pressure responses from air pollutant or combination of pollutants could be impacting the blood pressure responses seen in this study. The lack of a PM-mass exposure-response across the various stove treatments, which could be reasonably expected given that the treatments span a large window of PM_{2.5} exposure levels, suggests this may be the case. However, it is alternatively possible that the exposure-response seen for health outcomes from longer-term exposures to diverse air pollution sources may not translate to an acute exposure-response for cookstove air pollution and blood pressure.

Additional Strengths and Limitations

Our study population was young, predominantly white, healthy individuals with limited air pollution exposures outside of the study; therefore, the generalizability of results to cookstove users may be limited. This population was feasible to study in this context in terms of participant safety and allowed us to minimize confounder or interactions by age, co-morbid disease status, or other pollution exposures. Our study has strong internal validity accomplished by the controlled exposure design, which strengthens the study's ability to balance data gaps of potentially more generalizable, but less internally valid, observational studies.

We attempted to blind participants to the treatments they were receiving by not informing them of the treatment for each of their study days. However, full blinding was not feasible as higher-PM treatment levels (e.g., fan-rocket, rocket elbow, three stone fire) have a distinct wood

smoke smell that participants could identify when they entered the exposure chamber. Additionally, staff administering the clinical measurements were aware of the treatment for each study day. It is possible that the lack of complete blinding may have contributed to increased stress reactions on stove treatment days compared to the control days (particularly for higher-PM level treatments). Because stress is associated with changes in blood pressure, this could have resulted in some effect of treatment on blood pressure mediated through a stress pathway; however, similar patterns for systolic blood pressure were also observed for the LPG treatment compared to control which would not have been impacted by the lack of participant blinding.

This was the largest air pollution controlled exposure study conducted to date and the first, to our knowledge, that considered exposures to residential cookstove-generated air pollution. Similar studies conducted previously generally incorporated only one or two exposure treatments, had small sample sizes (10 to 20 individuals), and focused on air pollution generated from ambient particles or modern style wood-burning stoves (e.g., Barregard et al. 2008; Bonlokke et al. 2014; Brook et al. 2002; Brook et al. 2009; Byrd et al. 2016; Forchhammer et al. 2012; Langrish et al. 2014; Mills et al. 2005; Muala et al. 2015; Riddervold et al. 2011; Riddervold et al. 2012; Sallsten et al. 2006; Stockfelt et al. 2012; Unosson et al. 2013; Urch et al. 2005). Our study expands on these designs by incorporating more exposure levels allowing for assessment of incremental health impacts, including more subjects to allow for higher power, and generating treatment exposures from a variety of cookstove types. The Williams square crossover design allowed for within-person comparisons and, combined with study protocols that restricted participant behavior, eliminated many potential confounders to allow for efficient analyses comparing more stove types and exposure levels than is possible in observational designs. Additionally, four of the five cookstove treatments administered in this study were generated by wood-burning stoves, allowing results to translate to wood smoke exposures from residential heating stoves used in the United States and Europe as well as other wood smoke exposures such as wildfires.

Conclusions

We demonstrated that short-term exposures to cookstove-generated air pollution acutely perturbs blood pressure, eliciting a small, non-significant decrease in systolic pressure immediately post-exposure and a 2 to 3 mmHg increase in systolic pressure at 24 hours post-exposure. Responses were consistent across a range of stove treatment types with $PM_{2.5}$ levels ranging from 10 to 500 µg/m³, which suggests that household air pollution may be detrimental to cardiovascular health even at low $PM_{2.5}$ levels. Further work is needed to better characterize the multipollutant exposures and consider how the acute exposure-response relationships seen in this controlled study translate to real-world scenarios of chronic exposure.

CHAPTER 6: EFFECTS ON LUNG FUNCTION FOLLOWING CONTROLLED EXPOSURE TO AIR POLLUTION EMITTED FROM COOKSTOVES IN THE SUBCLINICAL TESTS OF VOLUNTEERS EXPOSED TO SMOKE (STOVES) STUDY

Summary

Exposure to household air pollution generated by the use of cookstoves is a leading contributor to global disease, yet uncertainties exist regarding the expected cardiopulmonary responses across different stove types. We conducted a controlled human exposure study to investigate lung function responses in volunteers following exposure to cookstove-generated air pollution. Forty-seven healthy adult volunteers underwent two-hour exposures to six different treatments: filtered air (fine particulate matter mass $[PM_{2.5}]$ target 0 μ g/m³), liquid petroleum gas stove (10 μ g/m³), gasifier stove (35 μ g/m³), fan rocket stove (100 μ g/m³), rocket elbow stove $(250 \ \mu g/m^3)$, and three stone fire $(500 \ \mu g/m^3)$. Health measurements were conducted each morning prior to the exposure, immediately after exposure, and at three and 24 hours after exposure. Immediately after exposures, forced vital capacity (FVC), forced expiratory volume in one second (FEV₁), and mid-expiratory flow (FEF₂₅₋₇₅) were slightly lower among the three highest PM_{2.5} stove treatments compared to the control; no differences in FEV₁/FVC were observed. The largest mean differences in all three metrics were for the fan rocket stove (FVC: -60 mL, 95% CI -135, 15; FEV1: -51 mL, 95% CI -117, 16; FEF25-75: -116 mL/s, 95% CI -239, 8). At three hours post-exposure, decreases were observed for all five stove treatments compared to the control for FVC, FEV₁, and FEF₂₅₋₇₅; effect estimates were largest for the LPG stove (FVC: -39 mL, 95% CI -114, 35; FEV1: -68 mL, 95% CI -128, -7; FEF25-75: -122 mL/s, 95% CI -255, 11). At 24 hours post-exposure, results were consistent with a null association for FVC, FEV₁, and FEV₁/FVC; FEF₂₅₋₇₅ was decreased compared to control for the gasifier, fan rocket, and three stone fire treatments but not the LPG or rocket elbow (LPG: 39 mL/s, 95% CI -278, 156; gasifier: -63 mL/s, 95% CI -182, 56; fan rocket: -81 mL/s, 95% CI -199, 37; rocket elbow: 35 mL/s, 95% CI -83, 153; three stone fire: -88 mL /s, 95% CI -204, 27). These results suggest that lung function is reduced

by exposure to cookstove emissions even at low PM_{2.5} exposure levels, potentially through an inflammatory pathway.

Introduction

Nearly 40% of the world's population cooks over open fires or with rudimentary solid fuel-burning stoves (Bonjour et al. 2013). This practice generates household air pollution, which is responsible for a high burden of disease globally: over 77 million disability-adjusted life years (DALYs), including 2.5 million deaths, in 2016 (Gakidou et al. 2017). The combination of lower respiratory infections, respiratory cancers, and chronic obstructive pulmonary disease were estimated to account for over 46 million of these DALYs, including 1.36 million deaths (Gakidou et al. 2017).

Approaches to reduce exposures and diseases from household air pollution have included switching to cookstoves designed to emit lower levels of fine particulate matter mass (PM_{2.5}) and/or carbon monoxide (CO). Although some research has demonstrated the potential for improved stoves to reduce human exposures (Thomas et al. 2015) and associated cardiovascular and pulmonary health effects (e.g., Alexander et al. 2017; Clark et al. 2013a; da Silva et al. 2012; Fullerton et al. 2011; McCracken et al. 2011; McCracken et al. 2007), other studies have not demonstrated the expected health benefits following dissemination of improved stoves (e.g., Clark et al. 2009; Mortimer et al. 2017; Romieu et al. 2009; Smith-Sivertsen et al. 2009; Smith et al. 2011). Gaps in our understanding of the health responses make it difficult to predict whether an improved stove design will result in a health-relevant reduction in exposure. Field observational and intervention studies in communities of cookstove users are often limited to investigating only a single stove technology compared to traditional practices. Field studies can be further hindered by a lack of adherence to intervention protocols and confounding by factors such as other air pollution exposures, diet, or co-morbid disease status. Interpretation of the body of literature is hindered by a number of factors, such as disconnects between laboratory and real-world emissions (Roden et al. 2009), barriers to adoption and sustained use during stove interventions

(Ruiz-Mercado et al. 2011), and challenges to conducting accurate exposure assessment (Clark et al. 2013b). Controlled exposure studies can address some of these challenges, through strictly controlled protocols that limit confounding, well defined exposures, and the ability to compare health effects across multiple treatments under similar conditions.

Reductions in lung function signify impairment of ventilation and respiratory mechanical function, which could be due to inflammation or tissue injury (U.S. EPA 2009). Spirometry is a useful tool for understanding general pulmonary function and diagnosing respiratory diseases (Miller et al. 2005). For example, spirometry is an effective means of differentiating restrictive and obstructive lung function impairments (Dempsey and Scanlon 2018; Lange et al. 2009). While spirometry is regularly used in clinical settings for disease diagnosis and management, it has also been employed in epidemiological studies as an indicator of respiratory health.

The objective of this work was to investigate acute changes in markers of respiratory health following controlled exposure to cookstove-generated air pollution. We measured lung function in healthy adult subjects at multiple follow-up times over 24 hours after two hours of controlled exposure to air pollution from five different cookstoves (ranging from 10 to 500 μ g/m³) and compared to filtered air exposure.

Methods

Study protocols were approved by the Colorado State University Institutional Review Board. All participants provided written informed consent. The study design, exposure facility, and study session protocols are described below and in more detail elsewhere (see Chapter 5 and Appendices C and D).

Study Design

We exposed participants to six different treatments, each lasting two hours: filtered air $(0 \ \mu g/m^3 \ PM_{2.5}$ target level), LPG stove $(10 \ \mu g/m^3)$, gasifier stove $(35 \ \mu g/m^3)$, fan rocket stove $(100 \ \mu g/m^3)$, rocket elbow stove $(250 \ \mu g/m^3)$, and three stone fire $(500 \ \mu g/m^3)$. The stoves are described in Appendix D.

Treatment order was determined using a Williams square, which is a Latin square crossover design that is balanced across treatments and first-order carry-over effects (Williams 1949). Under this design, we designated six sequences (treatment orders) with eight participants per sequence. Each treatment occurred one time per sequence. Across all sequences, each treatment occurred exactly once in each period (e.g., the first treatment in the sequence will not be the first treatment in any other sequence and so on) and was preceeded and followed by each other treatment exactly once. We conducted the study in three rounds (October 2016 to February 2017; March to June 2017; August 2017 to January 2018); within each round, two sequence groups (n = 16) alternated weeks until completion of all six treatments.

Participants within the same sequence (n = 8) completed their study sessions in groups of four per day, staggered at 30 minute intervals. To reduce effects due to diurnal variability or day-of-week patterns, we scheduled participants to complete each study session in the same order, on the same weekday (Monday or Wednesday). We scheduled sessions within the sequence with a minimum two-week washout period between visits (maximum six weeks due to holidays, though typically just two or three weeks). Participants were scheduled to complete all six treatments within 13 to 16 weeks. Missed treatments were made up at the end of the sequence, ten days to 14 weeks after the last regularly-scheduled session and not necessarily on the same day of the week or starting at the same time of morning. More details on the administration of the Williams square design are provided in Appendix D.

We administered whole-body exposures within a 26 m³ controlled-environment chamber. The target PM_{2.5} concentration was attained prior to participants entering the chamber. The PM_{2.5} concentration was controlled by periodically injecting pre-diluted emissions into the high-efficiency particulate air (HEPA) filtered air exchange system, which maintained a constant airflow through the chamber. The injection time was actively controlled to maintain PM_{2.5} levels at the target concentrations via a feedback loop whereby PM_{2.5} levels in the chamber were monitored using a calibrated nephelometer (Dustrak DRX 8533, TSI Incorporated, USA). Temperature, humidity,

oxygen, and CO were also monitored continuously during the treatments (Siemens Ultramat 6E, Siemens AG, Germany; Omega HX94BC transmitter and Type K thermocouple, OMEGA Engineering, U.S.A.). We instructed participants to sit quietly at individual desks within the chamber during each treatment. A registered nurse monitored participants' heart rate, blood pressure, and oxygen saturation levels every 15 minutes during the two-hour treatments.

Study Population

We recruited 48 young, healthy volunteers in Fort Collins, Colorado. Prior to enrollment, participants underwent a physical examination with the study physician that included review of personal and family medical history, an electrocardiogram, spirometry, height and weight measurements, and a standard blood panel, to ensure that the participant met health-based eligibility criteria. Full eligibility criteria were: 18 to 35 years old at the time of recruitment; never smokers; body mass index between 19 to 28 kg/m² with body weight greater than 110 pounds; no history of heart disease (including normal electrocardiogram and non-hypertensive blood pressure as determined during the screening exam), diabetes, kidney disease, systemic sclerosis, or any chronic inflammatory disease such as asthma, arthritis, or severe allergies; normal blood test results, including no evidence of iron-deficient anemia (as determined by blood results from the screening exam); spirometry values greater than 70% of the predicted value for the age/gender during the screening exam; not currently taking statins, anti-inflammatory medication, or other medications unless cleared by the study physician during the screening exam (oral contraceptives and some daily anti-depressant/anti-anxiety medications were approved for continued use); no use of tetrahydrocannabinol (marijuana) or illicit drugs within the past three months; no ear or abdominal/thoracic surgery in the past month; no current cancer or in remission for less than six months; do not have a central intravenous line or port; have never had a mastectomy; do not have a pacemaker; not currently pregnant, breastfeeding, or planning a pregnancy within six months; not regularly exposed to smoke, dust, fumes, or solvents (occupationally or recreationally/at home), or regularly burned candles or incense, within the last three months; no history of claustrophobia; no fear of needles; not planning to donate blood during the timeframe of participation; not allergic to latex; and live within 20 miles of the study facility and not planning to move more than 20 miles away within six months.

Participants were asked to abstain from alcohol, caffeine, smoke exposures, and strenuous exercise starting 24 hours prior to each study session through the end of the 24-hour post-exposure time point. They also were asked to abstain from over-the-counter medications starting 72 hours prior to each study session. Surveys were administered to determine compliance with these protocols (see "Health and Covariate Measurements"). On each study session, we performed a series of health measurements prior to the exposure treatment ("baseline"), immediately post exposure, and at three and 24 hours post-exposure (see Appendix D); only spirometry measurements are described herein. Participants remained on site from the baseline measurements through the end of the three-hour measurements and returned for 24-hour post-exposure health measurements on the following day. We provided a low-fat lunch and snacks during the study day and asked participants to be consistent from one session to the next in their choice of breakfast, dinner, and other snacks throughout the study session when not on-site.

Health and Covariate Measurements

Participants performed a pulmonary function test during each of the four health measurement time points using an ultrasonic spirometer (Easy on-PC, ndd Medizintechnik AG, Zurich, Switzerland). Spirometry measurements were conducted at the end of a series of health measurements not reported here (see Appendix D) and occurred approximately 30 minutes after the start of the series (see Results). Health measurements occurred at approximately the same time of day across sessions. Participants arrived at the facility at the same time (between approximately 7:30AM to 9AM) and followed the same protocols each study session.

Pulmonary function tests were performed according to American Thoracic Society/European Respiratory Society (ATS/ERS) guidelines (Miller et al. 2005). We conducted

a multi-flow calibration of the spirometer daily, at flow rates between 0.5 and 12 L/s using a three-liter syringe. Each test consisted of several trials of an expiratory-only maneuver, performed from a seated position with both feet on the ground, wearing a nose clip. Within each test, we required a minimum of three acceptable trials (e.g., free from artifacts due to cough, glottis closures, or obstructive mouthpieces and with full exhale) with the two largest FEV₁ and FVC values across trials within 150 mL (Miller et al. 2005). We allowed up to eight attempts in a single test. If a participant did not meet the requirement within eight attempts, we stopped the test. The health outcomes used from the pulmonary function test were forced vital capacity (FVC; the total volume of air exhaled in a forceful, complete expiration following maximum inspiration), the forced expiratory volume in one second (FEV₁; the volume of air exhaled in the first second of an FVC maneuver), the ratio of FEV₁ to FVC (FEV₁/FVC) and mid-expiratory flow (FEF₂₅₋₇₅; the mean flow rate between the 25th and 75th percent of the FVC). For all tests that met the quality criteria (Miller et al. 2005), we chose the largest FVC and FEF_{25-75} values from the acceptable trials within that test and the FEV₁ and FEV₁/FVC value that came from the trial with the largest FVC. For tests that did not meet the minimum quality criteria, a board-certified pulmonologist reviewed the spirographs to determine which values, if any, could be used in analyses.

We administered surveys at each health measurement time point to assess other covariates, including participants' recent exposures to medications, caffeine, smoke/fumes, and alcohol, mode of commute to the facility, and sleep duration. Hourly ambient PM_{2.5} and CO data from a local county monitoring site was downloaded via the U.S. EPA's Air Quality Data API (https://aqs.epa.gov/api). Ambient temperature data was obtained from Colorado State University Atmospheric Science Department's Christman Field Weather Station located approximately four miles from the study location (https://www.atmos.colostate.edu/fccwx/fccwx_data_form.php).

Statistical Analysis

We modeled the effect of the stove treatments on lung function at each post-exposure time point using mixed-effect regression models (repeated measures ANCOVA) that included a term for the baseline (pre-exposure) value, to account for differences in pre-exposure lung function across the six sessions (Vickers and Altman 2001). We ran separate models for each lung function metric (FVC, FEV₁, FEV₁/FVC, and FEF₂₅₋₇₅) and each post-exposure time point and used the categorical treatment as the exposure variable. Models included a random person intercept to account for repeated measures among participants and random day intercept to account for non-independence of data for participants of the same group who experienced treatments on the same dates. Because each individual receives all treatments exactly once, time-invariant factors (e.g., participants' age, sex, body mass index) cannot be confounders under this design. Further, the randomization of treatment using a balanced Williams square is designed to be robust to first-order carryover effects and external parameters that might change across study sessions or from one day to the next (e.g., ambient PM and CO, caffeine/alcohol use) as the distributions of these factors will be similar across treatments (Jones and Kenward 2014; Kenward 2015). We also conducted descriptive analyses and ran univariate models to confirm that no associations between these variables and the exposures occurred by chance or due to imbalance as a result of missing data.

Additionally, we ran a mixed-effects model (as an alternative to the previously described main model) that contained additional random effects such as participants' assigned sequence group and day of week. We ran this model using only data from the study sessions completed within the originally-scheduled sequence (i.e., we removed data from makeup sessions). We also ran the main model only using data from sessions completed within sequence. We conducted sensitivity analyses in which we ran the main model but (1) excluded all tests that did not meet the quality criteria but had been approved by the study pulmonologist, and (2) removed study sessions where the exposure average was outside a range of the target level (see Appendix D).

Data analyses were conducted using R (version 3.3.1, The R Foundation for Statistical Computing) and RStudio (Version 1.0.136). We used the Ime4 package for mixed effect models (Bates et al. 2015). Model assumptions were evaluated.

Results

Participants

A total of 269 exposure sessions were administered throughout the course of the study. We recruited 48 participants; one participant was removed from the study due to a decline in lung function between recruitment and baseline measurements on study days (i.e., their lung function during a baseline measurement was below our eligibility criterion). Of the remaining 47 participants (22 male, 25 female), 81% contributed data to all six treatments, either in sequence or using the allotted makeup sessions. Three participants withdrew from the study prior to completing six study sessions. Additionally, an error applying the exposure protocol resulted in the loss of data relevant to single sessions which were not repeated for three participants. Some participants missed individual post-exposure time points (n = 6) and some spirometry data was removed as the pulmonary function test did not meet minimum quality criteria and was not approved for use by the study pulmonologist (n = 9). As a result, the overall missing data rate was 8% (more detail provided in Appendix D).

Consistent with the eligibility criteria, participants were young (average age 27 years, range 21 to 36), with normal BMI (average 23.4 kg/m², range 19.4 to 28.7) and baseline lung function (average FVC of 4.9 L [range 3.1 to 8.1], average FEV₁ of 3.9 L [range 2.9 to 6.1], and average FEF₂₅₋₇₅ of 3.9 L/s [range 2.2 to 6.3]). Participants predominately self-identified as non-Hispanic and white (42/47 participants; 89%). As expected, baseline lung function differed by sex. Study participants are described in Table 6.1.

Variable (units)	Statistic	Total (n=47)	Female (n=22)	Male (n=25)	
BMI (ka/m ²)	Mean [SD]⁵	23.4 [2.3]	23.5 [2.6]	23.3 [2.0]	
Bivii (kg/iii)	min, max	19.4, 28.7	19.7, 28.7	19.4, 26.0	
	Mean [SD]⁵	27.4 [3.6]	27.5 [3.4]	27.4 [3.9]	
Age (years)	min, max	20.5, 36.1	22.8, 34.0	20.5, 36.1	
Number of sessions	Total	269	129	140	
conducted ^a	sessions	200	125		
Participants with data for all	Percent	81	86	76	
six treatments ^a	1 oroont	01	00	10	
Baseline EVC (liters)	Mean ^c [SD] ^b	4.9 [1.1]	4.2 [0.5]	5.5 [1.0]	
Dasenne i VO (inters)	min, max	3.1, 8.1	3.1, 5.4	4.0, 8.1	
Basolino EEV. (litors)	Mean ^c [SD] ^b	3.9 [0.8]	3.4 [0.4]	4.3 [0.7]	
	min, max	2.9, 6.1	2.9, 4.4	3.3, 6.1	
Recoling EEV(/EV(C (ratio)	Mean ^c [SD] ^b	0.8 [0.1]	0.8 [0.1]	0.8 [0.1]	
Daseline FEV1/FVC (Tallo)	min, max	0.6, 1.0	0.7, 1.0	0.6, 0.9	
Bacolino EEE	Mean ^c [SD] ^b	3.9 [1.1]	3.5 [0.9]	4.2 [1.1]	
Dasenne FEF25-75 (IIIerS/S)	min, max	2.2, 6.3	2.2, 5.4	2.2, 6.3	

Table 6.1 Description of Study Participants.^a

^aA participant session was counted if they had data for baseline measurement and at least one post-exposure measurement.

^bSD = standard deviation.

^cMean of each individuals' average baseline health measurement across their completed study sessions.

Treatments/Exposure Levels and Health Measurement Times

The six treatments are described in Table 6.2. The mean $PM_{2.5}$ levels for the two-hour treatments per participant was close to the target levels for each treatment (average difference from target: filtered air control, +1 µg/m³; LPG: -2 µg/m³; gasifier: +11 µg/m³; fan rocket: -5 µg/m³; rocket elbow: +4 µg/m³; three stone fire: -36 µg/m³). The minimum and maximum value for a single participant's two-hour mean maintained separation between treatment types (i.e., an individual's $PM_{2.5}$ concentration means did not overlap between different treatment types). Carbon monoxide levels, which were not controlled but monitored, ranged from a mean of 2 ppm for the control treatment to 9 ppm for the three stone fire and generally increased with increasing $PM_{2.5}$. The number of completed sessions per treatment type was lowest for the gasifier stove (43 participants), followed by fan rocket (44 participants), LPG and rocket elbow (45 each), and then three stone fire and control (46 each).

Due to our study protocols, which involved several other health measurements not reported here, there was a systematic difference in the spirometry measurement time compared to the nominally reported measurement timepoint. Baseline pre-exposure measurements occurred on average 17 minutes before entering the exposure facility (range 3 to 54 min). The immediate post-exposure measurements occurred on average 38 minutes (range 33 to 62 min) after exiting the facility. The average time of the three-hour post-exposure measurements was 3 hr 33 min (range 3 hr 20 min to 3 hr 50 min) after exiting the exposure facility, and the average time of the 24-hour measurements was 24 hr 22 min (range 22 hr 18 min to 25 hr 50 min) after exiting the facility. While participants' start times were staggered, we attempted to keep each individual on the same timeline during each study session to maintain consistency. We calculated the maximum difference in the health measurement timing for each person at each time point across all of their study sessions; the mean maximum difference was 11 minutes for the baseline measurements, 13 minutes for both immediate post-exposure and three-hour post-exposure measurements.

	Fuel	Participants Who Completed Treatment (n)	ΡΜ _{2.5} level (μg/m³)				CO level (ppm)		
Treatment ^a			Mean [SD]⁵	Min, Max Individual Exposure ^ь	Standard Deviation Mean [SD] ^c	5 th Percentile Mean [SD] ^c	95 th Percentile Mean [SD] ^c	Mean [SD]⁰	Min, Max Individual Exposureª
Control	None	46	1 [2]	-1 ^d , 9	1 [1]	0 [2]	2 [3]	2 [2]	1, 10
LPG	Propane	45	8 [3]	3, 13	5 [3]	3 [3]	17 [8]	3 [1]	1,6
Gasifier	Wood chips	43	46 [9]	30, 76	10 [10]	33 [12]	63 [24]	5 [3]	1, 14
Fan rocket	Wood sticks	44	95 [9]	77, 111	16 [6]	72 [11]	122 [21]	8 [2]	5, 12
Rocket elbow	Wood sticks	45	254 [9]	236, 276	28 [9]	209 [17]	297 [20]	6 [2]	3, 11
Three stone fire	Wood sticks	46	464 [39]	367, 531	63 [19]	350 [67]	556 [45]	9 [4]	4, 20

Table 6.2 Distributions of the Individual Mean Two-Hour Pollutant Exposures Measured during Treatments among 47 participants.

^aTarget PM_{2.5} levels for each treatment were: HEPA-filtered air (0 µg/m³ PM_{2.5} target level), LPG stove (10 µg/m³), gasifier stove (35 µg/m³), fan rocket stove (10 µg/m³), rocket elbow stove (250 µg/m³), and three stone fire (500 µg/m³). CO did not have a target level.

^bMeasured pollutant mean is of the participants' two-hour average values, calculated by determining the two-hour average of the one-second exposure data for each participant and then averaging across all participants for each treatment. Standard Deviation (SD) is the standard deviation for this mean. Min and max individual values are the lowest and highest two-hour average value measured for a single participant.

^cStandard Deviation Mean is the average of the standard deviations calculated for each participant's two-hour average exposure window; Mean 5th and 95th percentiles are the average of the 5th and 95th percentile values of one-second data for each participant's two-hour average exposure window. ^dNegative values are a result of a DustTrak calibration artifact.

SD = standard deviation; LPG = liquefied petroleum gas; PM_{2.5} = fine particulate matter mass less than 2.5 µm in diameter, CO = carbon monoxide

Model Results

The effect estimates and 95% confidence intervals (CIs) for the mean difference in each lung function metric at each post-exposure time point for each treatment type compared to the filtered air control are presented in Table 6.3 and Figure 6.1. No potential confounder covariates were included in the models. The descriptive statistics for these variables (medication use, caffeine and alcohol intake, smoke/fume exposure, sleep, mode of commute to facility, ambient PM_{2.5}, ambient CO, ambient temperature) showed consistency across treatments. Univariate analysis did not show any evidence of meaningful associations with the treatment (see Appendix D). Sensitivity analyses and secondary models described in the methods had results consistent with the primary model (see Appendix D).

FVC values at the immediate post-exposure measurements (average 38 minutes after the end of the exposure period) were lower for the three higher-PM_{2.5} treatments than the control, by 40 to 60 mL (fan rocket: -60 mL, 95% CI -135, 15; rocket elbow: -40 mL, 95% CI -114, 35; three stone fire: -42 mL, 95% CI -116, 32). FVC values for the two lower-PM_{2.5} level treatments did not appear different from the control (LPG: 5 mL, 95% CI -69, 80; gasifier: 19 mL, 95% CI -56, 93). At the three-hour post-exposure measurement (average time: 3 hrs 33 minutes after exposure ended), the difference in FVC compared to the control seen immediately post-exposure at the higher PM treatments was diminished, as all stoves showed effect estimates of -39 mL or less (LPG: -39 mL, 95% CI -11, 35; gasifier: -21 mL, 95% CI -95, 54; fan rocket: -30 mL, 95% CI -105, 45; rocket elbow: -8 mL, 95% CI -82, 67; three stone fire: -21, 95% CI -96, 53). The magnitude of the FVC effect showed no discernable pattern with increasing PM_{2.5} levels for the treatments. At 24 hours post exposure, no differences in FVC were observed between the stove treatments and the control.

Similar to FVC, FEV₁ values at the immediate post-exposure time point were suggestive of a reduction in lung function compared to the control for the three higher PM_{2.5}-level treatments (fan rocket: -51 mL, 95% CI -117, 16; rocket elbow: -24 mL, 95% CI -91, 42; three stone fire: -27

mL, 95% CI -93, 39), but not the two lower PM_{2.5} level treatments (LPG: 3 mL, 95% CI -64, 69; gasifier: 7 mL, 95% CI -59, 74). At three hours post exposure, the reduction was more pronounced: all stove treatments had lower FEV₁ values than the control, by 39 to 68 mL. The difference in FEV₁ was largest for the three lower PM_{2.5} level treatments (LPG: -68 mL, 95% CI -128, -7; gasifier: -53 mL, 95% CI -114, 8; fan rocket: -68 mL, 95% CI -129, -7) than for the higher two (rocket elbow: -39 mL, 95% CI -99, 22; three stone fire: -39 mL, 95% CI -99, 21). No effect on FEV₁ was observed at 24 hours post exposure.

FEF₂₅₋₇₅ was consistent with FVC and FEV₁ in that immediately post-exposure, results were suggestive of reductions compared to the control, particularly for the three higher PM_{2.5} level treatments (LPG: -44 mL/s, 95% CI -167, 79; gasifier: -13 mL/s, 95% CI -137, 110; fan rocket: -116 mL/s, 95% CI -239, 8; rocket elbow: -68 mL/s, 95% CI -191, 55; three stone fire: -103 mL/s, 95% CI -225, 19). At three hours post exposure, the estimates of decreased FEF₂₅₋₇₅ for stove treatments compared to the control ranged from -30 mL/s to -122 mL/s (LPG: -122 mL/s, 95% CI -244, 11; gasifier: -74 mL/s, 95% CI -208, 59; fan rocket: -114 mL/s, 95% CI -249, 21; rocket elbow: -56 mL/s, 95% CI -190, 77; three stone fire: -31 mL/s, 95% CI -164, 102). Again similar to FEV₁, the difference in FEF₂₅₋₇₅ was larger for the three lower PM_{2.5} level treatments than the two higher PM_{2.5} level treatments. At 24 hours post-exposure, the gasifier, fan rocket, and three stone fire treatments, but not the LPG or rocket elbow, maintained a lower FEF₂₅₋₇₅ value compared to the control (LPG: 39 mL/s, 95% CI -278, 156; gasifier: -63 mL/s, 95% CI -182, 56; fan rocket: -81 mL/s, 95% CI -199, 37; rocket elbow: 35 mL/s, 95% CI -83, 153; three stone fire: -88 mL/s, 95% CI -204, 27).

No clear patterns of associations were seen in the FEV₁/FVC ratio across stoves at the immediate or 24-hour post exposure measurements, though effect estimates suggest a slightly higher ratio for the three stone fire compared to the control immediately post-exposure (0.5%, 95% CI -0.4, 1.4). At three hours post-exposure, effect estimates indicate small decreases in ratio compared to the control for all treatments other than the three stone fire (LPG: -0.8%, 95%

CI -1.7, 0.2; gasifier: -0.9%, 95% CI -1.9, 0.0; fan rocket: -0.5%, 95% CI -1.4, 0.5; rocket elbow: -0.6%, 95% CI -1.6, 0.3; three stone fire: 0.2%, 95% CI -0.8, 1.1).

Table 6.3 Mean Differences in Lung Function for Each Stove Treatment Compared to Control at Each Measurement Time. Effect estimate is the difference in lung function value for the stove treatment compared to the control at the given post-exposure measurement time, accounting for the baseline (pre-exposure) lung function.

	Bacolino ^a	Effect Estimate (95% confidence interval) as compared					
Treatment	value mean	to control					
neutinent	(SD)	Immediate ^a	3 hours ^a	24 hours ^a			
	(02)	post- exposure	post-exposure	post-exposure			
	FVC (ml)	Difference in FVC (ml)					
LPG	4854 (1024)	5 (-69, 80)	-39 (-114, 35)	9 (-74, 91)			
gasifier	4867 (1043)	19 (-56, 93)	-21 (-95, 54)	12 (-72, 96)			
fan rocket	4879 (1148)	-60 (-135, 15)	-30 (-105, 45)	12 (-72, 95)			
rocket elbow	4898 (1064)	-40 (-114, 35)	-8 (-82, 67)	1 (-83, 84)			
three stone fire	4860 (1070)	-42 (-116, 32)	-21 (-96, 53)	26 (-56, 108)			
	FEV₁ (ml)	Difference in FEV1 (ml)					
LPG	3860 (750)	3 (-64, 69)	-68 (-128, -7)	7 (-62, 76)			
gasifier	3873 (803)	7 (-59, 74)	-53 (-114, 8)	-4 (-74, 66)			
fan rocket	3873 (852)	-51 (-117, 16)	-68 (-129, -7)	-15 (-84, 55)			
rocket elbow	3895 (816)	-24 (-91, 42)	-39 (-99, 22)	0 (-69, 69)			
three stone fire	3887 (793)	-27 (-93, 39)	-39 (-99, 21)	-14 (-82, 54)			
	FEV ₁ /FVC (%)	Difference in FEV ₁ /FVC (%)					
LPG	79.8 (6.4)	0.0 (-0.9, 0.8)	-0.8 (-1.7, 0.2)	0.0 (-0.9, 0.9)			
gasifier	79.4 (5.8)	-0.3 (-1.2, 0.6)	-0.9 (-1.9, 0.0)	0.0 (-1.0, 0.9)			
fan rocket	79.4 (6.7)	-0.1 (-1.0, 0.7)	-0.5 (-1.4, 0.5)	-0.4 (-1.3, 0.5)			
rocket elbow	79.4 (6.4)	-0.2 (-1.0, 0.7)	-0.6 (-1.6, 0.3)	0.2 (-0.8, 1.1)			
three stone fire	79.7 (6.8)	0.5 (-0.4, 1.4)	0.2 (-0.8, 1.1)	-0.2 (-1.1, 0.7)			
	FEF ₂₅₋₇₅ (ml/s)	Difference in FEF ₂₅₋₇₅ (ml/s)					
LPG	3836 (987)	-44 (-167, 79)	-122 (-255, 11)	39 (-78, 156)			
gasifier	3844 (1151)	-13 (-137, 110)	-74 (-208, 59)	-63 (-182, 56)			
fan rocket	3823 (1079)	-116 (-239, 8)	-114 (-249, 21)	-81 (-199, 37)			
rocket elbow	3862 (1148)	-68 (-191, 55)	-56 (-190, 77)	35 (-83, 153)			
three stone fire	3915 (1126)	-103 (-225, 19)	-31 (-164, 102)	-88 (-204, 27)			

All estimates are adjusted for baseline (pre-exposure) values.

^aBaseline pre-exposure measurements occurred on average 17 minutes before entering the exposure facility (range 3 to 54 min). The immediate post-exposure measurements occurred on average 38 minutes (range 33 to 62 min) after exiting the facility. The average time of the three hour post-exposure measurements was 3 hr 33 min (range 3hr 20 min to 3hr 50min) after exiting the exposure facility, and the 24-hour measurements were 24 hr 22 min (range 22hr 18min to 25hr 50min) after exiting the facility.

Control value at baseline [mean(SD)]: FVC: 4875 (1081) mL; FEV₁: 3864 (787) mL; FEV₁/FVC ratio: 79.4 (6.6)%; FEF₂₅₋₇₅: 3832 (1101) mL/s.



Figure 6.1 Effect Estimates and 95% Confidence Intervals for Spirometry Metrics by Stove Type and Post-Exposure Time Point. Effect estimate is the difference in lung function value for the stove treatment compared to the filtered air control at the given post-exposure measurement time, accounting for the baseline (pre-exposure) lung function. Units: FVC = mL, $FEV_1 = mL$, $FEV_1/FVC = \%$, $FEF_{25-75} = mL/s$.

Discussion

This work investigated short-term effects of exposure to cookstove air pollution on lung function. We observed small reductions in FVC, FEV₁, and FEF₂₅₋₇₅ that were suggestive of decreased lung function compared to filtered air exposures at the immediate post-exposure health measurements (on average 38 minutes after the end of the exposure period) and the three-hour post-exposure health measurements (on average 3 hours and 33 minutes after the end of the exposure period), but not at 24 hours post-exposure measurements. Results for a given treatment were consistent across each metric but were generally not statistically significant. No evidence of

an exposure-response was seen and similar effects were observed across all treatments, including the LPG stove at a $PM_{2.5}$ -level of 10 µg/m³. However, the effects were more apparent immediately following exposure for the three treatments at the higher $PM_{2.5}$ levels (fan rocket, rocket elbow, and three stone fire) than the two at the lower $PM_{2.5}$ levels (LPG, gasifier) for all three lung function metrics. At three hours post-exposure, decrements to FEV₁ and FVC were generally larger for the lower-emitting cookstoves (LPG, gasifier, and fan rocket). Differences in FEV₁ and FEF₂₅₋₇₅ were larger at three hours post-exposure than immediately post-exposure; for FVC, effects were largest immediately post-exposure.

These results are different than those of previous controlled exposure studies with wood smoke. Rivvervold et al. (2012) found no differences in the change in FVC, FEV₁, or peak expiratory flow (PEF) from pre-exposure to 3.5 and six hours after exposure among 20 volunteers following three-hour exposures to filtered air control (less than 20 µg/m³ PM), low wood smoke exposure (approximately 200 μ g/m³ PM), or high wood smoke exposure (approximately 400 µg/m³ PM). Sehlstedt et al. (2010) similarly found no changes in FVC or FEV₁ among 19 volunteers following three-hour exposures to filtered air and diluted wood smoke at concentrations of 5 to 15 ppm CO, 0.2 to 0.4 ppm NO_x, and 180 to 300 μ g/m³ PM_{2.5}. Ghio et al. (2012) found no changes in FVC, FEV₁, or PEF among ten volunteers following two-hour exposures to filtered air or wood smoke particles (approximately 500 µg/m³ concentrations). However, several studies have indicated potential for wood smoke air pollution to elicit acute respiratory tract inflammation and irritation, as measured by airway inflammatory markers, exhaled NO, and clinical irritation symptoms (Barregard et al. 2008; Riddervold et al. 2012; Stockfelt et al. 2012), while other studies did not observe these effects (Ghio et al. 2012; Sehlstedt et al. 2010). The difference between previous studies and ours may be attributable to our larger sample size, which gives more power to detect small changes, and our balanced Williams square crossover design and restrictive protocols, which reduced potential confounding.

Observational field studies generally support an association between exposure to cookstove-generated air pollution and reduced lung function, though evidence is inconsistent. Cross-sectional studies in Malawi, Brazil, India, Nigeria, and Mexico have found decreased FVC and FEV₁ among biomass/wood users compared to other fuel types (e.g., kerosene, charcoal, LPG) (da Silva et al. 2012; Desalu et al. 2010; Fullerton et al. 2011; Ibhafidon et al. 2014; Regalado et al. 2006; Revathi et al. 2012). Yet, similar-design studies in Ecuador, Honduras, and Guatemala have found no associations or inverse associations between lung function (FVC and FEV₁) and cookstove exposures (Clark et al. 2009; Diaz et al. 2007; Rinne et al. 2006). Findings for FEV₁/FVC ratios across these studies are mixed. Studies that have considered acute changes following short duration biomass cooking exposures (comparing pre-cooking spirometry measures to during-cooking measures) in Nigeria and Bangladesh have not found any significant changes in FEV₁ or FVC (Medgyesi et al. 2017; Oluwole et al. 2013). Longer-term intervention studies in China, Mexico, and Guatemala have been more consistent in demonstrating improved lung function, or lessened decline in lung function, following use of improved (lower PM_{2.5} emitting) stoves (Romieu et al. 2009; Smith-Sivertsen et al. 2009; Zhou et al. 2014). In the RESPIRE trial in Guatemala, no associations were found between assignment of the plancha stove intervention and FEV₁, FVC, or FEV₁/FVC at follow-up through 18 months (Smith-Sivertsen et al. 2009). However, when exposure was defined continuously as exhaled CO in breath measured at the same time as spirometry was conducted during follow-up, investigators observed a 35 mL decrease in FEV₁ (95% CI -61, -9 mL) and a 26 mL decrease in FVC (95% CI -57, 6 mL) – yet no change in FEV₁/FVC – for each one unit increase in natural log transformed CO (Pope et al. 2015).

Pathways/Mechanisms

The deposition of air pollutants throughout the respiratory pathway can cause irritation and tissue damage, provoking a cascade of cellular injury and inflammation that leads to development and exacerbation of obstructive lung diseases like COPD and asthma, as well as gene mutations and tumorogenesis responsible for development of lung/respiratory cancers (Gordon et al. 2014; Perez-Padilla et al. 2010; U.S. EPA 2009). PM may also contribute to reactive oxygen species (ROS) in the respiratory tract, either through the direct oxidative potential of PM components (e.g., metals, organic species) or indirect stimulation of epithelial and immune cells to produce ROS (U.S. EPA 2009). Oxidative stress and inflammation in the respiratory system can increase susceptibility to bacterial or viral infection and lead to altered infection responses following PM exposures (Gordon et al. 2014; Perez-Padilla et al. 2010). Further, fine particles can deposit deep in the airways and the ultrafine fraction may translocate beyond the respiratory system, which can lead to systemic inflammation and non-respiratory effects such as cardiovascular impacts (Brook et al. 2010; Gordon et al. 2014; Perez-Padilla et al. 2010; U.S. EPA 2009). The larger decrease at three hours post-exposure than immediately post-exposure for FEV1 and FEF25-75 also supports an inflammatory pathway, as the cascade of cellular signaling leading to an inflammatory-type response are not immediate. Pulmonary inflammation can trigger activation of the autonomic nervous system and contribute to development of systemic inflammation, which leads to cardiovascular effects (U.S. EPA 2009). However, the immediate decrease we observed primarily among the three higher-PM_{2.5} level stoves may be indicative of an acute pulmonary irritant mode of action for PM_{2.5}, which could lead to later inflammation.

Reduced FEV₁ and FVC without changes in the FEV₁/FVC ratio is considered a restrictive (non-obstructive) pattern (Barreiro and Perillo 2004; Dempsey and Scanlon 2018; Godfrey and Jankowich 2016; Ranu et al. 2011). Such restrictive spirometry patterns, which have been observed in cigarette smokers and populations with high prevalence of biomass fuel use, have been associated with increased inflammatory markers, hypertension, and cardiovascular disease (Godfrey and Jankowich 2016). Obstructive ventilation patterns, identified by lowered FEV₁/FVC ratios and reduced FEV₁, are hallmark in disease states like asthma and COPD (Averame et al. 2009; Barreiro and Perillo 2004; Mohamed Hoesein et al. 2011; Ranu et al. 2011). Reduced mid-expiratory flow rate (FEF₂₅₋₇₅) is also indicative of obstruction, though this metric is highly

dependent on FVC (Barreiro and Perillo 2004). The decreased FVC and FEV₁ yet minimal changes in the FEV₁/FVC ratio suggest that air pollution exposures generated by the cookstove treatments act through a restrictive, rather than obstructive, pathway.

Pollutant Mixture

Results indicate acute effects of cookstove exposures on lung function compared to filtered air, without an apparent PM_{2.5}-based exposure-response relationship. These findings suggest that a component of the cookstove exposures other than PM_{2.5} mass may be responsible for eliciting the observed health response, either alone or in combination. Previous research has shown that CO may be associated with both lung and systemic inflammation (Abolhassani et al. 2009). The variation in mean CO across the different stove treatments was less than the variation in PM_{2.5} and there was more overlap in CO concentrations between the different treatments and with the control. As such, a CO-dependent effect would likely not be distinguishable within our study design. Other pollutants, such as NO_x or ultrafine levels, may also play a role in pulmonary inflammation (Traboulsi et al. 2017). However, we only measured PM_{2.5} mass and CO levels during each study session, which limits interpretation of the results.

Other Strengths and Limitations

The small, non-significant changes in lung function observed in this study may not be clinically meaningful on an individual level. While the observed effect sizes were small, given the large number of individuals exposed globally, implications for public health could be considerable. Additionally, it is possible that we did not capture the strongest health responses. The post-exposure times for health measurements were chosen because of a combination of logistical considerations within our study protocols and also because they represent potentially key response times within the mechanistic pathway for other health endpoints measured within our study that are not reported here (Langrish et al. 2012). It is possible that these times do not correspond to peak responses within the mechanistic pathway for lung function changes. Further,

the method of measuring lung function using spirometry lacks precision, resulting in measurement error that reduces our statistical power and potentially biases our results towards the null.

The two-hour exposure duration was chosen both because of logistical and ethical considerations for participants and also because two hours approximately represents the duration of a single cooking event. It is unclear how longer duration exposures (e.g., three to four hours) or multiple exposures in a 24-hour period (e.g., representing several cooking events) would impact lung function. Additionally, exposures in real-world settings are more variable than the controlled exposures in this study; this must be considered when comparing our results to field studies.

The results of this study have strong internal validity, as the balanced Williams square crossover design and protocols that restricted participant behavior contributed to a lack of confounders and allowed for more efficient analyses. However, this study may have limited generalizability to the population of cookstove users that is of most interest because the study population was predominately white individuals from a single community. Responses to air pollution exposures may be different among cookstove users of different racial groups (Jones et al. 2015; Sack et al. 2017). Further, we did not obtain any genetic information from our population that could provide insight into questions of genetic susceptibility.

Our research was designed to explore adverse health responses to air pollution in healthy individuals whose normal exposure is low (Good et al. 2016). From a public health perspective, the more interesting research question is whether reducing or eliminating exposures among individuals who have been exposed to higher levels throughout their lifetime – such as through changes in cooking practices in communities using traditional cookstoves – can result in reduced or eliminated health burden. We assume that the health impacts of air pollution are reversible (e.g., removing exposure will result in an inverse response to that seen from being exposed) and that responses in those chronically exposed follow a similar mechanistic pathway as responses in those without chronic exposure; however, this may not be the case.

We included more treatments and a larger sample size than previous studies of wood smoke, ambient air pollution, and diesel exhaust (e.g., Barregard et al. 2008; Bonlokke et al. 2014; Brook et al. 2002; Brook et al. 2009; Byrd et al. 2016; Forchhammer et al. 2012; Langrish et al. 2014; Mills et al. 2005; Muala et al. 2015; Riddervold et al. 2011; Riddervold et al. 2012; Sallsten et al. 2006; Stockfelt et al. 2012; Unosson et al. 2013; Urch et al. 2005), which provided us with the ability to compare effects across a wider exposure range within a single study. We included lower PM_{2.5} levels than previous studies, comparable to ambient air pollution levels in cities throughout the U.S. and Europe. Natural gas or propane is burned for residential heating and cooking in many high-income countries such as the U.S.; the observed short-term effect of the LPG treatment on lung function within this study may be relevant in these settings. Additionally, four of the five cookstove treatments were generated by wood-burning stoves, which may parallel exposures seen from residential heating stoves used in the U.S. and Europe and wildfires. Results may therefore be useful for a wider application than just cookstove-generated air pollution exposures.

Conclusions

We demonstrated that acute exposures to cookstove-generated air pollution elicits responses in lung function including decreases in FVC and FEV₁. While statistically non-significant, effects were observed immediately (approximately 30 minutes) post-exposure and with increased strength at three hours post-exposure, without an apparent exposure-response consistent with increasing PM_{2.5} levels across the various treatments. These results suggest that household air pollution may be detrimental to pulmonary function even at levels as low as 10 μ g/m³. More work is needed to understand how the acute responses demonstrated by this work translate to effects in chronically-exposed populations.

CHAPTER 7: CONCLUSIONS

This body of work contributed knowledge to two broad areas in cookstove research: 1) identification of startup materials and quantification of their emissions and 2) understanding health responses following controlled exposure to cookstove emissions from different stove technologies.

Cookstove Startup Materials and Emissions

Anecdotal knowledge indicated that a variety of non-standard fuels are used to ignite cookstoves, yet little was known about the types of materials used. Limited laboratory data suggested that the startup process could contribute substantially to overall cooking emissions. We helped fill these data gaps by surveying experts about their observations on startup practices and then characterizing emissions from these materials.

The use of researchers and practitioners as survey respondents allowed us to cover a range of locations and gather information relative to diverse practices more easily than would be the case if we had targeted responses from cookstove users. Responses indicated that a variety of materials are used to ignite cookstoves, some of which are very different from the typical forms of biomass used as main fuels, which gave further justification for the importance of characterizing emissions from these materials. The large number of startup materials identified could be grouped into a few categories based on their composition. For example, nearly half the mentioned materials across all responses fell into three categories (accelerants, paper, and plastic). The fact that startup materials can be grouped into a few categories suggests that a reduced set of emissions factors based on a subset of representative materials could be used to develop a useful startup emissions inventory. We may have overrepresented practices used with more efficient, advanced stove designs, and underrepresented practices in East/Southeast Asia and South America. Further work to substantiate our results is necessary before drawing any wider conclusions about global startup practices. Additionally, more detailed information on the startup

practices beyond the fuel types used – such as how seasonality, primary stove type, or other factors affect material choice – could help inform and refine estimates about the relative importance of startup materials. Surveys about startup practices could be incorporated into ongoing field research to help gather this information.

Informed by the survey results, we conducted a laboratory-based emissions measurement campaign. The testing protocol developed mimicked the startup process while isolating emissions from the startup fuels only, which had not been done in previous work. Emissions factors were calculated using the novel unit of emissions per startup event in addition to the standard emissions per mass of fuel burned. Emissions factors for these startup materials can be used to determine startup materials' contribution to overall cookstove emissions. These emissions factors can also be scaled to provide emissions estimates in other settings such as backyard open burning.

It is possible that the use of startup materials in different stove designs alters the combustion dynamics and, therefore, emissions. Further work should be done to test this hypothesis, as the health and climate effects estimated from startup emissions may vary depending on the predominant stove types used. Additionally, quantification of the amount of startup fuel used under different scenarios is critical. We used a conservative amount of material, which means "per-startup" emissions may be underestimates. However, this further illustrates the importance of better understanding of startup fuel practices and emissions, as even with a conservatively low estimate of emissions, our results indicated that the startup emissions can play a substantial role in overall cooking emissions. Further work should be done to characterize more material types and pollutants and consider whether the use of startup materials in different stove types or operation methods result in different combustion dynamics that alter emissions.

Health Effects across a Spectrum of Exposures

While household air pollution is estimated to have a substantial burden on global respiratory and cardiovascular health, empirical evidence describing this relationship across a range of technology types and exposure levels is limited. Our controlled human exposure study

evaluated markers of cardiovascular and respiratory health in the 24 hours following short-term exposures to cookstove air pollution that spanned $PM_{2.5}$ exposure levels from 10 µg/m³ (for liquid petroleum gas [LPG]) up to 500 µg/m³ (for traditional three stone fire). The controlled exposure design allowed for comparisons to be made across a much wider range of exposure levels and stove types than is feasible in most field settings. The ability to make these wide-ranging comparisons in a single study adds even more value, as comparing across different observational studies is complicated by the variety of differences that exist between studies and may influence the results of each study – such as different study populations, time periods, study designs, and measurement methods. Another strength of our study is that the crossover design allowed for efficient comparisons within person, eliminating concerns about confounding that are inherent to most other observational epidemiologic designs.

Results suggest that both blood pressure and lung function are impacted by cookstove emission exposures on acute timescales. Systolic pressure was 2 to 3 mmHg lower at 24 hours post-exposure for stove treatments compared to the control; lung capacities (FVC, FEV₁) were approximately 1 to 2% lower and mix-expiratory flow (FEF₂₅₋₇₅) was approximately 2 to 3% lower at the immediate and three-hour post-exposure measurements. Surprisingly, effects occurred at a similar magnitude across all treatments – the supposedly clean technologies of LPG and gasifier stoves produced effects at similar levels as the traditional three stone fire.

A major limitation of our study is that we assessed acute health responses resulting from short, transient exposures, yet real-world exposures to household air pollution are chronic, occurring multiple times per day for years or an entire lifetime. While understanding acute health effects from single, short exposures can provide evidence that allows for inference about the clinical health responses following chronic exposures, it is not reasonable to draw strong conclusions from our work about the larger global health burden of cookstoves. The results from this study provide insight into the mechanisms and early responses that occur following exposures to household air pollution and allow us to compare these responses across different stove types

and exposure levels. We can reasonably hypothesize that chronic exposures will result in continuous activation of the observed pathways, eventually contributing to sustained physiological responses that lead to larger cardiovascular and pulmonary disease, and that differences between stove types or exposure levels will be amplified along this pathway. However, we do not actually know how longer exposures, sustained exposures, or continuous re-exposures over a longer time frame will modulate health responses. Evaluating evidence from this controlled exposure study in light of results from field studies that consider similar endpoints can help bridge the gap and tie our findings into the larger picture of cookstove health burden.

Results for both blood pressure and spirometry support an inflammatory pathway mechanism through which PM_{2.5} impacts health in the short-term. Additional work is underway to evaluate inflammatory markers in blood samples that may help further elucidate these mechanistic pathways. Further, we measured several other health endpoints not reported here, including pulse wave velocity, augmentation index, heart rate variability, and blood lipid levels. Analyses of these endpoints, for consideration with blood pressure and lung function, will help contribute to the overall picture of how air pollution impacts health.

We found no evidence of a PM_{2.5} exposure-response relationship for both the blood pressure and respiratory endpoints. There are a number of potential explanations for this related to our design. For example, it is possible that an exposure-response curve does not exist on the acute time scale studied for these health endpoints; observation of a response at multiple PM_{2.5} concentration levels could be the result of an acute response triggered by any cookstove air pollution exposure. Alternatively, measurement error and imprecision in the health outcomes could have obscured observation of an existing exposure-response. For example, blood pressure measurements were taken at a resolution of 1 mmHg, yet the observed differences in blood pressure were 1 to 3 mmHg. Also, we defined our treatments as categorical stove type, yet there was variation in the individual exposure concentration averages within each category, which induces error because exposures across the same treatment are not identical. Evaluation of other
health metrics, such as blood inflammatory markers, may reveal exposure-response relationships on the timescales studied. It is also possible that a component of the exposures other than PM_{2.5} mass may be responsible for eliciting the observed health responses – either alone, or in combination with other component pollutants.

The method in which the treatments were administered in this study may have resulted in exposures that are not consistent with real-world exposures. In real-world settings, household exposures are a mix of stove emissions and air pollution from other sources. In our study, emissions from the stove treatments were mixed with laboratory and building air; for the lowest exposure treatment (LPG), target PM_{2.5} exposure levels approached background conditions. The differences in real-world versus the controlled exposures could result in a difference in the health responses seen in the laboratory versus in the field despite the same stove being used, particularly for low exposure stoves. Additional work is underway to further characterize the pollutant mixture within the chamber under each stove treatment, including measurement of ultrafine particles, NO_x, elemental and organic carbon, and VOCs, which will help interpret results.

Overall Contribution and Impact

The startup materials survey and emissions campaign presented in this dissertation are the first attempt to systematically identify startup material types and quantify emissions. Results demonstrated that many material types are used during startup and the emissions from these materials may contribute substantially to total cookstove emissions. Fuller understanding of startup practices is needed to guide quantitative emissions inventories and facilitate effective exposure mitigation strategies.

The controlled exposure study demonstrated that exposure to cookstove air pollution from different stove types can elicit acute cardiovascular and respiratory effects. Effects occurred across $PM_{2.5}$ exposure levels from 10 to 500 µg/m³, with limited evidence of a PM-based exposure-response. These results raise questions on the potential efficacy of clean cookstove interventions using the technologies tested in this study.

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APPENDIX A: SUPPLEMENTAL MATERIAL FOR CHAPTER 3 (STARTUP SURVEY)

Startup Materials Survey

The following pages contain screenshots of the online-hosted survey administered to collect data for this research. The survey was created and hosted through a Google Forms (Google, 2016). Individuals recruited to take the survey were contacted between December 15, 2015 and January 15, 2016. The survey remained open for responses until February 15, 2016.



This survey is part of a effort by researchers at Colorado State University in Fort Collins, Colorado, USA to understand emissions from cookstoves and related health impacts. The purpose of this survey is to gather information about fuels used during start-up of cookstoves/cooking fires around the world. You have been asked to participate in this survey because you have experience working with/living in communities that use cookstoves.

The information you provide will be used to inform research related to the human health and climate relevant emissions from cookstoves and cooking practices. Results from this survey may be published, however we will keep any identifying information private. Please note that responses are anonymous, as there is no contact information linked to your responses.

This survey should take between 10 and 30 minutes to complete.

For this survey, we are interested in information about materials/fuels that are used in cookstove start-up, including those used very commonly and those used only occasionally or rarely.

- 'Start up' refers to the period in which the cook is actively trying to turn on or start their stove from a dormant state, raising the heat or firepower in order to initiate a cooking session.
- "Fuels" or "materials" refer to any object that is used to power this process and is consumed by the stove to contribute to the generation of the energy output.

Start up materials may also be thought of as kindling

Where do you currently work?

Please name your primary or main affiliation.

What is your current job title?

How long have you been involved in cookstove-related work? (enter the number of years).

Please briefly describe how you are involved in the cookstove field.

e.g., "I have been involved in air pollution research for 15 years. Over the past 5 years, I've been involved with several projects monitoring health impacts of cookstove interventions in Central America." or "I perform research and development on energy-efficient cookstove design."

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Cookstove Startup Fuels Survey

In what locations do you feel like you have first-hand knowledge of current, commonplace cooking practices?

Please list locations where you feel like you have expert-level insight through: living in the area, personally visiting for cookstove-related work, communicating with locally-based groups about cookstoves, working on a team where colleagues visited to conduct cookstove-related research, etc.

"Location" is loosely defined so that you can be as specific or broad as you think is justified. In subsequent questions, you will be asked to provide information about cooking practices in these locations as you've defined them, and will have space to discuss 3 or more types of startup materials used in the location. Define the scale of your location (country, state, specific region, urban or rural area, etc.) based on having observed similar cooking practices across the area. While more specific details on the location will be helpful in comparing your input to that from others (i.e., rural areas in Honduras), broad locations will help us understand if a certain practice is very widespread (i.e., Central America).

Please enter only one region/location in each entry space. You may enter up to 3 locations. If you have worked in more than 3 distinct locations, please list the 3 that you feel you have the most experience, and consider whether certain locations can be grouped together based on similarities in cooking practices.

Please note that in the subsequent questions, we will refer to the locations you've listed according to the location number. Take a moment to jot down a note of which locations you have listed under each location number, to reference as you continue through this survey.

Location #2	
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Location #3	
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Cookstove Startup Fuels Survey

Fuel Use in Location #1

In the questions on this page and the following 2 pages, you will be asked to provide information on the two most common stove types used in Location #1. You will be asked to provide information on the two most common start-up fuels used for each stove type. You will also be able to list all start-up materials you are aware are used, beyond the two most common.

The answer to all questions should be based on your opinion and expertise, there is no right or wrong answer.

Based on your experience, what is the main (primary or most common) stove type used in Location #1? Please give the common name as well as a description, for example: "three stone fires" or "small ceramic jiko-style stoves."

What is the primary fuel source burned in the stove type you listed above (the primary or most common stove type) in this location?

Based on your experience, what is the most common material/fuel source used to help start a fire/cookstove in this style stove in this location? Please provide a description of the material and how it is used.

How common is the use of the startup material you named above (the most commonly used material), in your experience?

- 0 This material is used by more than 75% of people in this region to start fires/stoves.
- O This material is used by 50-75% of people in this region to start fires/stoves.
- $\odot\,$ This material is used by 25-50% of people in this region to start fires/stoves.
- O This material is used by less than 25% of people in this region to start fires/stoves.

Based on your experience, what is the second most common material/fuel source used to help start a fire/cookstove in this style stove in this location? Please provide a description of the material and how it is used.

How common is the use of the startup material you named above (the second most commonly used material), in your experience?

- O This material is used by more than 75% of people in this region to start fires/stoves.
- This material is used by 50-75% of people in this region to start fires/stoves.
- O This material is used by 25-50% of people in this region to start fires/stoves.
- ③ This material is used by less than 25% of people in this region to start fires/stoves.

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NE	
	Cookstove Startup Fuels Survey
	Fuel Use in Location #1, continued
	Please list as many additional materials as you are aware of that are used in location #1, including unique materials that may be used infrequently or uncommonly in the region but nevertheless occur. Please indicate in your response how commonly you think the material is used and what types of stoves/primary fuels it's use is associated with.
	Based on your experience, does the use of different startup materials in this location vary depending on things such as season, availability of fuel types, or other factors?
	© Yes © No
Xoxi	If you said "yes", the choice of what startup material is used depends on other factors, please explain.
	Have you ever collected data on the startup processes and fuels used in location #1?
	Have you ever collected data on the startup processes and fuels used in location #1? Ves, I have collected data. I looked at that data to inform my answers to these questions. Ves, I have collected data. I did not look at that data to inform my answers to these questions.
	Have you ever collected data on the startup processes and fuels used in location #1? Yes, I have collected data, I looked at that data to inform my answers to these questions. Yes, I have collected data, I did not look at that data to inform my answers to these questions. No, I have not ever collected data on this. I based my answers to these questions on what I've seen.
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	Have you ever collected data on the startup processes and fuels used in location #1? Yes, I have collected data, I looked at that data to inform my answers to these questions. Yes, I have collected data, I did not look at that data to inform my answers to these questions. No, I have not ever collected data on this. I based my answers to these questions on what I've seen. Back Continue 38% completed

Cookstove Startup Fuels Survey

Fuel Use in Location #2

In the questions on this page and the following 2 pages, you will be asked to provide information on the two most common stove types used in Location #2. You will be asked to provide information on the two most common start-up fuels used for each stove type. You will also be able to list all start-up materials you are aware are used, beyond the two most common.

The answer to all questions should be based on your opinion and expertise; there is no right or wrong answer.

Based on your experience, what is the main (primary or most common) stove type used in Location #2? Please give the common name as well as a description, for example: "three stone fires" or "small ceramic jiko-style stoves."

What is the primary fuel source burned in the stove type you listed above (the primary or most common stove type) in this location?

Based on your experience, what is the most common material/fuel source used to help start a fire/cookstove in this style stove in this location? Please provide a description of the material and how it is used.

How common is the use of the startup material you named above (the most commonly used material), in your experience?

- O This material is used by more than 75% of people in this region to start fires/stoves.
- This material is used by 50-75% of people in this region to start fires/stoves.
- This material is used by 25-50% of people in this region to start fires/stoves.
- ③ This material is used by less than 25% of people in this region to start fires/stoves.

Based on your experience, what is the second most common material/fuel source used to help start a fire/cookstove in this style stove in this location? Please provide a description of the material and how it is used.

How common is the use of the startup material you named above (the second most commonly used material in the most common stove type), in your experience?

- O This material is used by more than 75% of people in this region to start fires/stoves.
- This material is used by 50-75% of people in this region to start fires/stoves.
- This material is used by 25-50% of people in this region to start fires/stoves.
- O This material is used by less than 25% of people in this region to start fires/stoves.

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	Cookstove Startup Fuels Survey
50	Fuel Use in Location #2, continued Based on your experience, what is the second most common stove type used in Location #2? Please give the common name as well as a description, for example: "three-stone fires" or "small ceramic piko-style stoves."
	What is the primary fuel source burned in the stove type you listed above in this location? Based on your experience, what is the most common material/fuel source used to help start a
	fire/cookstove in this style stove (the second most common stove type) in this location? Please provide clear descriptions of the material and how it is used. How common is the use of the startup material you named above (the most commonly used material in the second most common stove type), in your experience?
WSWI	 This material is used by more than 75% of people in this region to start fires/stoves. This material is used by 50-75% of people in this region to start fires/stoves. This material is used by 25-50% of people in this region to start fires/stoves. This material is used by less than 25% of people in this region to start fires/stoves.
	Based on your experience, what is the second most common material/fuel source used to help start a fire/cookstove in this style stove (the second most common stove) in this location? Please provide a clear description of the material and how it is used. How common is the use of the startup material you named above (the second most commonly used
	 This material is used by more than 75% of people in this region to start fires/stoves. This material is used by 50-75% of people in this region to start fires/stoves. This material is used by 25-50% of people in this region to start fires/stoves. This material is used by 25-50% of people in this region to start fires/stoves. This material is used by less than 25% of people in this region to start fires/stoves.
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Cockstove Startup Fuels Survey Fuel Use in Location #2, continued Passe indicate in your response work or our only you think the material is used and what type or every primary fuels it is use is associated with. Passe on your experience, does the use of different startup materials in this location vary dep triggs such as season, evailability of fuel types, or other factors? I yes No If you said 'yes', the choice of what startup material is used depends on other factors, please ending the indicate in place of the startup material is used depends on other factors, please ending the indicate of the startup material is used depends on other factors, please ending the indicate of the startup processes and fuels used in location #2? It yeu endicated data on the startup processes and fuels used in location #2? A set on the startup processes and fuels used in location #2? I we collected data on the startup processes and fuels used in location #2? I we collected data on the startup processes and fuels used in location #2? I we collected data on the startup processes and fuels used in location #2? I we collected data in did not look at that data to inform my answers to these questions on what I we collected data on the is a the data to inform my answers to these questions on what I we collected data on the is a the data to inform my answers to these questions on what I we collected data on the is a the data to inform my answers to these questions on what I we collected data on the is a startup material is used on form my answers to these questions on what I we collected data on the is a based my answers to these questions on what I we collected data on the is a startup material is used inform my answers to these questions on what I we collected data on the is a the data to inform my answers to these questions on what I we collected data on the is a the data on the is a startup material is used inform my answers to these questions on w	
Full Use in Location #2, continued Please list as many additional materials as you are aware of that are used in location 32, inclusterials that may be used infrequently or uncommonly in the region but nevertheless occur. Please indicate in your response how commonly you think the material is used and what types of the transmission of the transmission of the transmission. Please on your experience, does the use of different startup materials in this location vary deptions such as season, availability of fuel types, or other factors? Pres No In our experience of the transmission o	1
Based on your experience, does the use of different startup materials in this location vary deptings such as season, availability of fuel types, or other factors? Yes No If you said "yes", the choice of what startup material is used depends on other factors, please estimates and the season of the startup processes and fuels used in location #2? Yes, I have collected data. I looked at that data to inform my answers to these questions. Yes, I have collected data. I did not look at that data to inform my answers to these questions. No, I have not ever collected data on the. I based my answers to these questions on what I we have to the set of the	ing unique
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Cookstove Startup Fuels Survey

Fuel Use in Location #3

In the questions on this page and the following 2 pages, you will be asked to provide information on the two most common stove types used in Location #3. You will be asked to provide information on the two most common start-up fuels used for each stove type. You will also be able to list all start-up materials you are aware are used, beyond the two most common,

The answer to all questions should be based on your opinion and expertise; there is no right or wrong answer.

Based on your experience, what is the main (primary or most common) stove type used in Location #3? Please give the common name as well as a description, for example: "three stone fires" or "small ceramic jiko-style stoves."

What is the primary fuel source burned in the stove type you listed above (the primary or most common stove type) in this location?

Based on your experience, what is the most common material/fuel source used to help start a fire/cookstove in this style stove in this location? Please provide a description of the material and how it is used.

How common is the use of the startup material you named above (the most commonly used material), in your experience?

- This material is used by more than 75% of people in this region to start fires/stoves.
- This material is used by 50-75% of people in this region to start fires/stoves.
- This material is used by 25-50% of people in this region to start fires/stoves.
- This material is used by less than 25% of people in this region to start fires/stoves.

Based on your experience, what is the second most common material/fuel source used to help start a fire/cookstove in this style stove in this location? Please provide a description of the material and how it is used.

How common is the use of the startup material you named above (the second most commonly used

- material in the most common stove type), in your experience?
 This material is used by more than 75% of people in this region to start fires/stoves.
- This material is used by 50-75% of people in this region to start fires/stoves.
- This material is used by 25-50% of people in this region to start fires/stoves.
- p This material is used by 20-50% of people in this region to start mes/stoves.



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	Cookstove Startup Fuels Survey	
	Fuel Use in Location #3, continued	
	Based on your experience, what is the second most common stove type used in Location #3? Please give the common name as well as a description, for example: "three-stone fires" or "small ceramic jiko-style stoves."	
A.	What is the primary fuel source burned in the stove type you listed above in this location?	
Z	Based on your experience, what is the most common material/fuel source used to help start a fire/cookstove in this style stove (the second most common stove type) in this location? Please provide clear descriptions of the material and how it is used.	
1	How common is the use of the startup material you named above (the most commonly used material in the second most common stove type), in your experience?	1
	This material is used by more than 75% of people in this region to start fires/stoves.	
Note Comment	This material is used by 50-75% of people in this region to start fires/stoves.	Soles-
	This material is used by 25-50% of people in this region to start fires/stoves.	1000
WWW I	This material is used by less than 25% of people in this region to start fires/stoves.	
	Based on your experience, what is the second most common material/fuel source used to help start a fire/cookstove in this style stove (the second most common stove) in this location? Please provide a clear description of the material and how it is used.	
	How common is the use of the startup material you named above (the second most commonly used material in the second most common stove type), in your experience?	-
同学成	This material is used by more than 75% of people in this region to start fires/stoves.	Service 1
	This material is used by 50-75% of people in this region to start fires/stoves.	Carlos -
	This material is used by 25-50% of people in this region to start fires/stoves.	100 C
	O This material is used by less than 25% of people in this region to start fires/stoves.	1. 18
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	Cookstove Startup Fuels Survey	- And
	Fuel Use in Location #3, continued	
	Please list as many additional materials as you are aware of that are used in location #3, including unique materials that may be used infrequently or uncommonly in the region but nevertheless occur. Please indicate in your response how commonly you think the material is used and what types of stoves/primary fuels it's use is associated with.	
	Based on your experience, does the use of different startup materials in this location vary depending on things such as season, availability of fuel types, or other factors?	K
	No If you said "yes", the choice of what startup material is used depends on other factors, please explain.	AC,
-30) ²		-
	Have you ever collected data on the startup processes and fuels used in location #3?	
	 Yes, I have collected data. I looked at that data to inform my answers to these questions. Yes, I have collected data. I did not look at that data to inform my answers to these questions. No, I have not ever collected data on this. I based my answers to these questions on what I've seen. 	
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	Cookstove Startup F	Fuels Survey	L
	Thank you for your response Please contact me if you have any quest and the use of your responses. <u>Kristen f</u>	SE! tions or concerns regarding this survey redakacolostate edu	
	If you have any additional comments related to coo	ikstove startup fuels, please enter them here.	
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APPENDIX B: SUPPLEMENTAL MATERIAL FOR CHAPTER 4 (STARTUP EMISSIONS)

Detailed Methods

Emissions tests were performed at the Colorado State University Advanced Cookstove Testing Laboratory (Fort Collins, CO). Tests were performed within a total-capture encapsulating hood; a constant volume of filtered air (4 m³/min) was drawn through the hood to achieve natural dilution and allow emissions to reach typical indoor concentrations. Emissions were then drawn through isokinetic sampling probes to size-selective filter samplers, whole air sample collection canisters, or real-time instruments (as appropriate for each specific pollutant). Flow rates were measured at the sampling probe sites before and after each test to track partial capture proportions and system dilution ratios and ensure isokinetic sampling.

Integrated filter-based collection methods were used to measure particulate matter mass with diameters less than 2.5µm (PM_{2.5}), elemental carbon (EC), and organic carbon (OC). Integrated cartridge-based collection was used for gas-phase carbonyls (e.g., formaldehyde, acetaldehyde). Integrated canisters were used to collect an air sample for the volatile organic compounds benzene, toluene, ethylbenzene, and xylenes (BTEX). Carbon monoxide (CO), carbon dioxide (CO₂), and methane (CH₄) were measured at 1 Hz temporal resolution (Siemens Ultramat 6E, Siemens AG, Germany).

PM_{2.5} was collected on polytetrafluoroethylene (PTFE) membrane filters (Tisch Environmental, USA) placed downstream of 2.5 μm aerodynamic size cut-point cyclones. Filter mass was measured gravimetrically using a microbalance (Mettler Toledo MX5, USA) after filters had equilibrated to consistent temperature/humidity (filters were pre-weighed after a minimum of 24 hours of equilibration and used within two weeks of pre-weight date; filters were placed into the weight room for equilibration within six hours of collection and post-weighted within 24 to 36 hours of collection).

Elemental and organic carbon were collected on pre-baked quarts filters (Tissuequartz, Pall Life Sciences, USA) downstream of 2.5-µm cut-point cyclone and analyzed using a Sunset Laboratory ECOC Analyzer following the NIOSH 5040 method. To correct for the semi-volatile organic carbon artifact, a quartz filter sampled behind a PTFE filter as well as a stand-alone quartz filter were collected. The carbon measured on the quartz behind PTFE filter was subtracted from the stand alone quartz filter to provide the final OC concentrations.

Equipment used to house all filters (e.g., stainless steel cartridges, plastic filter cassettes) were cleaned prior to deployment using soap and water followed by rinsing with a mixture of dichloromethane, hexane, and methanol, then air-dried under a chemical fume hood. Cleaned equipment was stored in air-tight plastic bags prior to use (no more than one week; typically 24 hours). Quartz filters were baked at 800 degrees Celsius for 13 hours then stored in sealed glass jars (baked at 350 degrees Celsius for 8 hours to clean) prior to use (within two weeks of bake date).

The most common method for collecting and analyzing samples for carbonyls is the EPA Method TO-11A. Given that the sampling cartridges and column mentioned in this method are no longer available, we developed our own approach that follows the same principles as the EPA method and is outlined in detail below. Carbonyls were collected on 2,4-dinitrophenylhydrazine (DNPH) silica-based cartridges (Waters Sep-Pak) downstream of an ozone scrubber (Waters) at a flowrate of approximately 1 L/min across each burn. The DNPH cartridges and ozone scrubbers were stored in their manufacturer-sealed packages at -20 degrees Celsius prior to sampling. Once sampled, the DNPH cartridges were recapped, placed in 50 mL plastic centrifuge tubes, and stored at -80 °C until analyzed. Each DNPH cartridge was extracted using 3 mL acetonitrile pulled through the cartridge using a vacuum manifold. The extract was collected in a 5 mL volumetric flask. Once extraction was complete, the volumetric flask was filled to the line with acetonitrile to provide a final extraction volume of 5 mL. Each sample was analyzed immediately following extraction on an Agilent 1260 Infinity high-performance liquid chromatography (HPLC) equipped

with a dual channel pump and UV detector set to monitor at a wavelength of 360 nm. The separation was conducted on a Waters Nova-Pak C-18 4 μ m column (3.9 x 150 mm). The eluents were 60% deionized water:30% acetonitrile:10% tetrahydrofuran (A) and 40% water:60% acetonitrile (B). The complete run time was 32 minutes and included two steps. For the first 25 minutes, a linear gradient from 100% A to 100% B was conducted. For the final five minutes, a re-equilibration step was run to return to the starting conditions of 100% A. The flowrate during each step was 1.5 mL/min. A sample injection volume of 10 μ L was used. The limit of detection (LOD) for the various carbonyls was approximately 0.1 μ g/m³.

Whole air samples for BTEX analyses were collected using two-liter electropolished stainless steel canisters equipped with Silonite-coated flow controller valves (Entech Instruments Inc.). The flow regulator allowed for air to be consistently sampled from the airflow stream during an emissions test (so that time-weighted average concentrations could be calculated) and was set to a flow rate that resulted in the canister filling up to 60-80% by the end of the test. Canisters were cleaned prior to sampling by evacuating and refilling with ultra-high purity nitrogen that had passed through an activated charcoal molecular sieve, eight times. After the final flush, the canisters were evacuated to less than one torr. Canisters were used within two weeks of cleaning. Sampled canisters were stored at room temperature until analyzed (average turnaround time of less than two weeks). The canisters were analyzed using a five-channel gas chromatography (GC) coupled with two flame ionization detectors (FID), two electron capture detectors (ECD), and a quadrupole mass spectrometer (MS). Benzene and toluene were quantified using the GC-FID channel, whereas the GC-MS system was used to determine ethylbenzene and the xylenes. The LODs for the BTEX compounds were: benzene, 0.01 ppb; toluene: 0.017 ppb; ethylbenzene: 0.019 ppb; m,p-xylene: 0.014 ppb; o-xylene: 0.006 ppb.

CO, CH₄, and CO₂ were measured using a nondispersive infrared detection (NDIR) analyzer (Siemens Ultramat 6E, Siemens AG, Germany) that took a reading every second. The

instrument was zeroed using ultra-high purity nitrogen and spanned using pure gas before each testing day.

Measured pollutant levels (concentration, mixing ratio, mass) were converted to total pollutant mass emitted per test based on the hood flow rate (4 m³/min), sampling flow rates (shown in Figure B.1), and test duration, as appropriate. In addition to emissions factors reported on a per-startup basis (as described in the main text), emissions factors of mass of pollutant emitted per mass of material burnt were also calculated. Emissions factors were calculated per test, using the total mass of material consumed per test as the mass of the startup bundle prior to burning minus the mass of any remaining unburnt material or ash, added across all bundles burned in a single test. We then averaged the emissions factors across all tests of the same material type.

Supplementary Figures/Results

Fuel Type	Number of tests	Number of startups per test*		
kerosene	4	5, 5, 5, 6		
kindling	3	3, 3, 3		
footwear	3	2, 2, 3		
inner tubes	3	4, 4, 4		
newspaper	5	6, 6, 7, 7, 7		
wood shims	3	2, 2, 2		
fabric	4	6, 6, 6, 6		
food packaging	4	4, 5, 7, 7		
plastic bags	3	6, 6, 6		

Table B.1 Number of startup replicates per test.

*While we aimed for each startup bundle to be similar in size, small variations in mass meant that the exact number of bundles that would be burned within the 20 to 30 minute test time frame could vary by one or two across tests of the same material type.

Table B.2 Mean pollutant emissions per startup event for additional carbonyls. Emissions per startup event, calculated as the total mass of pollutant emitted per test divided by the standard mean mass of fuel consumed per startup bundle and averaged over the test replicates. Values are background adjusted. Materials are listed in order of highest to lowest $PM_{2.5}$ emissions (left to right). n=number of tests, each test contained multiple startup events (2-7, see table S1). <LOD = below limit of detection.

	Average Emissions Per Startup Event, mg (min, max)									
Pollutant	kerosene (n=4 tests; 21 total startups)*	kindling (n=3 tests; 9 total startups)	footwear (n=3 tests; 7 total startups)	inner tubes (n=3; 12 total startups)	newspaper (n=5 tests; 33 startups)*	wood shims (n=3 tests; 6 startups)	fabric (n=4 tests; 24 startups)	food packaging (n=4 tests; 23 startups)	plastic bags (n=3 tests; 18 startups)	
2,5-dimethyl-be nzaldehyde	0.18 (0.14, 0.22)	0.95 (0.86, 1.03)	<lod< td=""><td><lod< td=""><td>1.03 (0.78, 1.39)</td><td>0.17 (0.15, 0.19)</td><td>0.22 (0.02, 0.77)</td><td>0.01 (0.01, 0.02)</td><td><lod< td=""></lod<></td></lod<></td></lod<>	<lod< td=""><td>1.03 (0.78, 1.39)</td><td>0.17 (0.15, 0.19)</td><td>0.22 (0.02, 0.77)</td><td>0.01 (0.01, 0.02)</td><td><lod< td=""></lod<></td></lod<>	1.03 (0.78, 1.39)	0.17 (0.15, 0.19)	0.22 (0.02, 0.77)	0.01 (0.01, 0.02)	<lod< td=""></lod<>	
acetone	1.76 (1.1, 2.54)	5.30 (0.13, 8.35)	2.96 (2.87, 3.09)	2.72 (1.2, 5.34)	0.80 (0.06, 2.09)	30.97 (3.37, 83.69)	1.02 (0.41, 1.93)	1.90 (1.07, 3.04)	27.19 (0.9, 79.09)	
benzaldehyde	1.02 (0.96, 1.05)	1.02 (0.47, 1.41)	0.13 (0.1, 0.2)	0.12 (0.09, 0.14)	0.44 (0.25, 0.73)	0.49 (0.38, 0.6)	0.09 (0.09, 0.09)	0.13 (0.11, 0.16)	0.11 (0.07, 0.15)	
butanone	0.56 (0.26, 1.02)	1.72 (0.37, 2.85)	0.18 (0.11, 0.22)	0.21 (0.09, 0.36)	0.21 (0.04, 0.55)	0.55 (0.44, 0.67)	0.11 (0.07, 0.13)	0.25 (0.12, 0.42)	0.16 (0.07, 0.31)	
butyraldehyde	0.22 (0.15, 0.27)	2.81 (2.48, 3.25)	0.07 (0.06, 0.08)	0.11 (0.09, 0.14)	1.04 (0.49, 1.46)	0.28 (0.2, 0.33)	0.05 (0.03, 0.09)	0.08 (0.06, 0.1)	0.12 (0.1, 0.14)	
hexaldehyde	1.24 (1.12, 1.42)	12.12 (9, 16.24)	0.37 (0.27, 0.49)	0.29 (0.19, 0.36)	4.78 (3.61, 6.27)	7.48 (5.47, 9.08)	1.09 (0.98, 1.21)	0.43 (0.23, 0.56)	0.36 (0.34, 0.39)	
isovaleralde- hyde	0.19 (0.14, 0.21)	2.48 (1.38, 3.7)	0.10 (0.07, 0.13)	0.18 (0.05, 0.37)	1.14 (0.92, 1.49)	1.09 (0.65, 1.48)	0.15 (0.09, 0.18)	0.11 (0.07, 0.15)	0.11 (0.07, 0.14)	
methacrolein	0.39 (0.09, 0.6)	0.44 (0.25, 0.78)	0.12 (0.03, 0.19)	0.22 (0.17, 0.26)	0.18 (0.04, 0.28)	0.41 (0.36, 0.49)	0.17 (0.16, 0.2)	0.12 (0.05, 0.17)	0.24 (0.18, 0.29)	
m, p-tolualdehyde	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""></lod<></td></lod<>	<lod< td=""></lod<>	
o-tolualdehyde	1.15 (1, 1.28)	27.01 (22.74, 29.24)	0.53 (0.27, 0.99)	0.24 (0.02, 0.4)	18.24 (14.6,20.56)	12.43 (8.48,16.23)	2.04 (1.26, 2.45)	0.39 (0.15, 0.61)	0.31 (0.25, 0.34)	
propion- aldehyde	0.33 (0.28, 0.38)	4.15 (2.48, 5.07)	1.24 (0.09, 3.42)	0.09 (0.06, 0.13)	0.65 (0.53, 0.95)	0.9 (0.59, 1.12)	0.16 (0.09, 0.27)	0.33 (0.11, 0.94)	0.16 (0.13, 0.19)	
valeraldehyde	0.12 (0.08, 0.15)	0.19 (0.15, 0.24)	0.11 (0.07, 0.19)	0.06 (0.04, 0.1)	0.05 (0.05, 0.08)	0.17 (0.15, 0.2)	0.03 (0.01, 0.04)	0.06 (0.05, 0.07)	0.18 (0.15, 0.22)	
•			Emi	ssions per mas	s of fuel (g/kg; /	Average [min, m	nax])	•		
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pollutant	kerosene (n=4 tests; 21 total startups)*	kindling (n=3 tests; 9 total startups)	footwear (n=3 tests; 7 total startups)	inner tubes (n=3; 12 total startups)	newspaper (n=5 tests; 33 startups)*	wood shims (n=3 tests; 6 startups)	fabric (n=4 tests; 24 startups)	food packaging (n=4 tests; 23 startups)	plastic bags (n=3 tests; 18 startups)	
				Carbon	Gases					
CO ₂	2906 (2786, 3029)	1257 (1136, 1318)	1353 (1000, 1932)	2601 (2490, 2800)	1588 (1527, 1646)	1577 (1516, 1630)	1608 (1567, 1646)	2143 (1505, 3311)	1512 (1179, 1687)	
CO	31 (30, 32)	50 (47, 52)	7 (5, 8)	28 (25, 33)	45 (35, 53)	24 (18, 28)	22 (19, 24)	19 (12, 34)	16 (14, 21)	
CH ₄	3 (2, 6)	3 (3, 3)	1 (<0.5, 1)	2 (1, 3)	2 (2, 5)	2 (1, 2)	4 (<0.5, 13)	11 (<0.5, 44)	1 (<0.5, 2)	
				Particula	te Matter		x : <i>x</i>	· · · · · ·		
PM2.5 mass	32 (24, 36)	18 (17, 20)	33 (11, 73)+	35 (31, 40)	7 (5, 8)	3 (2, 3)	5 (4, 6)	3 (<0.5, 5)	1 (<bg, 4)<="" td=""></bg,>	
elemental carbon	12 (5, 19)	<0.5 (<0.5, 1)	11 (8, 13)	16 (14, 19)	<0.5 (<0.5, 1)	<0.5 (<0.5, 1)	1 (<0.5, 1)	2 (<bg, 4)<="" td=""><td><0.5 (<bg, <0.5)</bg, </td></bg,>	<0.5 (<bg, <0.5)</bg, 	
organic carbon	15 (9, 22)	11 (9, 13)	1 (<0.5, 1)	14 (8, 20)	4 (3, 5)	2 (1, 2)	2 (1, 2)	<0.5 (<bg, 1)<="" td=""><td>3 (2, 4)</td></bg,>	3 (2, 4)	
Volatile Organic Compounds										
benzene	1.18 (1.08, 1.28)	0.43 (0.4, 0.47)	0.05 (0.04, 0.06)	0.42 (0.23, 0.68)	0.42 (0.21, 0.56)	0.12 (0.1, 0.15)	0.26 (0.14, 0.43)	0.3 (0.22, 0.37)	0.18 (0.11, 0.3)	
ethylbenzene	0.09 (0.08, 0.09)	0.06 (0.05, 0.06)	0.02	0.01 (0.01, 0.02)	0.02	0.01 (0.01, 0.01)	0.02	0.03	0.02	
m+p-xylenes	0.17	0.08	0.07	0.05	0.02		0.06	0.09	0.06	
o-xylene	0.12	0.03	0.03	0.02	0.01	0.01	0.03	0.05	0.03 (
toluene	0.23	0.20	0.05	0.09	0.09	0.06	0.09	0.11	0.02, 0.03)	
	(0.2, 0.25)	(0.2, 0.21)	(0.03, 0.06)	(0.07, 0.11)	(0.05, 0.12)	(0.05, 0.07)	(0.03, 0.17)	(0.08, 0.13)	(0.05, 0.1)	
0 E alian a thuil	0.01	0.05		Carbo	onyis	0.01	0.00	0.01		
2,5-dimetriyi benzaldehyde	(0.01, 0.01)	(0.05 (0.05)	<lod< td=""><td><lod< td=""><td>(0.04, 0.07)</td><td>(0.01, 0.01)</td><td>(0.01, 0.29)</td><td>(0, 0.01)</td><td><lod< td=""></lod<></td></lod<></td></lod<>	<lod< td=""><td>(0.04, 0.07)</td><td>(0.01, 0.01)</td><td>(0.01, 0.29)</td><td>(0, 0.01)</td><td><lod< td=""></lod<></td></lod<>	(0.04, 0.07)	(0.01, 0.01)	(0.01, 0.29)	(0, 0.01)	<lod< td=""></lod<>	
acetaldehyde	0.14 (0.13, 0.15)	0.88 (0.26, 1.21)	0.09 (0.09, 0.09)	0.14 (0.08, 0.24)	0.20 (0.04, 0.38)	0.25 (0.19, 0.3)	0.42 (0.29, 0.49)	0.38 (0.29, 0.46)	0.35 (0.33, 0.37)	
acetone	0.11 (0.07, 0.16)	0.27	0.43	0.62	0.04	1.76	0.38 (0.15, 0.72)	0.77 (0.43, 1.24)	14.74 (0.49, 42.88)	
acrolein	<0.005 (<0.005, <0.005)	0.06 (0.04, 0.08)	0.01 (0, 0.02)	0.01 (0, 0.01)	0.03 (0.01, 0.04)	0.01 (0.01, 0.01)	0.01 (0.01, 0.02)	0.01 (0, 0.01)	0.02 (0.01, 0.02)	
benzaldehyde	0.07 (0.06, 0.07)	0.05 (0.02, 0.07)	0.02 (0.01, 0.03)	0.03 (0.02, 0.03)	0.02 (0.01, 0.04)	0.03 (0.02, 0.03)	0.03 (0.03, 0.03)	0.05 (0.04, 0.07)	0.06 (0.04, 0.08)	
butanone	0.04 (0.02, 0.07)	0.09 (0.02, 0.14)	0.03 (0.02, 0.03)	0.05 (0.02, 0.08)	0.01 (0, 0.03)	0.03 (0.03, 0.04)	0.04 (0.02, 0.05)	0.10 (0.05, 0.17)	0.09 (0.04, 0.17)	

 Table B.3 Mean pollutant emissions per mass of fuel.
 Emissions per mass fuel burned, calculated as the total mass of pollutant emitted per test divided by the total amount of startup material mass burned per test, averaged across all the test replicates.

butyralde-	0.01	0.14	0.01	0.03	0.06	0.02	0.02	0.03	0.07
hyde	(0.01, 0.02)	(0.12, 0.16)	(0.01, 0.01)	(0.02, 0.03)	(0.03, 0.08)	(0.01, 0.02)	(0.01, 0.03)	(0.03, 0.04)	(0.06, 0.08)
crotonalde-	0.01	0.07	0.01	0.01	0.02	0.02	0.02	0.02	0.03
hyde	(0.01, 0.01)	(0.04, 0.09)	(0.01, 0.02)	(0.01, 0.01)	(0.01, 0.03)	(0.02, 0.02)	(0.02, 0.03)	(0.01, 0.03)	(0.02, 0.04)
formoldobudo	0.3	1.35	0.19	0.38	0.91	0.47	0.62	0.46	0.57
Tormaidenyde	(0.29, 0.34)	(1.02, 1.58)	(0.18, 0.2)	(0.35, 0.42)	(0.82, 1.02)	(0.37, 0.53)	(0.59, 0.64)	(0.41, 0.57)	(0.51, 0.66)
	0.00	0.01	0.05	0.07	0.00	0.40	0.41	0.10	0.0
hexaldehyde	0.08	0.61	0.05	0.07	0.26	0.42	0.41	0.18	0.2
,, ,	(0.07, 0.09)	(0.45, 0.82)	(0.04, 0.07)	(0.04, 0.08)	(0.19, 0.34)	(0.31, 0.52)	(0.37, 0.45)	(0.09, 0.23)	(0.18, 0.21)
isovaler-	0.01	0.12	0.01	0.04	0.06	0.06	0.06	0.05	0.06
aldehyde	(0.01, 0.01)	(0.07, 0.19)	(0.01, 0.02)	(0.01, 0.09)	(0.05, 0.08)	(0.04, 0.08)	(0.03, 0.07)	(0.03, 0.06)	(0.04, 0.07)
un atha avalatio	0.03	0.02	0.02	0.05	0.01	0.02	0.06	0.05	0.13
methacrolein	(0.01, 0.04)	(0.01, 0.04)	(0, 0.03)	(0.04, 0.06)	(0, 0.01)	(0.02, 0.03)	(0.06, 0.07)	(0.02, 0.07)	(0.1, 0.16)
m,p-tolualde-									
hyde	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""><td><lod< td=""></lod<></td></lod<></td></lod<>	<lod< td=""><td><lod< td=""></lod<></td></lod<>	<lod< td=""></lod<>
o-tolualde-	0.07	1.36	0.08	0.06	0.97	0.71	0.76	0.16	0.17
hyde	(0.06, 0.08)	(1.14, 1.47)	(0.04, 0.14)	(0.01, 0.09)	(0.78, 1.1)	(0.48, 0.92)	(0.47, 0.91)	(0.06, 0.25)	(0.14, 0.18)
propionalde-	0.02	0.21	0.18	0.02	0.03	0.05	0.06	0.13	0.09
hyde	(0.02, 0.02)	(0.12, 0.25)	(0.01, 0.5)	(0.01, 0.03)	(0.03, 0.05)	(0.03, 0.06)	(0.03, 0.1)	(0.05, 0.38)	(0.07, 0.1)
	0.01	0.01	0.02	0.01	< 0.005	0.01	0.01	0.03	0.10
valeraldehyde					(<0.005,				(0.08.0.12)
	(0, 0.01)	(0.01, 0.01)	(0.01, 0.03)	(0.01, 0.02)	<0.005)	(0.01, 0.01)	(0.01, 0.01)	(0.02, 0.03)	(0.00, 0.12)

+73 g/kg PM_{2.5} value is an unexplained outlier, when removed, the average is 12.5 g/kg.

EC = elemental carbon; OC = organic carbon; PM_{2.5} = particulate matter mass less than 2.5 microns in diameter.

<BG = measured concentration was below mean background concentration, therefore emissions factors could not be calculated. Background concentrations were: methane: 12 mg/m³; PM_{2.5}: 270 µg/m³; EC: 90 µg C/m³; OC: 94 µg C/m³.

<LOD = below limit of detection.

Table B.4 Modified Combustion Efficiencies. Modified combustion efficiency, a metric that indicates the completeness of the combustion process, was calculated as the ratio of CO_2 divided by the sum of CO and CO_2 each material type. MCE was calculated for each individual startup event within a test, using the background-corrected one-second data for CO and CO_2 emissions, then averaged across the tests; 5th and 95th percentile values within a single startup event (one-second data) were also determined then averaged across the tests.

fuel	mean	median	5 th percentile mean	95 th percentile mean	
footwear	0.988	0.993	0.961	0.997	
food packaging	0.986 0.990		0.964	0.994	
kerosene	0.977	0.984	0.948	0.990	
kindling	0.927	0.933	0.864	0.983	
newspaper	0.952	0.966	0.862	0.991	
plastic bags	0.981	0.986	0.949	0.994	
rubber	0.978	0.988	0.926	0.994	
fabric	0.976	0.987	0.927	0.995	
wood shims	0.968	0.974	0.928	0.990	



Figure B.1 Experimental Setup for Emissions Characterization Tests. Flow rates are shown for the main exhaust plenum and each individual sampling line/instrument.



Fuel Type

Figure B.2 Emissions (g) per Mass (kg) of Fuel Burned for Select Pollutants. Each circle represents the calculated average emissions per mass of fuel for an individual test. Materials are ordered from left to right by highest to lowest mean PM_{2.5} mass.



Figure B.3 Comparison of the Relative Composition of PM2.5 Emitted across Different Startup Materials (per Mass Fuel Basis). Emissions in grams of pollutant per kilogram of fuel burned. Main plot shows total emissions; plot inset shows the lower emissions portions to emphasize values for lesser-emitting materials. Bar height shows total PM_{2.5} mass. Elemental carbon (EC) and organic carbon (OC) components are shown in black and green (respectively) and total PM_{2.5} mass, which encompasses EC and OC emissions, is shown in gray. The lighter grey bar shows PM_{2.5} footwear emissions with the outlier included.



Figure B.4 Comparison of the Relative Composition of BTEX Emitted across Different Startup Materials (per Mass Fuel Basis). Emissions in grams of pollutant kilogram of fuel burned. Main plot show total emissions; plot inset shows the lower emissions portions to emphasize values for lesser-emitting materials. The stacked bar height represents the total BTEX emissions.



Figure B.5 Comparison of the Relative Composition of PM2.5 Emitted across Different Startup Materials (per Startup Event Basis). Emissions in milligrams of pollutant per startup event. Inset highlights results for lower-emitting materials. Bar height shows total PM_{2.5} mass. Elemental carbon (EC) and organic mass (OM) components are shown in black and purple (respectively) and total PM_{2.5} mass, which encompasses EC and OC emissions, is shown in gray. The lighter grey bar shows PM2.5 footwear emissions with the outlier included. The lighter grey bar shows PM2.5 footwear emissions with the outlier included. Particulate organic mass was estimated from the organic carbon using ratios of total organic mass to organic carbon mass of 1.2 for hydrocarbon-based materials (kerosene, footwear, food packaging, inner tubes, and plastic bags) and 1.5 for wood and cellulose-based materials (wood shims, kindling, newspaper).



Figure B.6 Comparison of the Relative Composition of PM2.5 Emitted across Different Startup Materials (per Mass Fuel Basis). Emissions in milligrams of pollutant per kilogram of fuel burned. Main plot shows total emissions; plot inset shows the lower emissions portions to emphasize values for lesser-emitting materials. Bar height shows total PM_{2.5} mass. Elemental carbon (EC) and organic mass (OM) components are shown in black and purple (respectively) and total PM_{2.5} mass, which encompasses EC and OC emissions, is shown in gray. The lighter grey bar shows PM_{2.5} footwear emissions with the outlier included. Particulate organic mass was estimated from the organic carbon using ratios of total organic mass to organic carbon mass of 1.2 for hydrocarbon-based materials (kerosene, footwear, food packaging, inner tubes, and plastic bags) and 1.5 for wood and cellulose-based materials (wood shims, kindling, newspaper).

APPENDIX C: SUPPLEMENTAL MATERIAL FOR CHAPTER 5 (BLOOD PRESSURE)

Additional Details on Study Methods

Overall Design

Each participant received six exposure treatments (a clean air control and five levels of air pollution). There was a washout period between treatments that was typically two weeks but up to six weeks by design. Makeups of missed sessions were conducted within ten days to 14 weeks after the last scheduled treatment. Our study followed a Williams design, which is a Latin square crossover design that is balanced across treatments and first-order carry-over effects (Williams 1949). We specified six unique treatment sequences. Each sequence contained all six treatments administered in a unique order across the participant's six study sessions. Across all sequence groups, each treatment appeared once in each of the six 'periods' (visit numbers) of the treatment orders (e.g., first through last assigned study session) and was both followed by and preceded by each other treatment an equal number of times. This design controls for and time-variant personal level factors as well as is robust to time variant factors that might differ from one study session to the next and impacts of the treatment orders. Participants were blinded to the sequence they were placed in.

We conducted the study in three rounds; in each round, two sequence groups (8 participants each) completed their full set of six treatments on alternating weeks. Additionally, within each sequence group, we divided participants into two subgroups (4 participants each) who completed their session on different days of the week (Mondays versus Wednesdays). For the four participants who completed their study sessions on the same day, we staggered start times by 30 minutes. Participants who missed a scheduled study session (due to illness or unforeseen conflict) were allowed to make up the missed treatment at the end of the sequence.

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Recruitment and Screening Process

Individuals interested in participating in the study completed a screening questionnaire to determine potential eligibility. Individuals who appeared to meet criteria were asked to attend an in-person screening exam. At the screening exam, medical staff measured the individual's height, weight, and blood pressure. A physician reviewed the recruitment questionnaire with the individual, conducted a physical exam, and reviewed the individual's medical history, including family history of cardiovascular and respiratory disease. Additionally, the individual performed an electrocardiogram, spirometry test, and a blood draw for analysis of complete blood count, comprehensive metabolic panel, lipid levels, and serum ferratin. A physician reviewed all results from the screening exam to make a final determination of eligibility for the study.

Additionally, individuals received a tour of the study facility at the time of their screening exam. The goal of the tour was to familiarize potential participants with the exposure facility, protocols, and staff, to reduce drop-out rates and alleviate potential stress-related reactions to the contrived exposure experience during their participation. During the tour, study staff showed individuals examples of cookstoves such as those used for the treatments, explained how the exposure chamber worked and safety features of the chamber, and described what the participant experience was like during exposure periods and throughout the health measurements. Individuals were able to see the exposure chamber and, if not in use during the time of their screening exam, enter the exposure chamber.

Clinical Health Measurements

A set of cardiovascular and pulmonary health measurements were taken at four time points. It took approximately one hour to complete a set of health measurements. Measurements were taken in the same order at each time point and across each study session, as indicated below:

1. Apply Holter monitor for heart rate variability (HRV) measurement (at the baseline timepoint only; the holter remained in place for the first three time points).

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- 2. Rest period in supine position lasting twenty minutes during time points one, two, and three (HRV data collected) and ten minutes during time point 4.
- Blood pressure and pulse wave analysis (Augmentation Index) using SphygmoCor device (participant remained in supine position).
- Pulse wave velocity using SphygmoCor device (participant remained in supine position).
- 5. Spirometry using Easy-on device (participant in seated position).
- 6. Venous blood draw for later analysis of inflammatory markers, complete blood count, and lipid levels.

Stove Treatments

Stove makes/models were as follows:

- Liquid petroleum gas [LPG] stove: Classic Single Burner 25000 BTU, WokSmith, China),
- Gasifier: Ace 1 Gasifier, African Clean Energy (Pty) Ltd, Lesotho
- Fan-powered rocket elbow: HomeStove, Biolite, USA
- Rocket elbow: G3300, Envirofit International, USA)
- Traditional three stone fire: open fire, bricks in U-shape used to contain fuel

Additional Results

Study Completion/Missing Data

Of the 26 participants who missed study sessions, 12 missed one session. Approximately half of the missed study sessions were due to scheduling conflicts that arose after a participant enrolled in the study; one quarter were due to illnesses on scheduled study dates, and one quarter were due to the participants being enrolled in the study late, after the rest of their sequence cohort had completed the first study session.

Four participants withdrew from the study prior to completing six study sessions. Additionally, errors with data logging and our exposure chamber operation resulted in the loss of data relevant to single sessions which were not repeated for five participants. Finally, one participant completed six study sessions, however after the first session, we switched from left-side measurements to right-side to accommodate a medical implant in their left arm; their first session was therefore censored from the dataset. More study sessions were missing from the LPG, gasifier, and fan rocket treatments (44 of the 48 participants completed these treatments) than other treatments (rocket elbow: 45, three stone fire and control: 47). Within the sessions completed, 11 individual data points are missing. Reasons for missing individual data points included scheduling conflicts for participants that resulted in them leaving a study day without completing the three-hour or 24-hour follow-up time point or a data recording error that resulted in loss of data.

Health Measurement Timing

The three sets of post-treatment health measurements were scheduled to start immediately post-exposure, three hours post exposure, and 24 hours post-exposure. The average times of blood pressure measurements were 30 minutes (standard deviation: 4.2 minutes), 3 hours and 26 minutes (standard deviation: 4.8 minutes), and 24 hours and 13 minutes (standard deviation: 30 minutes) post-exposure. The mean of the range in measurement times per person across the six study sessions were 0.17 hrs, 0.19 hrs, and 0.63 hrs for immediately, three hours, and 24 hours post-exposure, respectively.

Potential Confounders

Alcohol Consumption

Participants were asked to avoid alcohol starting 24 hours before the start of each study session and continue throughout the end of the 24-hour health measurements. Reported alcohol consumption among participants was low during the 24 hours before each study session (8/271 reported occurrences, 3%; see Table C.1) and during the study sessions (3/266 reported occurrences, 1%; see Table C.2). Use of alcohol occurred with a mainly even distribution across the six treatments; use was slightly higher in the 24-hour prior to the rocket elbow treatment (three

out of 45 [7%] vs one out of 44 to 47 [2%] reported alcohol consumption). Thirty-three of the 48 participants did not change their habits regarding pre-study session alcohol use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Thirty-four of the 48 participants did not change their habits regarding during-study session alcohol use across the six study sessions. Univariate models of the association between alcohol use and treatment in the 24-hours prior to the study day, considering a within person random effect, found a non-statistically significant (p > 0.05) increased odds of alcohol use prior to the rocket elbow stove treatment compared to others (3.6, CI 0.3, 39).

Table C.1 Alcohol Consumption by Treatments: 24 Hours before Session Start.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	46	43	43	43	42	46	263
yes	1	1	1	1	3	1	8
total	47	44	44	44	45	47	271

Table C.2 Alcohol Consumption by Treatments: During the Study Session.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	44	43	42	43	44	47	263
yes	2	1	0	0	0	0	3
Not applicable*	1	0	2	1	1	0	5
Total	47	44	44	44	45	47	271

*Not applicable: Participant missed the 24-hour follow-up period/survey.

Caffeine Consumption

Participants were asked not to consume caffeine from 24 hours prior through completion of each study session. Reported caffeine consumption among participants was low during the 24-hours before (24/271 reported occurrences, 9%; see Table C.3) each study session and during the study sessions (13/266 reported occurrences, 5%; see Table C.4). Use of caffeine occurred with a mainly even distribution across the six treatments; use was slightly higher on the second study days when the treatment was LPG compared to others (4 out of 44 [9%] vs one or two out of 44 to 47 [2-5%]. Thirty-one of the 48 participants did not change their habits regarding pre-study session caffeine use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Thirty-one of the 48 participants did not change their habits regarding during-study session caffeine use across the six study sessions. Univariate models of the association between caffeine use and treatment during the study session (between the end of the three-hour measurements through the start of the 24-hour measurements), considering a within person random effect, found a non-significant increased odds of caffeine use for the LPG treatment compared to others (1.4, 95% CI 0.2, 12.4).

Table C.3 Caffeine Consumption by Treatments: 24 Hours before Session Start.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	43	40	40	39	40	45	247
yes	4	4	4	5	5	2	24
Total	47	44	44	44	45	47	271

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	44	40	40	42	42	45	253
yes	2	4	2	1	2	2	13
Not applicable*	1	0	2	1	1	0	5
Total	47	44	44	44	45	47	271

*Not applicable: Participant missed the 24-hour follow-up period/survey.

Medication use

Participants were asked not to use any medications that were not approved by the study physician prior to enrollment, starting 72 hours before the start of each study session and continue

throughout the end of the 24-hour health measurements. Reported medication use among participants was low during the 72-hours before each study session (37/271 reported occurrences, 14%; see Table C.5) and during the study sessions (34/266 reported occurrences, 13%; see Table C.6). Use of medication occurred with even distribution across the six treatments. Twenty-nine of the 48 participants did not change their habits regarding pre-study session medication use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Twenty-seven of the 48 participants did not change their habits regarding their habits regarding during-study session medication use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Twenty-seven of the 48 participants did not change their habits regarding the session medication use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Twenty-seven of the 48 participants did not change their habits regarding the session medication use across the six study sessions.

Table C.5 Medication Use by Treatments: 24 Hours before Session Start.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	43	37	38	39	38	39	234
yes	4	7	6	5	7	8	37
Total	47	44	44	44	45	47	271

Table C.6 Medication Use by Treatments: During the Study Session.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	41	40	34	37	40	40	232
yes	5	4	8	6	4	7	34
Not applicable*	1	0	2	1	1	0	5
Total	47	44	44	44	45	47	271

*Not applicable: Participant missed the 24-hour follow-up period/survey.

Smoke Exposures

Participants were asked to avoid smoke exposures from 24 hours prior through completion of each study session. Reported smoke exposures among participants was low during the 24-hours before each study session (2/271 reported occurrences, less than 1%; see Table C.7) and during the study sessions (3/266 reported occurrences, 1%; see Table C.8). Forty of the 48 participants did not change their habits regarding pre-study session smoke exposures across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Thirty-three of the 48 participants did not change their habits regarding during-study smoke exposures across the six study sessions.

Exposure	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	47	43	44	44	44	47	269
yes	0	1	0	0	1	0	2
Total	47	44	44	44	45	47	271

Table C.7 Smoke Exposures by Treatments: 24 Hours before Session Start.

Table C.8 Smoke Exposures by Treatments: During the Study Session.

Exposure	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	46	44	41	41	44	47	263
yes	0	0	1	2	0	0	3
Not applicable*	1	0	2	2	1	0	5
Total	47	45	49	44	44	47	271

*Not applicable: Participant missed the 24-hour follow-up period/survey.

Mode of Commute

Participants were asked to use the same mode of commute into the facility on each study day. Driving was the most common mode of commute (59% of all trips to the facility for the first study day and 56% of all trips for the second study day involved a car), followed by bike (36% of all trips on the first study day and 31% on the second study day; see Tables S9 and S10). Twenty-five of the 48 participants did not change their habits regarding first day commute mode across the six study sessions (i.e., consistently reported the exact same mode for all six sessions). Twenty-two of the 48 participants did not change their habits regarding the second day commute mode use across the six study sessions.

Mode	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Bike	15	16	17	16	18	14	96
Bike+walk	0	1	1	0	1	0	3
Bus	0	0	0	0	0	0	0
Bus+walk	0	0	1	0	1	0	2
Car	27	24	23	25	24	29	153
Car+walk	3	1	1	1	0	2	8
Walk	2	2	1	1	1	1	8
NA*	0	0	0	1	0	1	2
total	47	44	44	44	45	47	271

Table C.9 Mode of Commute to Facility by Treatments: Before Session Start.

*Not applicable: The participant did not report.

Table	C.10	Mode	of	Commute	to	Facility	by	Treatments:	Prior	to	the	24-Hour	Health
Measu	ireme	ents.				-	-						

Mode	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Bike	14	15	12	14	14	12	81
Bike+walk	0	0	1	0	1	1	3
Bus	1	1	0	2	0	0	4
Bus+walk	2	2	3	2	3	4	16
Car	26	23	24	23	21	29	147
Car+walk	2	1	1	0	2	0	6
Walk	1	1	0	1	2	1	6
NA*	1	1	3	2	2	0	9
Total	47	44	44	44	45	47	271

*Not applicable: The participant did not report or the participant was not present for the 24-hour measurements.

Sleep Quality

Most participants reported getting an "average" amount of sleep (74% for the night before the study session began and 75% for the night before the second study day); the amount of people reporting below-average sleep was less for the second study day than the first (19% for the night before the study session began vs. 10% for the night before the second study day; see Tables S11 and S12). Only 11 of the 48 participants did not change their habits regarding sleep prior to the start of a study session across the six study sessions (i.e., consistently reported the exact same sleep levels for all six sessions). Thirteen of the 48 participants did not change their habits regarding sleep the night before the 24-hour measurements across the six study sessions.

Sleep	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Above average	4	2	4	0	5	4	19
Average	33	36	32	36	32	32	201
Below average	10	6	8	8	8	11	51
Total	47	44	44	44	45	47	271

Table C.11 Sleep Quality by Treatment: Night Prior to Start of Study Session.

Table C.12 S	Sleep Quali	ty by Treati	ment: Night	Prior to the	24-Hour He	alth Measu	rements.

Sleep	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Above average	6	7	6	8	6	7	40
Average	35	34	33	31	32	35	200
Below average	5	3	3	4	6	5	26
Not applicable*	1	0	2	1	1	0	5
Total	47	44	44	44	45	47	271

*Not applicable: Participant missed the 24-hour follow-up period/survey.

Ambient PM_{2.5}

Mean ambient $PM_{2.5}$ in the 24-hours prior to the start of a study day ranged from 4.9 µg/m³ (control) to 9.8 µg/m³ (fan rocket; see Table C.13). Minimum recorded mean $PM_{2.5}$ was 0.9 µg/m³ (three stone fire) and maximum recorded mean $PM_{2.5}$ was 17.6 µg/m³ (fan rocket). As illustrated in Figure C.1, ambient $PM_{2.5}$ was significantly higher for all treatments compared to the control except the gasifier. However, the range of ambient $PM_{2.5}$ overall was determined to be narrow enough to not include this variable in the main model. We also considered average $PM_{2.5}$ levels for the six hours prior to the start of a study day; results were similar.

Table C.13 Ambient PM_{2.5} Levels* by Treatment: 24 Hours before Session Start.

PM _{2.5}	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone
mean	4.9	7.2	5.3	9.8	6.4	6.6
min	1.5	2.9	1.0	2.0	2.6	0.9
max	9.8	18.8	11.2	17.6	10.6	12.7

* 24-hour average in μg/m³



Figure C.1 Effect Estimates and 95% Confidence Intervals for Mean Difference in Ambient PM_{2.5} Levels for Stove Treatments Compared to Control. Effect is the absolute difference in ambient PM_{2.5} 24-hour average in μ g/m³.

Ambient CO

Mean ambient CO in the 24-hours prior to the start of a study day ranged from 0.25 (rocket elbow) to 0.35 ppm (three stone fire; see Table C.14). Minimum recorded mean CO was 0.13 ppm (LPG) and maximum recorded mean CO was 0.70 ppm (three stone fire). As illustrated in Figure C.2, ambient CO was significantly higher for the LPG and three stone fire treatments compared to the control. However, the range of ambient CO overall was determined to be narrow enough to not include this variable in the main model. We also considered average CO levels for the six hours prior to the start of a study day; results were similar.

Table C.14 Ambient CO Levels* by Treatment: 24 Hours before Session Start.

СО	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone
mean	0.27	0.31	0.28	0.29	0.25	0.35
min	0.16	0.13	0.19	0.17	0.17	0.17
max	0.46	0.48	0.45	0.50	0.44	0.70

* 24-hour average in ppm



Figure C.2 Effect Estimates and 95% Confidence Intervals for Mean Difference in Ambient CO Levels for Stove Treatments Compared to Control. Effect is the absolute difference in ambient CO 24-hour average in ppm.

Ambient Temperature

Mean temperature in the 24-hours prior to the start of a study day ranged from 6.0 °C (43 °F; three stone fire) to 15.9 °C (61°F; fan rocket). Minimum recorded mean temperature was -8.5 °C (17 °F; rocket elbow) and maximum recorded mean temperature was 24.2 °C (76 °F; rocket elbow; see Table C.15). As illustrated in Figure C.3, temperature was significantly higher for the LPG, fan rocket, and rocket elbow treatments compared to the control. However, the range of temperatures overall was determined to be narrow enough to not include this variable in the main model.

Temp	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone
mean	7.0	10.9	8.2	15.9	13.3	6.0
min	-7.3	2.9	0.5	4.6	-8.5	-3.1
max	20.2	23.9	14.0	22.3	24.2	15.3

Table C.15 Mean Temperature (°C) by Treatment: 24 Hours before Study Session.



Figure C.3 Effect Estimates and 95% Confidence Intervals for Mean Difference in Ambient Temperature for Stove Treatments Compared to Control. Effect is the absolute difference in ambient temperature 24-hour average in $^{\circ}C$.

Alternative Models

Baseline blood pressures were lowest for the fan-rocket treatment (115.0/68.2 mmHg) followed by the control (115.2/68.6 mmHg) and highest for the three stone fire (117.0/70.2 mmHg). As such, inclusion of a baseline term in the model is justified.

Model with more structured study design parameters, in sequence data only (no makeups)

We developed a mixed-effect model that considered more structured study design parameters relevant to our Williams square, such as each individual's assigned sequence group and the day of week (Monday vs. Wednesday), and only included data that was collected within the intended sequence. Results of this model for systolic pressure indicate no significance (p < 0.05) of the various fixed effect "design" terms (day of week, sequence group, or the sequence/day interaction term) (Table C.15; Figure C.3). There are no differences in main effect estimates compared to the main model with all data (Table C.15; Figure C.3).

Main model, in sequence only (no makeups)

We ran the main model but on a data set that excluded data collected outside of the intended treatment sequence. Results of this model for systolic pressure indicate no differences in main effect estimates compared to the main model with all data (Table C.15; Figure C.3).

Table C.16 Comparison of Model Results for Three Model Options: Effect Estimates and 95% Confidence Intervals for all Model Parameters.

Paramete r		MAIN MODEL		MAIN MO	DDEL, IN SEQUE	ENCE DATA	DESIGN MODEL, IN SEQUENCE DATA			
	l			SYSTOLIC	PRESSURE					
	immediately post-exposure	three hours post-exposure	24 hours post-exposure	immediately post-exposure	three hours post-exposure	24 hours post-exposure	immediately post-exposure	three hours post-exposure	24 hours post-exposure	
Random - date	0.7	0.2	0.0	0.8	0.5	0.0	0.9	0.6	0.0	
Random – ID (main model) or day:seque nce:id (design model)	5.1	5.7	7.1	5.4	5.55	7.3	5.7	5.9	7.3	
Residual	5.2	5.4	5.2	4.7	5.46	4.8	4.6	5.3	4.7	
Intercept	62.5 (50, 75)	71.5 (58.5, 84.6)	83.1 (69.5, 96.7)	65.3 (53.1, 77.6)	69.7 (56.1, 83.4)	87.8 (74.3, 101.2)	74.1 (60.1, 88.2)	84.1 (68.4, 99.7)	96.0 (80.5, 111.3)	
baseline	0.5 (0.4, 0.6)	0.4 (0.3, 0.5)	0.3 (0.2, 0.4)	0.4 (0.3, 0.5)	0.4 (0.28, 0.51)	0.2 (0.1, 0.3)	0.4 (0.3, 0.5)	0.3 (0.2, 0.4)	0.2 (0.1, 0.3)	
lpg	-0.2 (-2.5, 2.0)	1.1 (-1.1, 3.3)	3.1 (1.0, 5.3)	0.0 (-2.1, 2.2)	1.6 (-0.8, 4.1)	2.4 (0.3, 4.6)	0.0 (-2.2, 2.2)	1.6 (-0.8, 4.1)	2.3 (0.2, 4.5)	
gasifier	-1.81 (-4.0, 0.4)	1.0 (-1.2, 3.2)	2.3 (0.1, 4.5)	-1.0 (-3.3, 1.3)	1 (-1.61, 3.61)	2.2 (-0.1, 4.5)	-1.0 (-3.3, 1.3)	0.95 (-1.6, 3.5)	2.1 (-0.2, 4.4)	
fan rocket	-0.4 (-2.7, 1.8)	-1.8 (-4.0, 0.5)	2.5 (0.4, 4.7)	-0.5 (-2.7, 1.7)	-1.35 (-3.9, 1.19)	2.0 (-0.2, 4.2)	-0.6 (-2.8, 1.6)	-1.5 (-4.1, 1.0)	1.9 (-0.3, 4.1)	
rocket elbow	-0.6 (-2.8, 1.6)	-0.5 (-2.7, 1.7)	-0.1 (-2.2, 2.1)	-1.3 (-3.5, 0.9)	-0.31 (-2.78, 2.17)	-0.2 (-2.4, 1.9)	-1.3 (-3.4, 0.9)	-0.3 (-2.7, 2.2)	-0.3 (-2.4, 1.9)	
three stone	-2.3 (-4.5, -0.1)	-2.1 (-4.3, 0.2)	2.4 (0.3, 4.5)	-1.7 (-3.8, 0.5)	-1.7 (-4.1, 0.7)	2.6 (0.4, 4.7)	-1.6 (-3.7, 0.5)	-1.5 (-3.9, 0.9)	2.6 (0.5, 4.7)	
Day- Wed nesday							-0.0 (-8.6, 8.5)	0 (-9.0, 9.0)	3.4 (-7.2, 14.0)	
Sequence b							6.4 (-2.1, 14.9)	4.4 (-4.6, 13.3)	8.4 (-2.1, 19.0)	
Sequence c							-1.8 (-10.2, 6.7)	-4.5 (-13.3, 4.3)	-3.9 (-14.3, 6.6)	
Sequence d							-1.9 (-10.3, 6.5)	-1.2 (-10.0, 7.7)	0.1 (-10.3, 10.6)	
Sequence e							-3.5 (-12.0, 5.0)	-6.8 (-15.7, 2.1)	-2.1 (-12.6, 8.4)	
Sequence f							-1.4 (-9.9, 7.0)	-4.5 (-13.4, 4.4)	-5.6 (-16.1, 4.9)	

dayWedn esday: sequence b							-10.2 (-22.4, 2.0)	-12.0 (-24.9, 0.9)	-15.7 (-30.7, -0.6)			
dayWedn esday: sequence c							1.8 (-10.2, 13.8)	4.6 (-8.0, 17.2)	2.7 (-12.2, 17.5)			
dayWedn esday: sequence d							-4.0 (-16.2, 8.2)	-6.6 (-19.5, 6.3)	-12.9 (-27.9, 2.2)			
dayWedn esday: sequence e							-0.2 (-12.3, 11.8)	2.0 (-10.7, 14.6)	-5.0 (-19.9, 9.8)			
dayWedn esday: sequencef							-4.1 (-16.2, 8.0)	-2.5 (-15.2, 10.2)	-3.6 (-18.5, 11.3)			
	DIASTOLIC PRESSURE											
Random - date	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0			
Random – ID (main model) or day:seque nce:id (design model)	3.3	3.5	4.08	3.4	3.42	4.08	2.93	3.3	4.0			
Residual	3.6	4.2	4.5	3.5	4.2	4.01	3.56	4.2	4.0			
Intercept	35.3 (28.4, 42.2)	33.7 (25.8, 41.6)	44.8 (36.0, 53.6)	36.7 (30.0, 44.0)	33.6 (25.4, 41.9)	46.2 (37.8, 54.6)	38.1 (30.1, 46.1)	35.6 (26.2, 45.0)	50.0 (40.0, 60)			
baseline	0.5 (0.4, 0.6)	0.5 (0.4, 0.6)	0.4 (0.2, 0.5)	0.5 (0.4, 0.6)	0.5 (0.4, 0.6)	0.3 (0.2, 0.5)	0.5 (0.4, 0.6)	0.5 (0.4, 0.6)	0.3 (0.2, 0.4)			
lpg	-0.7 (-2.2, 0.8)	-0.0 (-1.7, 1.7)	0.3 (-1.6, 2.2)	-0.4 (-2.0, 1.2)	0.0 (-1.9, 1.9)	-0.6 (-2.4, 1.2)	-0.5 (-2.1, 1.1)	0.02 (-1.9, 1.9)	-0.8 (-2.6, 1.0)			
gasifier	-0.8 (-2.2, 0.74)	0.3 (-1.5, 2.0)	-0.4 (-2.3, 1.5)	-0.5 (-2.2, 1.2)	-0.4 (-2.4, 1.6)	-0.8 (-2.7, 1.2)	-0.6 (-2.2, 1.1)	-0.4 (-2.4, 1.6)	-0.8 (-2.7, 1.1)			
fan rocket	-0.13 (-1.63, 1.4)	-0.41 (-2.2, 1.3)	-0.1 (-1.9, 1.8)	0.1 (-1.5, 1.7)	-0.4 (-2.3, 1.5)	-1.2 (-3.0, 0.7)	0 (-1.6, 1.6)	-0.5 (-2.4, 1.5)	-1.3 (-3.2, 0.5)			
rocket elbow	0.4 (-1.1, 1.8)	0.2 (-1.5, 1.9)	-1.7 (-3.6, 0.2)	0.1 (-1.5, 1.7)	0.1 (-1.9, 2.0)	-2 (-3.8, -0.2)	0.1 (-1.5, 1.7)	0.2 (-1.7, 2.1)	-2.0 (-3.8, -0.2)			
three stone	-0.9 (-2.3, 0.6)	-0.8 (-2.5, 0.9)	0.8 (-1.0, 2.7)	-0.7 (-2.3, 0.9)	-0.9 (-2.8, 1.0)	0.5 (-1.3, 2.3)	-0.7 (-2.3, 0.8)	-0.9 (-2.7, 1.0)	0.5 (-1.2, 2.3)			

Day- Wed				-1.5 (-6.2, 3.2)	-0.5 (-5.9, 4.9)	1.2 (-4.9, 7.3)
Sequence						
b				2.1 (-2.6, 6.7)	1.0 (-4.3, 6.2)	4.2 (-1.9, 10.3)
Sequence				20(7517)	22(85.21)	33(03.27)
С				-2.9 (-7.3, 1.7)	-3.2 (-0.3, 2.1)	-3.3 (-9.3, 2.7)
Sequence				-4.1 (-8.8,	-04(-5649)	-08(-6852)
d				0.47)	0.1 (0.0, 1.0)	0.0 (0.0, 0.2)
Sequence				-2.9 (-7.6, 1.7)	-4.0 (-9.3, 1.3)	-0.9 (-6.9. 5.1)
e		 	 	- (- (; - /	
Sequence				-3.5 (-8.2, 1.1)	-1.9 (-7.2, 3.3)	-1.6 (-7.7, 4.4)
T alas XA(a alas						
daywedn				20/105		70/150
esuay.				-3.9 (-10.5,	-2.4 (-9.9, 5.0)	-7.3 (-15.9,
h				2.7)		1.0)
davWedn						
esday:				/ /		
sequence				2.0 (-4.5, 8.6)	4.2 (-3.3, 11.7)	1.4 (-7.1, 10.0)
C						
dayWedn						
esday:				17 (50 83)	-2.9 (-10.5,	-7.7 (-16.3,
sequence				1.7 (-5.0, 6.5)	4.7)	0.9)
d						
dayWedn						
esday:				-2.4 (-9.0.4.2)	-1.3 (-8.9, 6.2)	-3.2 (-11.7,
sequence				(, .)		5.4)
e						
dayWedn						-2.3 (-10.9,
esday:				0.34 (-6.3, 6.9)	-0.9 (-8.4, 6.7)	6.2)
sequencer						



Immediate Post
 Three Hour Post
 24 Hour Post

Figure C.4 Effect Estimates and 95% Confidence Intervals for Mean Difference in Systolic Pressure (mmHg) for Stove Treatments Compared to Control Across the Three Model Types.

Main model, remove when exposure value outside narrow range of target

We ran the main model excluded data from study sessions where the exposure mean was

outside of a narrowed range around the target value. The narrowed ranges were:

- Control: less than 5 μg/m³
- LPG: 5-15 μg/m³
- Gasifier: 20-60 µg/m³
- Fan rocket: 75-125 μg/m³
- Rocket elbow: 175-300 μg/m³
- Three stone fire: 350-600 μg/m³

Results indicated no considerable differences between the estimates for the treatment effects between this model and the main model (see Figure C.5).



◆ Immediate Post ◆ Three Hour Post ◆ 24 Hour Post

Figure C.5 Effect Estimates and 95% Confidence Intervals for Mean Difference in Systolic Pressure (mmHg) for Stove Treatments Compared to Control: Comparison of Main Model to Model with Exposure Outliers Removed.

Three Stone Fire as Reference

Effect estimates and 95% confidence intervals for the change in blood pressure post-exposure for each stove treatment compared to the filtered-air control treatment are presented in Table C.17 and Figure C.6.

At the immediate post-exposure measurement, systolic pressure was higher for the filtered air compared to the three stone fire (500 μ g/m³ PM_{2.5}) (2.29 mmHg, 95% CI 0.1, 4.48 mmHg).

There were no significant associations for the various other stove treatments compared to the three stone fire at this time point. However, effect estimates are suggestive of higher systolic pressure for all other stove treatments compared to the three stone fire (ranging from 0.48 to 2.07 mmHg), indicating a trend wherein the three stone fire suppresses systolic blood pressure compared to all other (lower-PM_{2.5} level) treatments and filtered air.

At three hours post exposure, systolic pressure remained lower compared to the three stone fire for all lower-PM_{2.5} level stove treatments and the filtered air control. The largest effect was seen for the LPG stove (10 μ g/m³ PM_{2.5}; 3.15 mmHg, 95% CI 0.90-5.39) followed by the gasifier (35 μ g/m³ PM_{2.5}; 3.04 mmHg, 95% CI 0.79, 5.30), both of which had larger effect estimates at this time point than the previous immediate post-exposure time point. Effect estimates for the other treatments were positive but lower than at the immediate post-exposure time point.

At 24-hours post-exposure, systolic blood pressure was significantly lower compared to the three stone fire among the filtered air control (-2.41 mmHg, 95% CI -4.53, -0.28) and the rocket elbow treatment (-2.48 mmHg, 95% CI -4.68, -0.33). The other treatments (LPG, gasifier, and fan rocket) showed no trend of difference compared to the three stone fire (effect estimates -0.11 to 0.70 mmHg; not statistically significant).

Immediately post-exposure, diastolic pressure was higher for all treatments compared to the three stone fire (effect estimates of 0.12 to 1.22 mmHg); at three hours post exposure, diastolic pressure remained increased, with similar though still non-significant effect estimates (0.37 to 1.03 mmHg). At 24 hours post-exposure, trends were reversed, with effect estimates suggesting slight lower blood pressure for all other $PM_{2.5}$ -level stove treatments and the filtered air control compared to the three stone fire (significant only for the rocket elbow treatment at 250 µg/m³ $PM_{2.5}$; -2.53 mmHg, 95% CI -4.40, -0.65). Mean pressure followed a similar pattern as diastolic pressure, with higher pressure compared to the three stone fire for all treatments at the immediate post-exposure and three hour post-exposure time points followed by lower pressure at the 24 hour post-exposure time point.

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Effect Estimate (05% OI) [mmHz difference estimated to three									
	Effect Estimate (95% CI) [mmHg difference compared to three								
	stone tirej								
	Immediate	3 hour	24 hour						
Treatment	post-exposure	post-exposure	post-exposure						
Systolic									
filtered air 2.29 (0.10, 4.48) 2.05 (-0.15, 4.25) -2.41 (-4.53, -									
LPG	2.07 (-0.16, 4.30)	3.15 (0.9, 5.39)*	0.70 (-1.45, 2.85)						
gasifier	0.48 (-1.74, 2.71)	3.04 (0.79, 5.3)*	-0.11 (-2.31, 2.09)						
fan rocket	1.85 (-0.39, 4.09)	0.29 (-2, 2.58)	0.14 (-2.04, 2.32)						
rocket elbow	1.71 (-0.5, 3.93)	1.58 (-0.65, 3.81)	-2.48 (-4.63, -0.33)						
	Diastolic								
filtered air	0.87 (-0.60, 2.34)	0.77 (-0.93, 2.48)	-0.81 (-2.67, 1.04)						
LPG	0.20 (-1.29, 1.7)	0.76 (-0.98, 2.5)	-0.49 (-2.37, 1.38)						
gasifier	0.12 (-1.37, 1.61)	1.03 (-0.72, 2.77)	-1.18 (-3.1, 0.73)						
fan rocket	0.74 (-0.77, 2.24)	0.37 (-1.4, 2.14)	-0.87 (-2.78, 1.03)						
rocket elbow	1.22 (-0.27, 2.7)	1.00 (-0.72, 2.73)	-2.53 (-4.40, -0.65)						
Mean									
filtered air	1.32 (-0.16, 2.81)	1.26 (-0.38, 2.89)	-1.36 (-3.10, 0.39)						
LPG	0.86 (-0.65, 2.37)	1.5 (-0.16, 3.16)	-0.2 (-1.96, 1.56)						
gasifier	0.14 (-1.37, 1.65)	1.68 (0.01, 3.35)	-0.84 (-2.63, 0.95)						
fan rocket	1.12 (-0.40, 2.65)	0.21 (-1.48, 1.91)	-0.53 (-2.32, 1.26)						
rocket elbow	1.20 (-0.30, 2.7)	1.15 (-0.5, 2.8)	-2.67 (-4.43, -0.91)						

Table C.17 Effect Estimates and 95% Confidence Intervals for Mean Difference in Blood Pressure (mmHg) for Treatments Compared to Three Stone Fire.

All models were adjusted for baseline (pre-exposure) blood pressure.



Figure C.6 Effect Estimates and 95% Confidence Intervals for Mean Difference in Blood Pressure (mmHg) for Treatments Compared to Three Stone Fire.

APPENDIX D. SUPPLEMENTAL MATERIAL FOR CHAPTER 6 (SPIROMETRY)

Additional Details on Study Methods

Overall Design

Each participant received six exposure treatments (a clean air control and five levels of air pollution, described in the main text). There was a washout period between treatments that was typically two weeks but up to six weeks by design. Makeups of missed sessions were conducted within ten days to 14 weeks after the last scheduled treatment. Our study followed a Williams design, which is a Latin square crossover design that is balanced across treatments and first-order carry-over effects (Williams 1949). We specified six unique treatment sequences. Each sequence contained all six treatments administered in a unique order across the participant's six study sessions. Across all sequence groups, each treatment appeared once in each of the six 'periods' (visit numbers) of the treatment orders (e.g., first through last assigned study session) and was both followed by and preceded by each other treatment an equal number of times. This design controls for and time-variant personal level factors as well as is robust to time variant factors that might differ from one study session to the next and impacts of the treatment orders. Participants were blinded to the sequence they were placed in.

We conducted the study in three rounds; in each round, two sequence groups (8 participants each) completed their full set of six treatments on alternating weeks. Additionally, within each sequence group, we divided participants into two subgroups (4 participants each) who completed their session on different days of the week (Mondays versus Wednesdays). For the four participants who completed their study sessions on the same day, we staggered start times by 30 minutes. Participants who missed a scheduled study session (due to illness or unforeseen conflict) were allowed to make up the missed treatment at the end of the sequence.

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Recruitment and Screening Process

Individuals interested in participating in the study completed a screening questionnaire to determine potential eligibility. Individuals who appeared to meet criteria were asked to attend an in-person screening exam. At the screening exam, medical staff measured the individual's height, weight, and blood pressure. A physician reviewed the recruitment questionnaire with the individual, conducted a physical exam, and reviewed the individual's medical history, including family history of cardiovascular and respiratory disease. Additionally, the individual performed an electrocardiogram, spirometry test, and a blood draw for analysis of complete blood count, comprehensive metabolic panel, lipid levels, and serum ferratin. A physician reviewed all results from the screening exam to make a final determination of eligibility for the study.

Additionally, individuals received a tour of the study facility at the time of their screening exam. The goal of the tour was to familiarize potential participants with the exposure facility, protocols, and staff, to reduce drop-out rates and alleviate potential stress-related reactions to the contrived exposure experience during their participation. During the tour, study staff showed individuals examples of cookstoves such as those used for the treatments, explained how the exposure chamber worked and safety features of the chamber, and described what the participant experience was like during exposure periods and throughout the health measurements. Individuals were able to see the exposure chamber and, if not in use during the time of their screening exam, enter the exposure chamber.

Clinical Health Measurements

A set of cardiovascular and pulmonary health measurements were taken at four time points. It took approximately one hour to complete a set of health measurements. Measurements were taken in the same order at each time point and across each study session, as indicated below:

1. Apply Holter monitor for heart rate variability (HRV) measurement (at the baseline timepoint only; the holter remained in place for the first three time points).

- 2. Rest period in supine position lasting twenty minutes during time points one, two, and three (HRV data collected) and ten minutes during time point 4.
- Blood pressure and pulse wave analysis (Augmentation Index) using SphygmoCor device (participant remained in supine position).
- Pulse wave velocity using SphygmoCor device (participant remained in supine position).
- 5. Spirometry using Easy-on device (participant in seated position).
- 6. Venous blood draw for later analysis of inflammatory markers, complete blood count, and lipid levels.

Stove Treatments

Stove makes/models were as follows:

- Liquid petroleum gas [LPG] stove: Classic Single Burner 25000 BTU, WokSmith, China),
- Gasifier: Ace 1 Gasifier, African Clean Energy (Pty) Ltd, Lesotho
- Fan-powered rocket elbow: HomeStove, Biolite, USA
- Rocket elbow: G3300, Envirofit International, USA)
- Traditional three stone fire: open fire, bricks in U-shape used to contain fuel

Additional Results

Study completion/missing data

One participant was removed from the study after three sessions due to consistently low lung function at baseline. The participant had met eligibility criteria regarding lung function at screening (spirometry values greater than 70% of the predicted value for the age/gender), however, on study days was consistently achieving values between 65-75% at the baseline (pre-exposure) measurements. As such, the study physician determined that removal of the participant from the study was advised. The data from this participant's three completed sessions was censored from the spirometry dataset.

Within the sessions completed, six individual data points were not collected due to scheduling conflicts for participants that resulted in them leaving a study day without completing the three-hour or 24-hour follow-up time point. An additional nine data points were collected but censored from the dataset because they did not meet minimum quality criteria as established by the ATS/ERS and were not approved for use by the study pulmonologist.

Of the 26 participants who missed scheduled study sessions, 12 missed only one session. Approximately half of the missed study sessions were due to scheduling conflicts that arose after a participant enrolled in the study; one quarter were due to illnesses on scheduled study dates, and one quarter were due to the participants enrolling in the study late, after the rest of their sequence cohort had completed the first study session. We attempted to schedule makeups for all missed sessions. Three participants withdrew from the study for personal reasons prior to completing six study sessions, resulting in nine sessions not made up. Additionally, errors with data logging and our exposure chamber operation resulted in the loss of data relevant to single sessions which were not repeated for five participants.

Health Measurement Timing

The three sets of post-treatment health measurements were scheduled to start immediately post-exposure, three hours post exposure, and 24 hours post-exposure. The average time of spirometry measurements were 0.63 hours (standard deviation 0.09 hours), 3.55 hours (standard deviation 0.49 hours) post-exposure.

The mean of the range in measurement times per person across the six study sessions were 0.21 hours, 0.21 hours, and 0.65 hours for immediately, three hours, and 24 hours post-exposure, respectively.

Potential Confounders

Alcohol Consumption

Participants were asked to avoid alcohol starting 24 hours before the start of each study session and continue throughout the end of the 24-hour health measurements. Reported alcohol

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consumption among participants was low during the 24 hours before each study session (8/269 reported occurrences, 3%; see Table D.1) and during the study sessions (3/264 reported occurrences, 1%; see Table D.2). Use of alcohol occurred with a mainly even distribution across the six treatments; use was slightly higher in the 24-hour prior to the rocket elbow treatment (three out of 45 [7%] vs one out of 42 to 46 [2%] reported alcohol consumption). Thirty-four of the 47 participants did not change their habits regarding pre-study session alcohol use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Thirty-five of the 47 participants did not change their habits regarding during-study session alcohol use across the six study sessions. Univariate models of the association between alcohol use and treatment in the 24-hours prior to the study day, considering a within person random effect, found a non-significant (p > 0.05) increased odds of alcohol use prior to the rocket elbow stove treatment compared to others (3.5, 95% CI 0.3, 38).

Table D.1 Alcohol Consumption by Treatments: 24 Hours before Session Start.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	45	44	42	43	42	45	261
yes	1	1	1	1	3	1	8
total	46	45	43	44	43	45	269

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	43	44	41	43	44	46	261
yes	2	1	0	0	0	0	3
Not applicable*	1	0	2	1	1	0	5
Total	47	45	43	44	45	47	269

*Not applicable: Participant missed the 24-hour follow-up period/survey.

Caffeine Consumption

Participants were asked not to consume caffeine from 24 hours prior through completion of each study session. Reported caffeine consumption among participants was low during the 24-hours before (24/269 reported occurrences, 9%; see Table D.3) each study session and during the study sessions (13/264 reported occurrences, 5%; see Table D.4). Use of caffeine occurred with a mainly even distribution across the six treatments; use was slightly higher on the second study days when the treatment was LPG compared to others (4 out of 45 [9%] vs one or two out of 43 to 46 [2 to 4%]. Thirty-one of the 47 participants did not change their habits regarding pre-study session caffeine use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Thirty-two of the 47 participants did not change their habits regarding during-study session caffeine use and treatment during the study session. Univariate models of the association between caffeine use and treatment during the study session (between the end of the three-hour measurements through the start of the 24-hour measurements), considering a within person random effect, found a non-significant increased odds of caffeine use for the LPG treatment compared to others (1.2, 95% CI 0.1, 10.5).

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	42	41	39	39	40	44	245
yes	4	4	4	5	5	2	24
Total	46	45	43	44	45	46	269

Table D.3 Caffeine Consumption by Treatments: 24 Hours before Session Start.
Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	43	41	39	42	42	44	251
yes	2	4	2	1	2	2	13
Not applicable*	1	0	2	1	1	0	5
Total	46	45	43	44	45	46	269

Table D.4 Caffeine Consumption by Treatments: During the Study Session.

Medication use

Participants were asked not to use any medications that were not approved by the study physician prior to enrollment, starting 72 hours before the start of each study session and continue throughout the end of the 24-hour health measurements. Reported medication use among participants was low during the 72-hours before each study session (37/269 reported occurrences, 14%; see Table D.5) and during the study sessions (32/264 reported occurrences, 13%; see Table D.6). Use of medication occurred with even distribution across the six treatments. Twenty-nine of the 47 participants did not change their habits regarding pre-study session medication use across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Twenty-eight of the 47 participants did not change their habits regarding their habits regarding during-study session medication use across the six study sessions.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	42	38	37	39	38	38	232
yes	4	7	6	5	7	8	37
Total	46	45	43	44	45	46	269

Table D.5 Medication Use by Treatments: 24 Hours before Session Start.

Use	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	41	41	34	37	40	39	232
yes	4	4	7	6	4	7	32
Not applicable*	1	0	2	1	1	0	5
Total	46	45	43	44	45	46	269

Table D.6 Medication Use by Treatments: During the Study Session.

Smoke Exposures

Participants were asked to avoid smoke exposures from 24 hours prior through completion of each study session. Reported smoke exposures among participants was low during the 24-hours before each study session (2/269 reported occurrences, less than 1%; see Table D.7) and during the study sessions (3/264 reported occurrences, 1%; see Table D.8). Forty of the 47 participants did not change their habits regarding pre-study session smoke exposures across the six study sessions (i.e., consistently reported either "no" or "yes" for all six sessions). Thirty-four of the 47 participants did not change their habits regarding during-study smoke exposures across the six study sessions.

Exposure	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	46	44	43	44	44	46	267
yes	0	1	0	0	1	0	2
Total	46	45	43	44	45	46	269

Table D.7 Smoke Exposures by Treatments: 24 Hours before Session Start.

Exposure	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
no	45	45	40	41	44	46	261
yes	0	0	1	2	0	0	3
Not applicable*	1	0	2	1	1	0	5
Total	47	45	43	44	45	46	269

Table D.8 Smoke Exposures by Treatments: During the Study Session

Mode of Commute

Participants were asked to use the same mode of commute into the facility on each study day. Driving was the most common mode of commute (59% of all trips to the facility for the first study day and 56% of all trips for the second study day involved a car), followed by bike (36% of all trips on the first study day and 31% on the second study day; see Tables S9 and S10). Twenty-six of the 48 participants did not change their habits regarding first day commute mode across the six study sessions (i.e., consistently reported the exact same mode for all six sessions). Twenty-three of the 48 participants did not change their habits regarding the second day commute mode use across the six study sessions.

Mode	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Bike	15	16	17	16	18	14	96
Bike+walk	0	1	1	0	1	0	3
Bus	0	0	0	0	0	0	0
Bus+walk	0	0	1	0	1	0	2
Car	26	25	22	25	24	28	150
Car+walk	3	1	1	1	0	2	8
Walk	2	2	1	1	1	1	8
NA*	0	0	0	1	0	1	2
total	46	45	43	44	45	46	269

Table D.9 Mode of Commute to Facility by Treatments: Before Session Start.

*Not applicable: The participant did not report.

Mode	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Bike	14	15	12	14	14	12	81
Bike+walk	0	0	1	0	1	1	3
Bus	1	1	0	2	0	0	4
Bus+walk	2	2	3	2	3	4	16
Car	25	24	23	23	21	28	145
Car+walk	2	1	1	0	2	0	6
Walk	1	1	0	1	2	1	6
NA*	1	1	3	2	2	0	9
Total	46	45	43	44	45	47	269

Table D.10 Mode of Commute to Facility by Treatments: Prior to the 24-Hour Health Measurements.

*Not applicable: The participant did not report or the participant was not present for the 24-hour measurements.

Sleep Quality

Most participants reported getting an "average" amount of sleep (75% for the night before the study session began and 76% for the night before the second study day); the amount of people reporting below-average sleep was less for the second study day than the first (19% for the night before the study session began vs. 9% for the night before the second study day; see Tables S11 and S12). Only 11 of the 47 participants did not change their habits regarding sleep prior to the start of a study session across the six study sessions (i.e., consistently reported the exact same sleep levels for all six sessions). Thirteen of the 47 participants did not change their habits regarding sleep the night before the 24-hour measurements across the six study sessions.

Sleep	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Above average	3	3	3	0	5	3	17
Average	33	36	32	36	32	32	201
Below average	10	6	8	8	8	11	51
Total	46	45	43	44	45	47	269

Table D.11 Sleep Quality by Treatment: Night Prior to Start of Study Session.

Table D.12 Sleep Quality by Treatment: Night Prior to the 24-Hour Health Measurements.

Sleep	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone	Total
Above average	6	7	6	8	6	6	39
Average	34	35	33	31	32	35	200
Below average	5	3	2	4	6	5	25
Not applicable*	1	0	2	1	1	0	5
Total	46	45	43	44	45	47	269

Ambient PM_{2.5}

Mean ambient PM_{2.5} in the 24-hours prior to the start of a study day ranged from $5.3 \,\mu\text{g/m}^3$ (control) to $9.5 \,\mu\text{g/m}^3$ (fan rocket; see Table D.13). Minimum recorded mean PM_{2.5} was $0.9 \,\mu\text{g/m}^3$ (three stone fire) and maximum recorded mean PM_{2.5} was $18.8 \,\mu\text{g/m}^3$ (LPG). As illustrated in Figure D.1, ambient PM_{2.5} was significantly higher for all treatments compared to the control except the gasifier. However, the range of ambient PM_{2.5} overall was determined to be narrow enough to not include this variable in the main model. We also considered average PM_{2.5} levels for the six hours prior to the start of a study day; results were similar.

PM _{2.5}	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone
mean	5.3	7.6	5.6	9.5	6.7	6.7
min	1.5	2.9	1.0	2.0	2.6	0.9
max	12.3	18.8	11.2	17.6	10.6	12.7

Table D.13 Ambient PM_{2.5} Levels* by Treatment: 24 Hours before Session Start.

* 24-hour average in $\mu g/m^3$



Figure D.1 Effect Estimates and 95% Confidence Intervals for Mean Difference in Ambient PM2.5 Levels for Stove Treatments Compared to Control. Effect is the absolute difference in ambient $PM_{2.5}$ 24-hour average in $\mu g/m^3$.

Ambient CO

Mean ambient CO in the 24-hours prior to the start of a study day ranged from 0.26 ppm (rocket elbow) to 0.35 ppm (three stone fire; see Table D.14). Minimum recorded mean CO was 0.13 ppm (LPG) and maximum recorded mean CO was 0.70 ppm (three stone fire). As illustrated in Figure D.2, ambient CO was significantly higher for the three stone fire and LPG compared to the control. However, the range of ambient CO overall was determined to be narrow enough to

not include this variable in the main model. We also considered average CO levels for the six hours prior to the start of a study day; results were similar.

СО	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone
mean	0.28	0.32	0.27	0.30	0.26	0.35
min	0.16	0.13	0.19	0.17	0.17	0.17
max	0.60	0.48	0.45	0.50	0.44	0.70

Table D.14 Ambient CO Levels* by Treatment: 24 Hours before Session Start.

* 24-hour average in ppm



Figure D.2 Effect Estimates and 95% Confidence Intervals for Mean Difference in Ambient CO Levels for Stove Treatments Compared to Control. Effect is the absolute difference in ambient CO 24-hour average in ppm.

Ambient Temperature

Mean temperature in the 24 hours prior to the start of a study day ranged from 6.4 °C (44 °F; three stone fire) to 15.4 °C (60 °F; fan rocket). Minimum recorded mean temperature was -8.5 °C (17 °F; rocket elbow) and maximum recorded mean temperature was 24.2 °C (76 °F;

rocket elbow; see Table D.15). As illustrated in Figure D.3, temperature was significantly higher for the LPG, fan rocket, and rocket elbow treatments compared to the control. However, the range of temperatures overall was determined to be narrow enough to not include this variable in the main model.

Temp	Control	LPG	Gasifier	Fan rocket	Rocket elbow	Three stone
mean	7.1	10.5	7.9	15.4	13.2	6.4
min	-7.4	2.9	-6.2	4.6	-8.5	-3.1
max	20.0	23.9	16.8	22.3	24.2	15.3

Table D.15 Mean Temperature (°C) by Treatment: 24 Hours before Study Session



Figure D.3 Effect Estimates and 95% Confidence Intervals for Mean Difference in Ambient Temperature for Stove Treatments Compared to Control. Effect is the absolute difference in ambient temperature 24-hour average in ^oC.

Alternative Models and Sensitivity Analyses

Baseline values for each endpoint are shown in Table D.16. Some differences occur across treatments, justifying the inclusion of a baseline term in the model.

Treatment	Mean FVC	Mean FEV ₁	Mean FEV ₁ /FVC	Mean FEF ₂₅₋₇₅
meatiment	(mL)	(mL)	(%)	(mL/s)
Control	4875 (1081)	3864 (787)	79.4 (6.6)	3832 (1101)
LPG	4854 (1024)	3860 (750)	79.8 (6.4)	3836 (987)
Gasifier	4867 (1043)	3873 (803)	79.4 (5.8)	3844 (1151)
Fan rocket	4879 (1148)	3873 (852)	79.4 (6.7)	3823 (1079)
Rocket elbow	4898 (1064)	3895 (816)	79.4 (6.4)	3862 (1148)
Three stone	4860 (1070)	3887 (793)	79.7 (6.8)	3915 (1126)

Table D.16 Mean Baseline (Pre-Exposure) Values for Spirometry Metrics.

Model with more structured study design parameters, in sequence data only (no makeups)

We developed a mixed-effect model that considered more structured study design parameters relevant to our Williams square, such as each individual's assigned sequence group and the day of week (Monday vs. Wednesday), and only included data that was collected within the intended sequence. Results of this model indicate no significance to the various fixed effect "design" terms (day of week, sequence group, or the sequence/day interaction term) (Tables D.17-D.19, Figure D.4). There are no differences in main effect estimates compared to the main model with all data (Tables D.17-D.19; Figure D.4).

Main model, in sequence only (no makeups)

We ran the main model but on a data set that excluded data collected outside of the intended treatment sequence. Results of this model indicate no differences in main effect estimates compared to the main model with all data (Tables D.17-D.19; Figure D.4).

	MAIN MODEL, ALL DATA*	MAIN MODEL, INSEQUENCE DATA ONLY*	DESIGN MODEL, INSEQUENCE DATA ONLY*
	Immediately Post-	Exposure	
(Intercept)	-0.039 (-0.148, 0.069)	-0.053 (-0.165, 0.059)	-0.009 (-0.161, 0.143)
baseline	1.007 (0.987, 1.026)	1.011 (0.991, 1.031)	1.005 (0.979, 1.03)
			0.014 (-0.096,
dayWednesday	NA	NA	0.123)
dayWednesday:sequenceb	NA	NA	0.054)
dayWednesday:sequencec	NA	NA	-0.049 (-0.209, 0.111)
dayWednesday:sequenced	NA	NA	-0.011 (-0.167, 0.144)
dayWednesday:sequencee	NA	NA	0.031 (-0.123, 0.186)
			-0.017 (-0.176,
daywednesday:sequencef	NA	NA	0.143)
sd_(Intercept).date	0.036	0.011	0.024
id	NA	NA	0
sd_(Intercept).id	0	0	NA
sd_Observation.Residual	0.167	0.168	0.168
sequenceb	NA	NA	0.017 (-0.094, 0.129)
sequencec	NA	NA	0.025 (-0.08, 0.129)
sequenced	NA	NA	-0.003 (-0.109, 0.103)
			-0.078 (-0.186,
sequencee	NA	NA	0.03)
sequencef	NA	NA	0.089)
treatment_assignedfan_rock et	NA	NA	-0.067 (-0.146, 0.011)
treatment assignedgasifier	NA	NA	-0.022 (-0.104,
treatment_assignedIng	ΝΔ	ΝΔ	0.015 (-0.062,
treatment_assignedrocket_el		NA	-0.055 (-0.133,
treatment_assignedthree_sto			-0.046 (-0.122,
ne	NA	NA	0.03)
treatmentfan_rocket	-0.06 (-0.135, 0.015)	-0.068 (-0.144, 0.008)	NA
treatmentgasifier	0.019 (-0.056, 0.093)	-0.025 (-0.103, 0.054)	NA
treatmentlpg	0.005 (-0.069, 0.08)	0.011 (-0.063, 0.085)	NA
treatmentrocket_elbow	-0.04 (-0.114, 0.035)	-0.06 (-0.136, 0.016)	NA
treatmentthree_stone	-0.042 (-0.116, 0.032)	-0.05 (-0.124, 0.024)	NA
3 Hours Post-Exposure			
(Intercept)	0.04 (-0.078, 0.158)	0 (-0.124, 0.124)	-0.038 (-0.207, 0.131)

 Table D.17 Comparison of Model Results for Three Model Options, FVC: Effect Estimates

 and 95% Confidence Intervals for all Model Parameters.

dayWednesday NA NA NA 0.04 (-0.084, 0.164) dayWednesday:sequenceb NA NA 0.169) 0.169) dayWednesday:sequenceb NA NA 0.026) 0.152 (-0.331, 0.026) dayWednesday:sequencec NA NA 0.028) 0.014 (-0.027, 0.038 (-0.211, 0.038 (-0.217, 0.011 0 0.0027 0.016 (-0.237, 0.015 NA sd (Intercept).date sd (Intercept).id 0.027 0.015 NA sd (Intercept).id 0.027 0.015 NA scquenceb NA NA 0.064 (-0.185, 0.064 (-0.185, 0.097) sequenceb NA NA 0.013 (-0.103, 0.013 (-0.103, 0.014 (-0.123, 0.011 (-0.013, 0.013 (-0.103, 0.013 (-0.103, 0.014 (-0.103, 0.003 (-0.008, 0.071) NA sequencef NA NA 0.003 (-0.108, 0.071) NA treatment_assignediga NA NA 0.0016 (-0.108, 0.071) 0.0016 (-0.088, 0.07	baseline	0.991 (0.97, 1.013)	0.997 (0.975, 1.019)	1.011 (0.982, 1.039)	
dayWednesday NA NA U144 dayWednesday.sequenceb NA NA 0.009 (-0.187, 0.009 (-0.187, 0.008 (-0.211, 0.008 (-0.211, 0.0134) dayWednesday.sequencee NA NA 0.059 (-0.211, 0.0134) dayWednesday.sequencee NA NA 0.0134 (-0.237, 0.015) dayWednesday.sequencer NA NA 0.016 (-0.237, 0.015) id 0.01 0 0.027 sd_(Intercept).iday:sequence: NA NA 0.016 id 0.027 0.015 NA sd_(Intercept).id 0.027 0.015 NA sequenceb NA NA 0.064 (-0.133, 0.0071) sequencec NA NA 0.062 (-0.181, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.123, 0.003 (-0.012, 0.064) NA treatment_assignedfan_rock et NA NA 0.062 (-0.181, 0.006 (-0.082, 0.067) treatment_assignedfan_rock et NA NA				0.04 (-0.084,	
dayWednesday:sequenceb NA NA 0.159 (dayWednesday	NA	NA	0.164)	
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treatment_assignedrocket_el -0.003 (-0.088, 0.082) bow NA NA 0.082) treatment_assignedthree_sto -0.005 (-0.088, 0.079) -0.005 (-0.088, 0.079) treatmentfan_rocket -0.03 (-0.105, 0.045) -0.019 (-0.102, 0.064) NA treatmentgasifier -0.021 (-0.095, 0.054) -0.023 (-0.108, 0.063) NA treatmentgasifier -0.039 (-0.114, 0.035) -0.02 (-0.101, 0.061) NA treatmentocket_elbow -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmentthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA treatmentthree_stone -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) -0.122 (-0.319, 0.074) (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA NA 0.161)	treatment_assignedlpg	NA	NA	0.064)	
bow NA NA 0.002/ treatment_assignedthree_sto NA NA -0.005 (-0.088, 0.079) treatmentfan_rocket -0.03 (-0.105, 0.045) -0.019 (-0.102, 0.064) NA treatmentgasifier -0.021 (-0.095, 0.054) -0.023 (-0.108, 0.063) NA treatmentpg -0.039 (-0.114, 0.035) -0.02 (-0.101, 0.061) NA treatmentpg -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmentrocket_elbow -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA treatmentthree_stone -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) -0.122 (-0.319, 0.074) (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA NA 0.016 (-0.128, 0.0161)	treatment_assignedrocket_el	ΝΔ	ΝΔ	-0.003 (-0.088,	
ne NA NA 0.079) treatmentfan_rocket -0.03 (-0.105, 0.045) -0.019 (-0.102, 0.064) NA treatmentgasifier -0.021 (-0.095, 0.054) -0.023 (-0.108, 0.063) NA treatmentlpg -0.039 (-0.114, 0.035) -0.022 (-0.101, 0.061) NA treatmentlpg -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmenthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA 24Hours Post-Exposure (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) -0.122 (-0.319, 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA NA 0.016 (-0.128, 0.016 (-0.128, 0.015 (-0.227))	treatment assignedthree sto			-0.005 (-0.088.	
treatmentfan_rocket -0.03 (-0.105, 0.045) -0.019 (-0.102, 0.064) NA treatmentgasifier -0.021 (-0.095, 0.054) -0.023 (-0.108, 0.063) NA treatmentlpg -0.039 (-0.114, 0.035) -0.02 (-0.101, 0.061) NA treatmentrocket_elbow -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmentthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA 24Hours Post-Exposure (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) -0.122 (-0.319, 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.013 (0.981, 1.045) dayWednesday NA NA 0.016 (-0.128, 0.161)	ne	NA	NA	0.079)	
treatmentgasifier -0.021 (-0.095, 0.054) -0.023 (-0.108, 0.063) NA treatmentlpg -0.039 (-0.114, 0.035) -0.02 (-0.101, 0.061) NA treatmentrocket_elbow -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmentthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA 24Hours Post-Exposure (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA NA 0.161)	treatmentfan_rocket	-0.03 (-0.105, 0.045)	-0.019 (-0.102, 0.064)	NA	
treatmentlpg -0.039 (-0.114, 0.035) -0.02 (-0.101, 0.061) NA treatmentrocket_elbow -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmentthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA 24Hours Post-Exosure (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) -0.122 (-0.319, 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.013 (0.981, 1.045) dayWednesday NA NA 0.016 (-0.128, 0.161)	treatmentgasifier	-0.021 (-0.095, 0.054)	-0.023 (-0.108, 0.063)	NA	
treatmentrocket_elbow -0.008 (-0.082, 0.067) -0.006 (-0.089, 0.076) NA treatmentthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA 24Hours Post-Exposure (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA NA 0.161)	treatmentlpg	-0.039 (-0.114, 0.035)	-0.02 (-0.101, 0.061)	NA	
treatmentthree_stone -0.021 (-0.096, 0.053) -0.01 (-0.091, 0.071) NA 24Hours Post-Exposure (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA 0.161 (-0.128, 0.161)	treatmentrocket elbow	-0.008 (-0.082, 0.067)	-0.006 (-0.089, 0.076)	NA	
24Hours Post-Exposure -0.122 (-0.319, 0.074) (Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.045) dayWednesday NA 0.161) -0.015 (-0.227)	treatmentthree stone	-0.021 (-0.096, 0.053)	-0.01 (-0.091, 0.071)	NA	
(Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) -0.122 (-0.319, 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.013 (0.981, 1.045) dayWednesday NA NA 0.161)		24Hours Post-Fy			
(Intercept) -0.057 (-0.181, 0.068) -0.077 (-0.214, 0.06) 0.074) baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.013 (0.981, 1.045) dayWednesday NA 0.016 (-0.128, 0.161) 0.016 (-0.128, 0.161)	-0.122 (-0.319,				
baseline 1.002 (0.979, 1.024) 1.002 (0.978, 1.027) 1.013 (0.981, 1.045) dayWednesday NA 0.016 (-0.128, 0.161) 0.015 (-0.227)	(Intercept)	-0.057 (-0.181, 0.068)	-0.077 (-0.214, 0.06)	0.074)	
dayWednesday NA NA 0.016 (-0.128, 0.161)	baseline	1.002 (0.979, 1.024)	1.002 (0.978, 1.027)	1.013 (0.981, 1.045)	
dayWednesday NA 0.161)				0.016 (-0.128,	
	dayWednesday	NA	NA	0.161)	
dayWednesday:sequenceb NA NA 0.198)	dayWednesday:sequenceb	NA	NA	-0.015 (-0.227, 0.198)	
davWednesdav:sequencec NA NA 0.101)	davWednesdav:sequencec	NA	NA	-0.11 (-0.321, 0.101)	

dayWednesday:sequenced	NA	NA	-0.036 (-0.241, 0.169)
dayWednesday:sequencee	NA	NA	0.023 (-0.181, 0.227)
dayWednesday:sequencef	NA	NA	-0.017 (-0.225, 0.192)
sd_(Intercept).date	0	0.007	0.054
sd_(Intercept).day:sequence: id	NA	NA	0.023
sd_(Intercept).id	0	0	NA
sd_Observation.Residual	0.198	0.204	0.201
sequenceb	NA	NA	-0.023 (-0.177, 0.131)
sequencec	NA	NA	-0.002 (-0.142, 0.138)
sequenced	NA	NA	0.019 (-0.12, 0.159)
sequencee	NA	NA	-0.042 (-0.185, 0.102)
sequencef	NA	NA	0.007 (-0.138, 0.152)
treatment_assignedfan_rock et	NA	NA	0.03 (-0.073, 0.132)
treatment_assignedgasifier	NA	NA	0.041 (-0.066, 0.147)
treatment_assignedlpg	NA	NA	0.033 (-0.067, 0.134)
treatment_assignedrocket_el bow	NA	NA	0.015 (-0.087, 0.116)
treatment_assignedthree_sto ne	NA	NA	0.046 (-0.054, 0.145)
treatmentfan_rocket	0.012 (-0.072, 0.095)	0.027 (-0.067, 0.121)	NA
treatmentgasifier	0.012 (-0.072, 0.096)	0.035 (-0.062, 0.131)	NA
treatmentlpg	0.009 (-0.074, 0.091)	0.029 (-0.062, 0.121)	NA
treatmentrocket_elbow	0.001 (-0.083, 0.084)	0.01 (-0.083, 0.103)	NA
treatmentthree_stone	0.026 (-0.056, 0.108)	0.041 (-0.05, 0.132)	NA

Table D.18 Comparison of Model Results for Three Model Options, FEV₁: Effect Estimates and 95% Confidence Intervals for all Model Parameters.

		MAIN MODEL, INSEQUENCE DATA	DESIGN MODEL, INSEQUENCE DATA
	MAIN MODEL	ONLY	ONLY
	Immediately pos	st-exposure	
(Intercept)	0.05 (-0.067, 0.166)	0.026 (-0.089, 0.14)	0.091 (-0.071, 0.253)
baseline	0.997 (0.97, 1.025)	1.006 (0.979, 1.032)	0.995 (0.959, 1.03)
dayWednesday	NA	NA	-0.03 (-0.144, 0.085)
dayWednesday:sequenceb	NA	NA	-0.025 (-0.189, 0.138)
dayWednesday:sequencec	NA	NA	-0.011 (-0.183, 0.161)
dayWednesday:sequenced	NA	NA	0.06 (-0.102, 0.222)
dayWednesday:sequencee	NA	NA	0.057 (-0.105, 0.219)
dayWednesday:sequencef	NA	NA	0.069 (-0.097, 0.236)

sd_(Intercept).date	0.037	0.021	0.022 F	
sd_(Intercept).day:sequence:	NA	NA	0.046	
sd_(Intercept).id	0.041	0.036	NA	
sd_Observation.Residual	0.145	0.144	0.143	
sequenceb	NA	NA	-0.008 (-0.124, 0.108)	
sequencec	NA	NA	0.032 (-0.078, 0.142)	
sequenced	NA	NA	-0.013 (-0.124, 0.098)	
sequencee	NA	NA	-0.081 (-0.195, 0.032)	
sequencef	NA	NA	-0.07 (-0.183, 0.044)	
treatment_assignedfan_rock et	NA	NA	-0.053 (-0.121, 0.014)	
treatment_assignedgasifier	NA	NA	-0.024 (-0.095, 0.046)	
treatment_assignedlpg	NA	NA	0.009 (-0.057, 0.074)	
treatment_assignedrocket_el bow	NA	NA	-0.044 (-0.111, 0.024)	
treatment_assignedthree_sto ne	NA	NA	-0.037 (-0.102, 0.029)	
treatmentfan_rocket	-0.051 (-0.117, 0.016)	-0.056 (-0.123, 0.011)	NA	
treatmentgasifier	0.007 (-0.059, 0.074)	-0.025 (-0.095, 0.045)	NA	
treatmentlpg	0.003 (-0.064, 0.069)	0.003 (-0.063, 0.068)	NA	
treatmentrocket_elbow	-0.024 (-0.091, 0.042)	-0.049 (-0.116, 0.018)	NA	
treatmentthree_stone	-0.027 (-0.093, 0.039)	-0.041 (-0.106, 0.024)	NA	
3 Hour Post-Exposure				
	3 Hour Post-	Exposure		
(Intercept)	3 Hour Post- 0.178 (0.029, 0.327)	C.135 (-0.021, 0.291)	0.122 (-0.104, 0.348)	
(Intercept) baseline	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005)	0.135 (-0.021, 0.291) 0.976 (0.939, 1.014)	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027)	
(Intercept) baseline dayWednesday	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA	0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA	0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA	0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA NA NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA	••••••••••••••••••••••••••••••••••••	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA	••••••••••••••••••••••••••••••••••••	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencee	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA NA	0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencef sd_(Intercept).date	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA NA O	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA NA NA NA NA NA NA NA NA 0	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencef sd_(Intercept).date sd_(Intercept).day:sequence: id	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA NA NA NA NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequencee dayWednesday:sequencee dayWednesday:sequencef sd_(Intercept).date sd_(Intercept).day:sequence: id sd_(Intercept).id	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA O NA 0 NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA 0 NA 0.083	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencee sd_(Intercept).date sd_(Intercept).date sd_(Intercept).id sd_Observation.Residual	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA NA NA NA 0 NA 0.082 0.145	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA 0 NA 0.083 0.146	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee sd_(Intercept).date sd_(Intercept).id sd_Observation.Residual sequenceb	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA NA NA NA NA 0 NA 0 NA 0.082 0.145 NA	0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA NA NA NA NA NA NA NA NA 0 NA 0.083 0.146	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146 -0.055 (-0.222, 0.112)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencef sd_(Intercept).date sd_(Intercept).date sd_(Intercept).id sd_Observation.Residual sequenceb sequencec	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA 0 NA 0.083 0.146 NA NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146 -0.055 (-0.222, 0.112) 0.067 (-0.091, 0.224)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee sequencee sequencee sequencee	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA NA NA NA NA NA NA NA 0 NA 0.083 0.146 NA NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146 -0.055 (-0.222, 0.112) 0.067 (-0.091, 0.224) 0.054 (-0.104, 0.213)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencee dayWednesday:sequencee sd_(Intercept).date sd_(Intercept).date sd_(Intercept).id sd_Observation.Residual sequenceb sequencec sequencee	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146 -0.055 (-0.222, 0.112) 0.067 (-0.091, 0.224) 0.054 (-0.104, 0.213) -0.038 (-0.199, 0.124)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencee dayWednesday:sequencee dayWednesday:sequencee sequencep).date sd_(Intercept).day:sequence: id sd_(Intercept).id sd_Observation.Residual sequenceb sequencec sequencee sequencee sequencef	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146 -0.055 (-0.222, 0.112) 0.067 (-0.091, 0.224) 0.054 (-0.104, 0.213) -0.038 (-0.199, 0.124) -0.008 (-0.169, 0.153)	
(Intercept) baseline dayWednesday dayWednesday:sequenceb dayWednesday:sequencec dayWednesday:sequenced dayWednesday:sequencee dayWednesday:sequencee dayWednesday:sequencee sd_(Intercept).date sd_(Intercept).day:sequence: id sd_(Intercept).id sd_Observation.Residual sequenceb sequenceb sequencec sequencee sequencee sequencee sequencef treatment_assignedfan_rock et	3 Hour Post- 0.178 (0.029, 0.327) 0.968 (0.932, 1.005) NA	xposure 0.135 (-0.021, 0.291) 0.976 (0.939, 1.014) NA NA NA NA NA NA NA NA 0 NA 0.083 0.146 NA NA NA	0.122 (-0.104, 0.348) 0.976 (0.925, 1.027) 0.096 (-0.068, 0.259) -0.022 (-0.258, 0.213) -0.146 (-0.391, 0.1) -0.103 (-0.332, 0.127) -0.077 (-0.307, 0.153) -0.091 (-0.323, 0.141) 0 0.094 NA 0.146 -0.055 (-0.222, 0.112) 0.067 (-0.091, 0.224) 0.054 (-0.104, 0.213) -0.038 (-0.199, 0.124) -0.055 (-0.122, 0.013)	

treatment_assignedlpg	NA	NA	-0.054 (-0.12, 0.011)
treatment_assignedrocket_el bow	NA	NA	-0.037 (-0.104, 0.03)
treatment_assignedthree_sto ne	NA	NA	-0.033 (-0.098, 0.032)
treatmentfan_rocket	-0.068 (-0.129, -0.007)	-0.054 (-0.122, 0.014)	NA
treatmentgasifier	-0.053 (-0.114, 0.008)	-0.044 (-0.114, 0.026)	NA
treatmentlpg	-0.068 (-0.128, -0.007)	-0.053 (-0.119, 0.012)	NA
treatmentrocket_elbow	-0.039 (-0.099, 0.022)	-0.037 (-0.104, 0.03)	NA
treatmentthree_stone	-0.039 (-0.099, 0.021)	-0.033 (-0.099, 0.032)	NA
	24 Hour Post	Exposure	
(Intercept)	0.035 (-0.114, 0.184)	-0.001 (-0.157, 0.155)	0.03 (-0.216, 0.277)
baseline	0.995 (0.959, 1.031)	1.001 (0.964, 1.039)	0.995 (0.94, 1.051)
dayWednesday	NA	NA	0.041 (-0.131, 0.213)
dayWednesday:sequenceb	NA	NA	-0.041 (-0.298, 0.216)
dayWednesday:sequencec	NA	NA	-0.101 (-0.362, 0.159)
dayWednesday:sequenced	NA	NA	0 (-0.242, 0.242)
dayWednesday:sequencee	NA	NA	-0.002 (-0.245, 0.241)
dayWednesday:sequencef	NA	NA	-0.056 (-0.301, 0.19)
sd_(Intercept).date	0	0	0
sd_(Intercept).day:sequence: id	NA	NA	0.096
sd_(Intercept).id	0.071	0.07	NA
sd_Observation.Residual	0.164	0.17	0.167
sequenceb	NA	NA	-0.039 (-0.228, 0.15)
sequencec	NA	NA	0.029 (-0.139, 0.197)
sequenced	NA	NA	-0.006 (-0.174, 0.163)
sequencee	NA	NA	-0.055 (-0.227, 0.116)
sequencef	NA	NA	-0.006 (-0.176, 0.165)
treatment_assignedfan_rock et	NA	NA	-0.001 (-0.079, 0.078)
treatment_assignedgasifier	NA	NA	0.016 (-0.065, 0.098)
treatment_assignedlpg	NA	NA	0.019 (-0.057, 0.095)
treatment_assignedrocket_el bow	NA	NA	0.006 (-0.071, 0.083)
treatment_assignedthree_sto ne	NA	NA	-0.003 (-0.079, 0.072)
treatmentfan_rocket	-0.015 (-0.084, 0.055)	-0.003 (-0.081, 0.076)	NA
treatmentgasifier	-0.004 (-0.074, 0.066)	0.012 (-0.069, 0.093)	NA
treatmentlpg	0.007 (-0.062, 0.076)	0.016 (-0.061, 0.092)	NA
treatmentrocket_elbow	0 (-0.069, 0.069)	0.001 (-0.077, 0.079)	NA
treatmentthree_stone	-0.014 (-0.082, 0.054)	-0.008 (-0.084, 0.068)	NA

 Table D.19 Comparison of Model Results for Three Model Options, FEF₂₅₋₇₅: Effect

 Estimates and 95% Confidence Intervals for all Model Parameters.

term		MAIN MODEL, INSEQUENCE DATA ONLY	DESIGN MODEL, INSEQUENCE DATA ONLY	
	Immediate Pos	t-Exposure	0	
(Intercept)	0.531 (0.317, 0.744)	0.478 (0.263, 0.694)	0.627 (0.305, 0.948)	
baseline	0.903 (0.853, 0.952)	0.915 (0.865, 0.965)	0.878 (0.814, 0.941)	
dayWednesday	NA	NA	-0.16 (-0.468, 0.148)	
dayWednesday:sequenceb	NA	NA	0.11 (-0.32, 0.541)	
dayWednesday:sequencec	NA	NA	0.119 (-0.346, 0.583)	
dayWednesday:sequenced	NA	NA	0.016 (-0.419, 0.451)	
dayWednesday:sequencee	NA	NA	0.089 (-0.344, 0.522)	
dayWednesday:sequencef	NA	NA	0.376 (-0.067, 0.819)	
sd_(Intercept).date	0.056	0.051	0.047	
sd_(Intercept).day:sequence:	NA	NA	0.176	
sd_(Intercept).id	0.156	0.147	NA	
sd_Observation.Residual	0.278	0.277	0.272	
sequenceb	NA	NA	-0.081 (-0.385, 0.223)	
sequencec	NA	NA	0.141 (-0.159, 0.441)	
sequenced	NA	NA	0.118 (-0.185, 0.422)	
sequencee	NA	NA	0.034 (-0.28, 0.348)	
sequencef	NA	NA	-0.127 (-0.428, 0.175)	
treatment_assignedfan_rock et	NA	NA	-0.098 (-0.228, 0.032)	
treatment_assignedgasifier	NA	NA	-0.022 (-0.158, 0.115)	
treatment_assignedlpg	NA	NA	-0.025 (-0.153, 0.102)	
treatment_assignedrocket_el bow	NA	NA	-0.071 (-0.201, 0.059)	
treatment_assignedthree_sto ne	NA	NA	-0.098 (-0.225, 0.029)	
treatmentfan_rocket	-0.116 (-0.239, 0.008)	-0.107 (-0.239, 0.026)	NA	
treatmentgasifier	-0.013 (-0.137, 0.11)	-0.021 (-0.159, 0.117)	NA	
treatmentlpg	-0.044 (-0.167, 0.079)	-0.033 (-0.163, 0.097)	NA	
treatmentrocket_elbow	-0.068 (-0.191, 0.055)	-0.076 (-0.209, 0.056)	NA	
treatmentthree_stone	-0.103 (-0.225, 0.019)	-0.102 (-0.231, 0.027)	NA	
3 Hour Post-Exposure				
(Intercept)	0.572 (0.318, 0.826)	0.531 (0.255, 0.806)	0.959 (0.469, 1.45)	
baseline	0.886 (0.826, 0.946)	0.887 (0.822, 0.951)	0.747 (0.653, 0.842)	
dayWednesday	NA	NA	-0.03 (-0.528, 0.468)	
dayWednesday:sequenceb	NA	NA	0.007 (-0.694, 0.708)	
dayWednesday:sequencec	NA	NA	0.355 (-0.395, 1.104)	
dayWednesday:sequenced	NA	NA	-0.21 (-0.913, 0.493)	
dayWednesday:sequencee	NA	NA	-0.127 (-0.829, 0.575)	

dayWednesday:sequencef	NA	NA	0.275 (-0.433, 0.984)
sd_(Intercept).date	0	0	0
sd_(Intercept).day:sequence: id	NA	NA	0.325
sd_(Intercept).id	0.198	0.21	NA
sd_Observation.Residual	0.319	0.325	0.306
sequenceb	NA	NA	-0.047 (-0.542, 0.448)
sequencec	NA	NA	0.149 (-0.339, 0.637)
sequenced	NA	NA	0.284 (-0.21, 0.777)
sequencee	NA	NA	0.338 (-0.169, 0.845)
sequencef	NA	NA	-0.081 (-0.57, 0.408)
treatment_assignedfan_rock et	NA	NA	-0.055 (-0.198, 0.088)
treatment_assignedgasifier	NA	NA	-0.051 (-0.2, 0.097)
treatment_assignedlpg	NA	NA	-0.085 (-0.223, 0.054)
treatment_assignedrocket_el bow	NA	NA	-0.029 (-0.17, 0.111)
treatment_assignedthree_sto ne	NA	NA	0.008 (-0.129, 0.146)
treatmentfan_rocket	-0.114 (-0.249, 0.021)	-0.063 (-0.213, 0.088)	NA
treatmentgasifier	-0.074 (-0.208, 0.059)	-0.045 (-0.201, 0.111)	NA
treatmentlpg	-0.122 (-0.255, 0.011)	-0.082 (-0.228, 0.064)	NA
treatmentrocket_elbow	-0.056 (-0.19, 0.077)	-0.034 (-0.183, 0.115)	NA
treatmentthree_stone	-0.031 (-0.164, 0.102)	0.005 (-0.141, 0.151)	NA
	24 Hour Post-	Exposure	
(Intercept)	0.696 (0.42, 0.972)	0.622 (0.332, 0.911)	1.996 (1.269, 2.724)
baseline	0.847 (0.782, 0.913)	0.859 (0.79, 0.927)	0.48 (0.37, 0.591)
dayWednesday	NA	NA	-0.237 (-1.1, 0.626)
dayWednesday:sequenceb	NA	NA	-0.136 (-1.405, 1.132)
dayWednesday:sequencec	NA	NA	0.968 (-0.318, 2.255)
dayWednesday:sequenced	NA	NA	-0.2 (-1.421, 1.021)
dayWednesday:sequencee	NA	NA	-0.239 (-1.46, 0.983)
dayWednesday:sequencef	NA	NA	0.646 (-0.579, 1.871)
sd_(Intercept).date	0	0	0
sd_(Intercept).day:sequence: id	NA	NA	0.611
sd_(Intercept).id	0.246	0.249	NA
sd_Observation.Residual	0.278	0.282	0.239
sequenceb	NA	NA	0.123 (-0.808, 1.054)
sequencec	NA	NA	-0.025 (-0.885, 0.835)
sequenced	NA	NA	0.311 (-0.553, 1.175)
sequencee	NA	NA	0.561 (-0.313, 1.435)
sequencef	NA	NA	-0.116 (-0.976, 0.744)
treatment_assignedfan_rock et	NA	NA	-0.019 (-0.132, 0.093)

treatment_assignedgasifier	NA	NA	-0.008 (-0.124, 0.109)
treatment_assignedlpg	NA	NA	0.049 (-0.06, 0.158)
treatment_assignedrocket_el bow	NA	NA	0.073 (-0.037, 0.184)
treatment_assignedthree_sto ne	NA	NA	-0.047 (-0.155, 0.061)
treatmentfan_rocket	-0.081 (-0.199, 0.037)	-0.035 (-0.167, 0.096)	NA
treatmentgasifier	-0.063 (-0.182, 0.056)	-0.012 (-0.148, 0.124)	NA
treatmentlpg	0.039 (-0.078, 0.156)	0.053 (-0.075, 0.18)	NA
treatmentrocket_elbow	0.035 (-0.083, 0.153)	0.062 (-0.068, 0.192)	NA
treatmentthree_stone	-0.088 (-0.204, 0.027)	-0.062 (-0.189, 0.065)	NA



◆ Immediate Post ◆ Three Hour Post ◆ 24 Hour Post



Figure D.4 Effect Estimates and 95% Confidence Intervals for Mean Difference in Endpoint for Stove Treatments Compared to Control Across the Three Model Types. Top: FVC, Middle Panel: FEV₁, Bottom Panel: FEF₂₅₋₇₅.

Sensitivity analyses: Main model, remove C/D quality tests

We conducted sensitivity analyses wherein data was removed for measurements that did not meet an A or B quality rating. This resulted in removing 21 data points (8% of the data) from the immediate-post exposure models, 25 data points (10% of the data) from the three-hour post-exposure models, and 17 data points (7% of the data) from the 24-hour post-exposure models. However, model results indicated no considerable differences between the estimates for the treatment effects (see Figure D.5).

Sensitivity analyses: Main model, remove when exposure value outside narrow range of target

We ran the main model excluded data from study sessions where the exposure mean was outside of a narrowed range around the target value. The narrowed ranges were:

- Control: less than 5 μg/m³
- LPG: 5-15 μg/m³
- Gasifier: 20-60 µg/m³
- Fan rocket: 75-125 μg/m³
- Rocket elbow: 175-300 μg/m³
- Three stone fire: 350-600 µg/m³

Results indicated no considerable differences between the estimates for the treatment effects between this model and the main model (see Figure D.5).





Figure D.5 Effect Estimates and 95% Confidence Intervals for Mean Difference in Systolic Pressure (*mmHg*) for Stove Treatments Compared to Control: Comparison of Main Model to Models with C/D Quality Tests Removed and Exposure Outliers Removed. Top: FVC, Middle Panel: FEV₁, Bottom Panel: FEF₂₅₋₇₅.