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Chemical composition and source apportionment of ambient, household, and personal exposures to $PM_{2.5}$ in communities using biomass stoves in rural China

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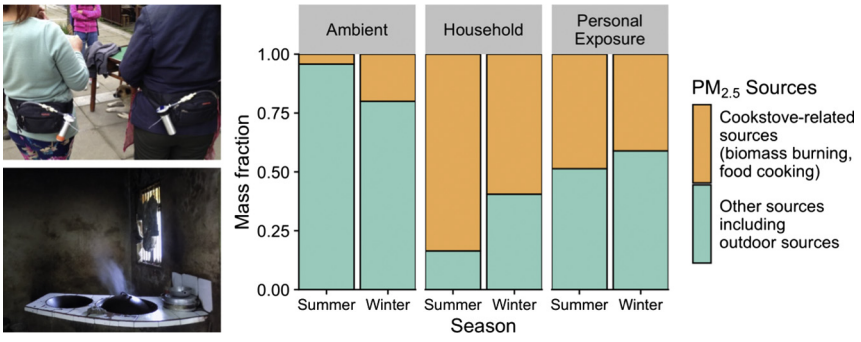
HIGHLIGHTS

- Biomass burning was the largest identified source of personal exposure $PM_{2.5}$.
- Household sources (cooking, biomass burning) contributed up to 20% of ambient $PM_{2.5}$.
- Up to 15% of indoor $PM_{2.5}$ was from outdoor sources (dust, vehicles, secondary PM).
- Personal activities influenced dust exposures beyond indoor/outdoor concentrations.

GRAPHICAL ABSTRACT



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ABSTRACT

Fine particulate matter ($PM_{2.5}$) has health effects that may depend on its sources and chemical composition. Few studies have quantified the composition of personal and area $PM_{2.5}$ in rural settings over the same time period. Yet, this information would shed important light on the sources influencing personal $PM_{2.5}$ exposures. This study investigated the sources and chemical composition of 40 personal exposure, 40 household, and 36 ambient $PM_{2.5}$ samples collected in the non-heating and heating seasons in rural southwestern China. Chemical analysis included black carbon (BC), water-soluble components (ions, organic carbon), elements, and organic tracers. Source apportionment was conducted using organic tracer concentrations in a Chemical Mass Balance model. Biomass burning was the largest identified $PM_{2.5}$ source contributor to household (average, SD: 48 ± 11%) and exposures (31 ± 6%) in both seasons, and ambient $PM_{2.5}$ in winter (20 ± 4%). Food cooking also contributed to household and personal PM, reaching approximately half of the biomass contributions. Secondary inorganic aerosol was the major identified source in summertime ambient $PM_{2.5}$ (32 ± 14%), but was present in all samples

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Biomass burning
Chemical mass balance
Solid fuels

(summer: 10 ± 3% [household], 13 ± 6% [exposures]; winter: 18 ± 2% [ambient], 7 ± 2% [household], 8 ± 2% [exposures]). Dust concentrations and fractional contribution to total $PM_{2.5}$ were higher in summer exposure samples (7 ± 4%) than in ambient or household samples (6 ± 1% and 2 ± 1%, respectively). Indoor sources comprised up to one-fifth of ambient $PM_{2.5}$, and outdoor sources (vehicles, secondary aerosols) contributed up to 15% of household $PM_{2.5}$. While household sources were the main contributors to $PM_{2.5}$ exposures in terms of mass, inorganic components of personal exposures differed from household samples. Based on these findings, health-focused initiatives to reduce harmful $PM_{2.5}$ exposures may consider a coordinated approach to address both indoor and outdoor $PM_{2.5}$ source contributors.

1. Introduction

Solid fuels such as biomass and coal are the primary household energy source for billions of people worldwide (Bonjour et al., 2013). Inefficient combustion of solid fuels releases air pollutants including fine particulate matter ($PM_{2.5}$), which contribute to both indoor and outdoor air pollution and are a leading environmental health risk factor (GBD 2016 Risk Factors Collaborators, 2017; Prüss-Üstün et al., 2016). To mitigate the health impacts of household air pollution, numerous governments and organizations have implemented programs to distribute cleaner-burning stoves and fuels (e.g. Anenberg et al., 2013; Dickinson et al., 2015; GACC, 2015; Shen, 2016; Thomas et al., 2015). While household pollution levels are doubtless major contributors to exposures in settings of household solid fuel combustion, personal PM exposures are also influenced by other factors including other pollutant sources, time-varying activity patterns, and variability between individuals. This complexity makes it challenging to assess how much changing a specific source actually affects personal PM exposures. Given that precise, quantitative exposure assessments are crucial for establishing exposure-response relationships that are transferable across diverse settings of household solid fuel use (Peel et al., 2015), mitigation strategies and epidemiologic research on household air pollution should include assessment of other sources and activities impacting personal exposures.

Detailed investigations of $PM_{2.5}$ chemical composition and sources have been conducted primarily in urban areas, even in regions where rural populations are using solid fuels for household energy (Pant et al., 2016; Zheng et al., 2016). Only a few personal exposure studies have measured chemical tracers of sources other than biomass burning (Baumgartner et al., 2014) and used that data to identify those sources (Huang et al., 2015; Secrest et al., 2016), and their results suggest that other sources including dust, vehicles, and food cooking also contribute substantially to personal exposures.

Understanding how closely personal exposures resemble individuals' microenvironments may also help to discern how much specific source mitigation impacts personal exposures. However, studies directly comparing the chemical composition of personal PM exposures, indoor PM, and outdoor PM are currently limited to urban settings (Brokamp et al., 2015; Gadkari and Pervez, 2007; Larson et al., 2004), with the exception of Downward et al. (2016), who measured indoor, outdoor, and personal BC exposures in settings of household solid fuel combustion in rural Yunnan, China. While Downward et al. found that personal BC exposures and household BC were well correlated, the three urban studies each identified at least one source contributing to personal exposures that was not well correlated with either outdoor or indoor concentrations. Given that field studies of solid fuel combustion analyze the chemical composition of only household PM more often than that of personal PM exposures or both (Secrest et al., 2017), direct comparison of the composition and sources of personal PM exposures to household and ambient PM is a crucial yet insufficiently studied research area.

In this study, we analyzed the chemical composition and sources of household, ambient, and personal $PM_{2.5}$ exposures in rural Sichuan province, China. To the authors' knowledge, this is the first published study of source apportionment of outdoor, indoor, and personal exposure $PM_{2.5}$ in a rural setting with widespread household solid fuel combustion. We address three main research questions: (1) the major sources contributing to household, ambient, and personal $PM_{2.5}$ exposures, (2) the influence of household air pollution on ambient air quality, compared to regional sources, and (3) the extent to which household and ambient sources each contribute to personal exposures, in addition to personal activity contributions.

2. Methods

2.1. Study setting

The $PM_{2.5}$ samples analyzed for this study were collected from May 2014 to January 2017 as part of a 3-year clean cookstove field study (Section S1). Details of study design, stove and energy use, and demographic data are described elsewhere (Carter et al., 2016; Ni et al., 2016). Briefly, the study site is comprised of 12 natural villages in southwestern Sichuan province. These villages were selected based on their inclusion in a planned clean energy initiative led by China's Ministry of Agriculture and Ministry of Science and Technology. Stove emissions performance in the lab and field and household adoption and uptake of the household energy package are reported elsewhere (Clark et al., 2017; Shan et al., 2017). The study villages are situated in a rural, mountainous region with few other local sources of air pollution. The nearest major road has low to moderate traffic density and most villages are at a higher elevation and physically isolated from the road. There is also a cement factory in the area. However, continuous (1-minute resolution) ambient monitoring of $PM_{2.5}$ indicated that daily and weekly variation in $PM_{2.5}$ did not correspond with the factory's operating schedule. Therefore, the factory was not anticipated to be a substantial contributor to filter-based $PM_{2.5}$ sample collection in the village homes and on study participants. Study protocols were approved by institutional review boards at McGill University (Canada), the University of Wisconsin-Madison (USA), and Tsinghua University (China).

2.2. Air pollution sampling methods

Personal $PM_{2.5}$ samples were collected at 48-h intervals on 37 mm PTFE filters (2 μm pore size; Zefluor; Pall Laboratory, USA) using Personal Exposure Monitors (PEMs) with greased impaction surfaces (Demokritou et al., 2001). Participants wore a small waistpack containing the sampling pump and PEM (<1 kg). Air was drawn through the PEM at a flow rate of 1.8 L per minute (LPM), using active sampling pumps (Apex Pro, Casella, UK) that were calibrated with a rotameter before each deployment.

Household $PM_{2.5}$ samples were collected for 48-h concurrently with personal exposure measurements, using either PEMs or a cyclone with an aluminum filter cassette (GK2.05 SH and GK2.05 cyclone models, Mesa Labs, USA). The cyclones operated at either 3.5 (for the GK2.05 SH cyclone) or 4 LPM (for the GK2.05 cyclone). Household sampling took place in kitchen areas within 1 m of the traditional stove and