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Assessing Exposure to Household Air Pollution: A Systematic Review and Pooled Analysis of Carbon Monoxide as a Surrogate Measure of Particulate Matter

Ellison Carter,[1] Christina Norris,[2] Kathie L. Dionisio,[3] Kalpana Balakrishnan,[4] William Checkley,[5,6] Maggie L. Clark,[7] Santu Ghosh,[4] Darby W. Jack,[8] Patrick L. Kinney,[8] Julian D. Marshall,[9] Luke P. Naeher,[10] Jennifer L. Peel,[7] Sankar Sambandam,[4] James J. Schauer,[11,12] Kirk R. Smith,[13] Blair J. Wylie,[14] and Jill Baumgartner[1,2,15]*

[1]Institute on the Environment, University of Minnesota, St. Paul, Minnesota, USA
[2]Department of Epidemiology, Biostatistics & Occupational Health, McGill University, Montreal, Quebec, Canada
[3]National Exposure Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina, USA
[4]Department Environmental Health Engineering, Sri Ramachandra University, Porur, Chennai, India
[5]Division of Pulmonary and Critical Care, School of Medicine, Johns Hopkins University, Baltimore, Maryland, USA
[6]Program in Global Disease Epidemiology and Control, Department of International Heath, Johns Hopkins Bloomberg School of Public Health, Baltimore, Maryland, USA
[7]Department of Environmental and Radiological Health Sciences, Colorado State University, Fort Collins, Colorado, USA
[8]Department of Environmental Health Sciences, Mailman School of Public Health, Columbia University, New York, New York, USA
[9]Department of Civil and Environmental Engineering, University of Washington, Seattle, Washington, USA
[10]Department of Environmental Health Science, College of Public Health, The University of Georgia, Athens, Georgia, USA
[11]Environmental Chemistry & Technology Program, University of Wisconsin-Madison, Madison, Wisconsin, USA
[12]Department of Civil & Environmental Engineering, University of Wisconsin-Madison, Madison, Wisconsin, USA
[13]Division of Environmental Health Sciences, School of Public Health, University of California, Berkeley, Berkeley, California, USA
[14]Division of Maternal-Fetal Medicine, Department of Obstetrics and Gynecology, Massachusetts General Hospital and Harvard Medical School, Boston, Massachusetts, USA
[15]Institute for Health and Social Policy, McGill University, Montreal Quebec, Canada

Background: Household air pollution from solid fuel burning is a leading contributor to disease burden globally. Fine particulate matter ($PM_{2.5}$) is thought to be responsible for many of these health impacts. A co-pollutant, carbon monoxide (CO) has been widely used as a surrogate measure of $PM_{2.5}$ in studies of household air pollution.

Objective: The goal was to evaluate the validity of exposure to CO as a surrogate of exposure to $PM_{2.5}$ in studies of household air pollution and the consistency of the $PM_{2.5}$–CO relationship across different study settings and conditions.

Methods: We conducted a systematic review of studies with exposure and/or cooking area $PM_{2.5}$ and CO measurements and assembled 2,048 $PM_{2.5}$ and CO measurements from a subset of studies (18 cooking area studies and 9 personal exposure studies) retained in the systematic review. We conducted pooled multivariate analyses of $PM_{2.5}$–CO associations, evaluating fuels, urbanicity, season, study, and CO methods as covariates and effect modifiers.

Results: We retained 61 of 70 studies for review, representing 27 countries. Reported $PM_{2.5}$–CO correlations (r) were lower for personal exposure (range: 0.22–0.97; median = 0.57) than for cooking areas (range: 0.10–0.96; median = 0.71). In the pooled analyses of personal exposure and cooking area concentrations, the variation in ln(CO) explained 13% and 48% of the variation in ln($PM_{2.5}$), respectively.

Conclusions: Our results suggest that exposure to CO is not a consistently valid surrogate measure of exposure to $PM_{2.5}$. Studies measuring CO exposure as a surrogate measure of PM exposure should conduct local validation studies for different stove/fuel types and seasons. https://doi.org/10.1289/EHP767

Introduction

Over 2.8 billion people are exposed to household air pollution from cooking and heating with solid fuels, which include biomass (e.g., wood, crop residues, animal dung, charcoal) and coal (Bonjour et al. 2013). Household air pollution comprises many pollutants (Zhang and Smith 2007; Naeher et al. 2007) and is a leading health risk factor, annually responsible for an estimated 2.9 million premature deaths (GBD 2013 Risk Factors Collaborators et al. 2015). Two widely studied air pollutants from solid fuel combustion are particulate matter (PM) and carbon monoxide (CO). Strong epidemiologic and experimental evidence point to the mass of PM with a diameter ≤2.5 μm ($PM_{2.5}$) as a pollutant that is causally associated with many health outcomes (Pope and Dockery 2006; U.S. EPA 2009) and is likely a strong driver of many health effects associated with household air pollution (Brook et al. 2010; WHO 2014). Evidence for adverse health outcomes related to low-to-moderate CO exposure is sparse and less consistent, with associations between infant low birth weight and women's CO exposure during pregnancy demonstrated in some studies (Ritz and Yu 1999; Ha et al. 2001; Gouveia et al. 2004; Salam et al. 2005), but not in others (Alderman et al. 1987; Koren et al. 1991; Chen et al. 2002; Parker et al. 2005; Wylie et al 2016). In epidemiologic and exposure studies of household air pollution, including those evaluating maternal exposure and birth outcomes (Thompson et al. 2011; Dix-Cooper et al. 2012), CO exposure is usually measured as a surrogate of $PM_{2.5}$ exposure (Balakrishnan et al. 2011; Clark et al. 2013).

Accurate exposure assessment is the basis for evaluating exposure–response relationships (Armstrong 1998, 2004), and in the context of household air pollution, critical to interpreting the effectiveness of stove-fuel interventions (Peel et al. 2015). Direct

Address correspondence to J. Baumgartner, Institute for Health and Social Policy and Department of Epidemiology, Biostatistics and Occupational Health, 1130 des Pins Ave. Ouest, Montréal, Québec H3A 1A3 Canada. Telephone: (514) 398-6688. Email: jill.baumgartner@mcgill.ca

*Current affiliation: Department of Civil and Environmental Engineering, Colorado State University, 400 Isotope Dr, Fort Collins, CO 80521 USA.

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measurement of personal exposure to $PM_{2.5}$ mass is considered the "gold standard" in epidemiologic studies (Smith 1993; Northcross et al. 2015), but is challenging to measure in large populations (Northcross et al. 2015) and in infants (Naeher et al. 2001; Dionisio et al. 2008). Questionnaires and cooking area $PM_{2.5}$ have been used alone or in combination as surrogates but were poorly associated with personal $PM_{2.5}$ exposure in validation studies (Ezzati et al. 2000; Bruce et al. 2004; Cynthia et al. 2008; Baumgartner et al. 2011; Ni et al. 2016). As an alternative, many health and intervention studies have measured personal exposure to CO as a surrogate for $PM_{2.5}$ given that it is also a major component of household air pollution but is easier and less costly to measure than $PM_{2.5}$ (Naeher et al. 2001; Dionisio et al. 2008; Smith et al. 2010).

The empirical evidence supporting the validity of personal CO exposure as a surrogate of personal $PM_{2.5}$ exposure is limited and inconclusive, despite its common use. Direct measurements of personal $PM_{2.5}$ and CO exposure were not correlated (Pearson $r = -0.04$) in children living in homes cooking with wood in The Gambia (Dionisio et al. 2012) and were only moderately correlated in women in Peru (Spearman $r = 0.41$; Commodore et al. 2013), Tanzania (Spearman $r = 0.34$; Wylie et al. 2016), and China (Spearman $r = 0.60$; Ni et al. 2016). In rural Guatemala, however, variation in personal CO exposures explained 78% of the variation in personal $PM_{2.5}$ exposures among women (McCracken et al. 2013). It is not known whether the strength of a $PM_{2.5}$–CO relationship in one setting is transportable to other settings. Here, we use definitions for a validation study and transportability adapted from Spiegelman (2010):

Validation study: a study in which data are simultaneously collected on the exposure surrogate (CO) and the gold standard method of exposure assessment ($PM_{2.5}$). This study may be external to the main epidemiologic study, or be a subsample internal to the main study.

Transportability: the extent to which the $PM_{2.5}$–CO relationship in a validation study is similar to the one that generates the surrogate exposure in the main study ($PM_{2.5}$).

Studies in Bolivia, Peru, Ghana, Kenya, Mexico, South Africa, the Philippines, and Burkina Faso (Röllin et al. 2004; Saksena et al. 2007; Riojas-Rodriguez et al. 2011; Ochieng et al. 2013; Commodore et al. 2013; Alexander et al. 2014; Thorsson et al. 2014; Jack et al. 2015; Yip et al. 2017) measured CO exposure as a surrogate for $PM_{2.5}$ without prior validation; one reason given was the strong $PM_{2.5}$–CO exposure relationship observed in Guatemala (McCracken et al. 2013; Naeher et al. 2000a). Similarly, it is unknown whether the $PM_{2.5}$–CO exposure relationship within a single study setting and population under one set of study conditions is transportable to other study conditions (e.g., pre- vs. postintervention; heating- vs. nonheating season) in the same setting and population, which is an approach taken in some studies (Smith-Sivertsen et al. 2009; Smith et al. 2011; Guarnieri et al. 2014; Pope et al. 2015). Finally, it is unclear whether the $PM_{2.5}$–CO correlation in cooking areas can be extrapolated to personal exposures in the same setting, which several studies have done (Bruce et al. 2004; Northcross et al. 2010; Dionisio et al. 2012; Alnes et al. 2014). Of these, only one (Dionisio et al. 2012) directly compared actual versus predicted $PM_{2.5}$ exposure, finding no relationship (Pearson $r = 0.01$).

Both PM and CO are products of incomplete combustion and co-emitted during solid fuel burning. The amount and relative proportion of these pollutants emitted from stoves can vary by factors including fuel type and moisture content; combustion efficiency and power throughout burn cycles; stove ventilation; and the behavior of energy users (Roden et al. 2009; Chen et al. 2012; Jetter et al. 2012; Carter et al. 2014). For personal exposures, the presence of other community or regional air pollution sources with different pollutant composition (Huang et al. 2015) could further impact the strength and consistency of a personal PM-CO association.

We systematically reviewed the methods and correlation coefficients reported in studies with paired measurements of $PM_{2.5}$ and CO personal exposures and/or cooking area concentrations in settings where biomass is the primary household fuel. We also obtained 2,048 paired $PM_{2.5}$ and CO measurements from previously completed studies along with relevant information on season, level of urbanicity, fuel type, and other energy use behaviors to conduct pooled analyses of the $PM_{2.5}$–CO relationship for personal exposures and cooking area concentrations. For the pooled analysis, our first objective was to evaluate the validity of exposure to CO as a surrogate of exposure to $PM_{2.5}$ in epidemiologic and intervention studies of household air pollution. Because most health studies aim to evaluate daily or "usual" exposure, we limited our pooled analysis to studies of $PM_{2.5}$ and CO concentration and/or exposure relationships for at least 24-hr in settings where biomass was the dominant household fuel. Our second objective was to evaluate whether the $PM_{2.5}$–CO relationships estimated under one set of conditions are transportable to other conditions.

We provide a timely assessment of CO exposure as a surrogate of $PM_{2.5}$ exposure, as a systematic review has been lacking but is critical to exposure measurement method selection for ongoing (Rosa et al. 2014; Klasen et al. 2013; Tielsch et al. 2014; Jack et al. 2015; NIH 2015) randomized controlled trials and other epidemiologic studies.

Methods

Search Strategy and Selection Criteria for the Systematic Review

We searched publications included in the electronic database PubMed (from 1966 to present; https://www.ncbi.nlm.nih.gov/pubmed/) and the Science Citation index, as well as the electronic databases Ovid MEDLINE® In-Process & Other NonIndexed Citations, Ovid MEDLINE® Daily, Ovid OLDMEDLINE®, (1946 to present, http://ospguides.ovid.com/OSPguides/medline.htm) and Embase Classic + Embase (1947 to present, http://www.elsevier.com/embase). We searched for combinations of the key words "carbon monoxide" or "CO" and "indoor air pollution" or "indoor*" or "house*" or "home*" or "personal exposure and particulate*" or "PM*" and "biomass or coal" or "fuel*" or "wood*" or "dung" or "crop" or "agricultural residue*." The search was restricted to articles available in English, French, Spanish, or Chinese. We retained articles for which cooking area or personal measurements of PM and CO were done concurrently in a setting where biomass was burned for cooking and/or heating. Two researchers independently extracted information from these articles and hand-searched their reference lists to identify additional publications for retrieval. Finally, we contacted 15 researchers to directly obtain data from published and unpublished studies with paired PM and CO measurements. These studies were identified from the literature review and recent conference proceedings or academic meetings. We adhered to systematic review guidelines from PRISMA-P guidelines and the Cochrane Collaboration (Van Tulder et al. 2003; Moher et al. 2015).

We classified studies retained for this review into two groups (Figure 1): studies with paired measurements of personal PM and

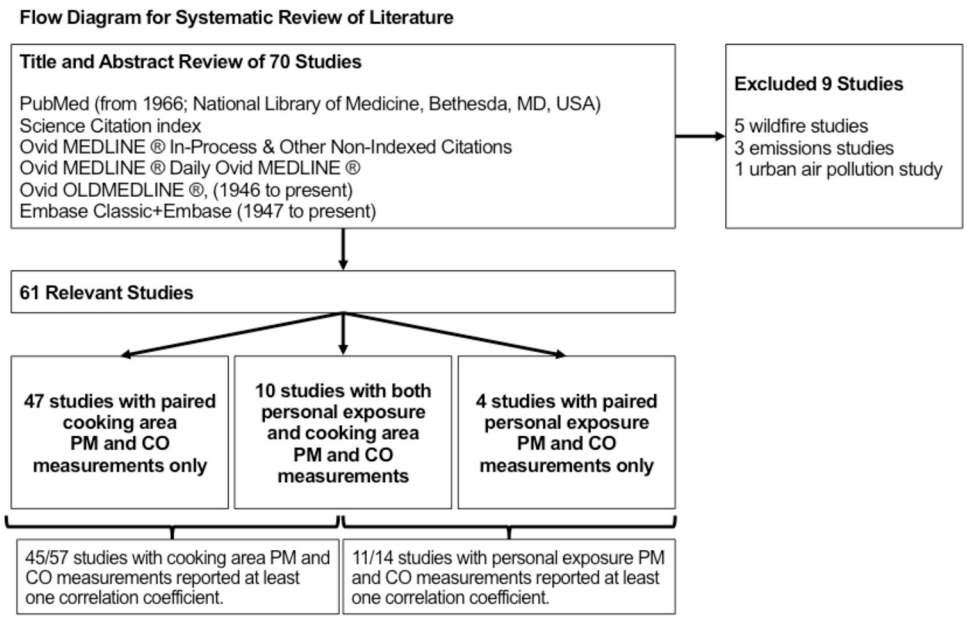

Figure 1. Flow diagram of systematic search of literature for review.

CO exposures or stationary PM and CO concentrations in cooking areas. Though personal exposures were our primary interest, we reviewed studies with paired cooking area measurements because they are more common than studies with personal exposure (Balakrishnan et al. 2011; Clark et al. 2013) and may shed additional light on the $PM_{2.5}$–CO relationship for personal exposures. For every study, the following information was extracted: authors, year of publication, year(s) of data collection, location, season(s), description of setting, elevation, description of study population (see Table S1), stove types, cooking location, cooking area ventilation, fuel types, other local air pollution sources, number of paired PM and CO measurements, pollutant measurement methods (i.e., protocols, instrumentation, quality assurance, quality control measures), and reported $PM_{2.5}$–CO correlation coefficients. When information was not reported, we requested it from corresponding authors.

Compiling Paired $PM_{2.5}$ and CO Measurements for Pooled Data Analysis

We contacted the corresponding authors of studies identified in our systematic review to obtain the paired $PM_{2.5}$ and CO data. If authors did not respond but the data were available in published studies, we downloaded those data. We also requested information on the stoves and fuels used, stove ventilation, other local air pollution sources, season of data collection, level of urbanicity (rural vs. peri-urban/urban), and $PM_{2.5}$ and CO measurement methods. If these variables were unavailable for individual observations, we assigned them at the study level based on the information reported in the manuscript or communication with authors.

We obtained 2,048 paired $PM_{2.5}$ and CO measurements and covariate data from 9 studies of personal $PM_{2.5}$ and CO exposures ($n = 714$ pairs) and 18 studies of cooking area $PM_{2.5}$ and CO concentrations ($n = 1{,}334$ pairs). Personal exposure data were obtained from authors for 6 studies and extracted from tables or figures from 3 studies (Fitzgerald et al. 2012; McCracken et al. 2013; Naeher et al. 2000b). For paired cooking area measurements, data were obtained from authors of 9 studies, and we extracted data from 9 studies (Naeher et al. 2000a; Naeher et al. 2001; Park and Lee 2003; Chengappa et al. 2007; Dutta et al. 2007; Henkle et al. 2010; Fitzgerald et al. 2012; Chowdhury et al. 2013; Huboyo et al. 2014). We created dichotomous variables for covariates (see Table S2), including fuel use (exclusive vs. nonexclusive biomass use), whether other local sources of air pollution were reported, level of urbanicity, season (nonheating vs. heating), and CO measurement method (colorimetric-based vs. sensor-based). PM measurement type was almost exclusively gravimetric for personal exposures and a mix of gravimetric and optical measurements in cooking areas. We summarized the protocols and quality assurance/control procedures for personal PM and CO in Tables S3 and S4.

Statistical Analysis of the Pooled Data

We conducted a series of univariate and multivariate regression models to evaluate the coefficient of determination (R^2) and the slope between $PM_{2.5}$ and CO, with separate models for personal exposure and cooking area measurements. Natural cubic spline functions with 2–5 degrees of freedom were used to evaluate whether the pollutant relationships were linear functions. Covariates including fuel use, other local sources of air pollution, urbanicity, season, and CO measurement method were added to the models to determine the extent to which their inclusion improved the R^2. We incorporated a random intercept for study into the linear regression models to account for clustering of data by study. The R^2 values were compared to quantify the proportion of variation in $\ln(PM_{2.5})$ explained by $\ln(CO)$ alone and after including other covariates in the models. Differences in the slope of $\ln(PM_{2.5})$ on $\ln(CO)$ by fuel use, urbanicity, season, and CO measurement method were also compared to evaluate transportability (i.e., similarity) of the $PM_{2.5}$–CO relationships between study conditions. Finally, in studies for which personal exposure and cooking area measurements of $PM_{2.5}$ and CO were concurrent, we graphically compared the slopes of the personal exposure versus cooking area $PM_{2.5}$–CO relationships to assess within-study transportability of the cooking area $PM_{2.5}$–CO relationship to personal exposures.

As sensitivity analyses, we conducted the same models with untransformed CO. We also conducted the analyses without nonwood biomass (e.g., dung, charcoal), which may differ from wood in its proportional contribution of PM and CO to

overall emissions (Jetter et al. 2012), and excluding studies not meeting the U.S. EPA (2016) Quality Assurance Guidelines for gravimetric PM analysis ($n=2$). Finally, we compared the R^2 values for univariate and multivariate models within studies to investigate the extent to which individual-level covariate heterogeneity improved explanation of variation in $\ln(PM_{2.5})$ by $\ln(CO)$. All model assumptions were verified by routine diagnostic analysis of the residuals. The statistical analysis was conducted using Stata 13.1 (Stata Corporation, College Station, Texas, USA).

Results

Systematic Review of the Literature

Our search criteria yielded 70 studies, including 2 unpublished studies that were eligible for review, representing measurements in 27 countries. Of these, we retained 61 studies for review after excluding 5 studies of outdoor wildfires, 3 studies of emissions measurements, and 1 urban outdoor air pollution study. Publication year ranged from 1968 to 2016, though most studies (92%) were published after 2000. Studies were conducted in Sub-Saharan Africa ($n=12$), Latin America ($n=23$), South and East Asia ($n=16$), Eastern Mediterranean ($n=1$), and Western Pacific ($n=9$).

Studies with paired measurements of personal exposures to $PM_{2.5}$ and CO in adults and/or children accounted for 23% ($n=14$) of all studies reviewed (Table 1). Sample sizes ranged from 10 to 268 paired measurements (median = 80 pairs). Twelve of the 14 studies enrolled women who were the primary household cooks; in 2 studies, all enrolled participants were pregnant (St. Helen et al. 2015; Wylie et al. 2016). One study enrolled children 15–61 months of age (Dionisio et al. 2012), and another (Naeher et al. 2000a) enrolled mother–child pairs in which both mother and child (<15 months) wore the CO and $PM_{2.5}$ monitors.

Personal $PM_{2.5}$ and CO exposures were integrated over 22-, 24-, or 48-hr periods to represent "usual" daily exposure. Most studies ($n=12$ of 14) measured personal $PM_{2.5}$ exposures with gravimetric instruments. Nine studies used a sensor-based method for CO measurement, and five used a colorimetric dosimeter.

Studies of paired cooking area $PM_{2.5}$ and CO concentrations comprised 93% ($n=57$) of those identified in this systematic review (see Table S5). Sample sizes ranged from 9 to 350 paired measurements (median = 60 pairs). Most stationary $PM_{2.5}$ and CO concentrations were measured in kitchens and cooking areas located in the same building as the living quarters, although some were conducted in rooms adjacent to the kitchen or in separate cookhouses. In five studies (Fischer and Koshland 2007; Pearce et al. 2009; Leavey et al. 2015; Muralidharan et al. 2015; Saksena et al. 1992), $PM_{2.5}$ and CO measurements were limited to cooking events, but the rest were integrated over 22-, 24-, or 48-hr periods. Light-scattering, optical techniques ($n=27$) and integrated, gravimetric techniques ($n=30$) were used for $PM_{2.5}$ measurements. Of the studies with optical $PM_{2.5}$ measurements, 85% measured CO with an electrochemical or optical sensor. Of studies with gravimetric measurements of $PM_{2.5}$, CO was measured with a sensor in 67% of studies ($n=20$) and with a colorimetric dosimeter in 33% of studies ($n=10$).

Correlations between paired $PM_{2.5}$ and CO personal exposure measurements. Correlation coefficients (Spearman r) were reported or calculated for 11 of the 14 studies measuring personal exposures (Table 1). The highest correlation ($r=0.97$) was observed in the study with the smallest sample size, namely 12 observations from mother–child pairs using biomass in open fires and traditional stoves in Guatemala (Naeher et al. 2000a). In the remaining studies, the correlations ranged from $r=0.22$ to $r=0.71$ [$n=10$ studies; median = 0.53; interquartile range (IQR) = 0.34–0.68]. Personal $PM_{2.5}$–CO correlations were generally higher for studies reporting exclusive use of biomass fuel ($n=9$ studies; median = 0.60; IQR = 0.49 – 0.71), conducted in rural settings ($n=10$ studies; median = 0.64; IQR = 0.53 – 0.77), and using sensor-based CO measurements ($n=9$ studies; median = 0.57; IQR = 0.41 – 0.71).

Correlations between paired measurements of cooking area $PM_{2.5}$ and CO concentrations. Correlation coefficients were reported or calculated in 45 of the 57 studies with paired $PM_{2.5}$

Table 1. Characteristics of studies with paired measurements of personal exposure to $PM_{2.5}$ and CO.

Author/year (country)	Fuel(s)[a]	Other local air pollution sources	CO/PM method		$PM_{2.5}$–CO correlation[f] (correlation coeff; r)
			CO S[b]/D[c]	PM G[d]/LS[e]	
Cynthia et al. 2008 (Mexico)	Wood	ETS[g]	S	LS	0.82 ($n=45$) preintervention 0.84 ($n=45$) postintervention
Balakrishnan et al. 2015 (India)	Wood, dung		S	G	0.49 ($n=45$)
Commodore et al. 2013 (Peru)	Wood		S	LS	0.41 ($n=19$[h])
Dionisio et al. 2012 (The Gambia)	Wood		D	G	0.22 ($n=29$)
Ellegård and Egnéus 1993 (Zambia)	Wood, charcoal, electricity	ETS	D	G	NR[i] ($n=268$)
Fitzgerald et al. 2012 (Peru)	Wood		S	G	0.68 ($n=80$)
Hartinger et al. 2013 (Peru)	Wood	ETS	S	G	NR ($n=79$)
McCracken et al. 2013 (Guatemala)	Wood		S	G	0.70 ($n=216$)
Mukhopadhyay et al. 2012 (India)	Wood, dung, LPG[j]		S	G	NR ($n=10$)
Naeher et al. 2000a (Guatemala)	Wood		D	G	0.97 ($n=12$)
Ni et al. 2016 (China)	Wood	ETS	D	G	0.60 ($n=22$)
Peel JL, written and oral communication, 2016 (Honduras)	Wood		S	G	0.57 ($n=105$)
St. Helen et al. 2015 (Peru)	Wood, coal, LPG, kerosene	ETS	S	G	0.33 ($n=93$)
Wylie et al. 2016 (Tanzania)	Wood, charcoal, kerosene	ETS; major road ≤200 m	D	G	0.34 ($n=118$)

[a]Biomass (e.g., wood, crop residue, dung) and non-biomass fuels.
[b]Sensor-based.
[c]Colorimetric/diffusion-based.
[d]Gravimetric.
[e]Light-scattering.
[f]Spearman correlation.
[g]Environmental tobacco smoke.
[h]4-Hr mean CO and $PM_{2.5}$ concentrations.
[i]Not reported.
[j]Liquefied petroleum gas.

and CO measurements in cooking areas (see Table S5) and ranged from $r = 0.10$ to $r = 0.96$ (median = 0.71; IQR = 0.54 – 0.80). Overall, the $PM_{2.5}$–CO correlations were higher for studies with exclusive biomass use ($n = 18$ studies; median = 0.74; IQR = 0.65 – 0.86) than use of multiple fuels ($n = 26$ studies; median = 0.64; IQR = 0.50–0.79) but the same in rural ($n = 37$ studies, median = 0.71, IQR = 0.53 – 0.80) and peri-urban settings ($n = 7$ studies; median = 0.72; IQR = 0.54–0.80). The $PM_{2.5}$–CO correlations were similar for all combinations of $PM_{2.5}$ and CO measurement techniques and in homes with or without a tobacco or pipe smoker. In one-third of studies reviewed, the authors reported $PM_{2.5}$–CO correlation coefficients for subscript-group analyses (see Table S5). Within studies, the $PM_{2.5}$–CO correlation was often higher for observations in rural settings or where wood was burned in open fires or traditional stoves.

Results from Pooled Data Analyses of Paired $PM_{2.5}$ and CO Measurements

Paired personal exposures to $PM_{2.5}$ and CO. The $PM_{2.5}$ and CO personal exposure means, ranges (see Table S6), and correlations varied between studies (Figure 2). The overall $PM_{2.5}$–CO correlation was $r = 0.36$ [95% confidence intervals (CI): 0.30, 0.42; $n = 714$ pairs] (Figure 2a). The majority of study participants lived in rural settings (68%) and used biomass fuels exclusively (80%). Participants who did not use biomass exclusively also used other fuels including liquefied petroleum gas (LPG; 9%), charcoal (6%), kerosene (3%), coal (2%), and electricity (0.1%). The $PM_{2.5}$–CO correlation among only those living with a tobacco or pipe smoker ($n = 46$ pairs) was low ($r = 0.12$, 95% CI: −0.17, 0.40). All $PM_{2.5}$ exposure measurements were gravimetric. Most (75%) CO observations were sensor-based, and the rest were colorimetric-based.

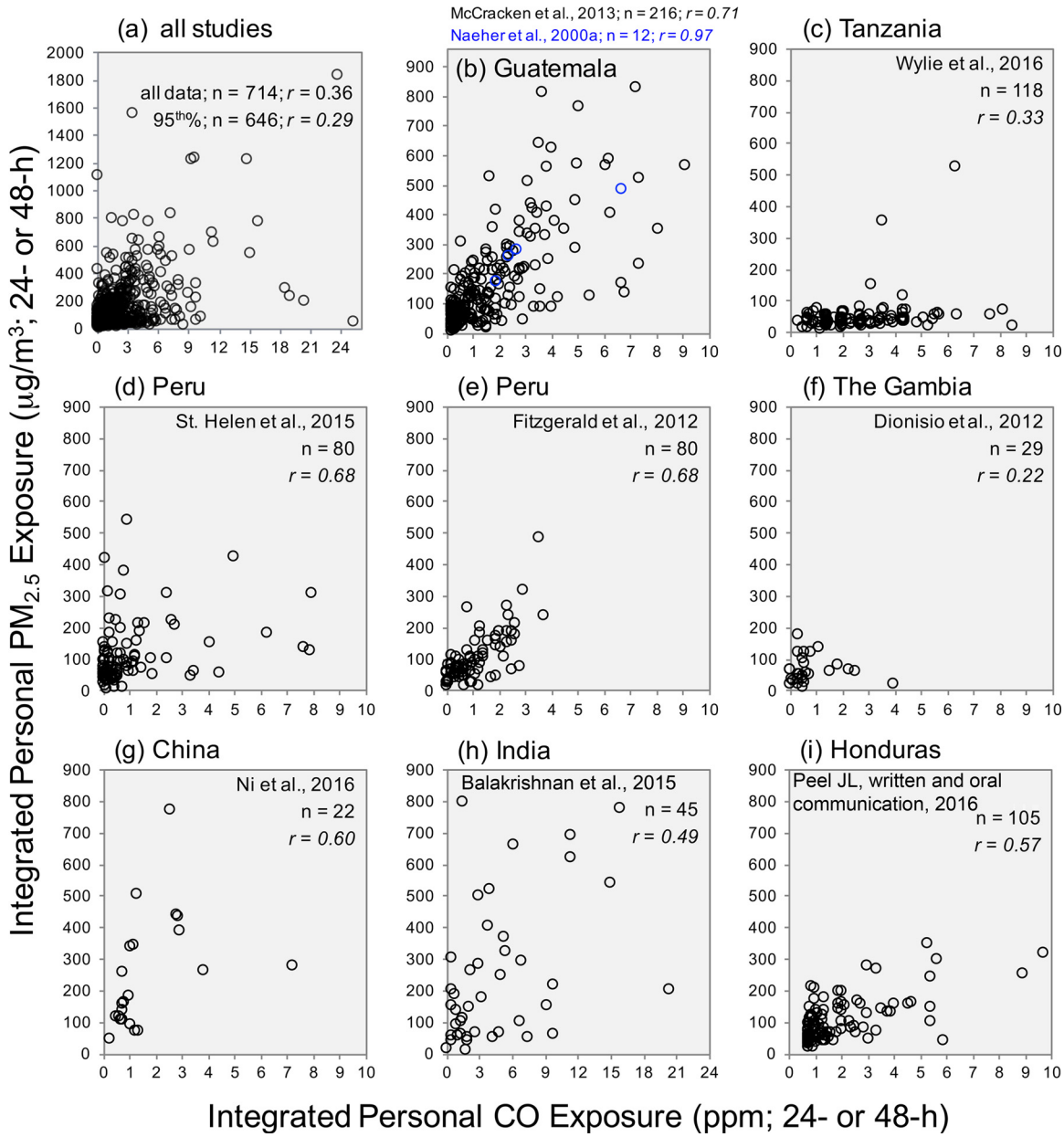

Figure 2. Paired personal $PM_{2.5}$ and personal CO exposure measurements for (*a*) all observations combined from nine studies and for (*b–i*) individual studies. One outlying data point for Tanzania (CO: 25.2 ppm, $PM_{2.5}$: 42.9 $\mu g/m^3$), one for Peru (CO: 25.2 ppm, $PM_{2.5}$: 42.9 $\mu g/m^3$), two for Guatemala (CO: 18.5 ppm, $PM_{2.5}$: 284 $\mu g/m^3$; CO: 23.6 ppm, $PM_{2.5}$: 1,843 $\mu g/m^3$), and two for India (CO: 14.7 ppm, $PM_{2.5}$: 1,226 $\mu g/m^3$; CO: 9.5 ppm, $PM_{2.5}$: 1,243 $\mu g/m^3$) are not pictured to improve data visualization. 2*h* has an expanded CO concentration range along the horizontal axis.

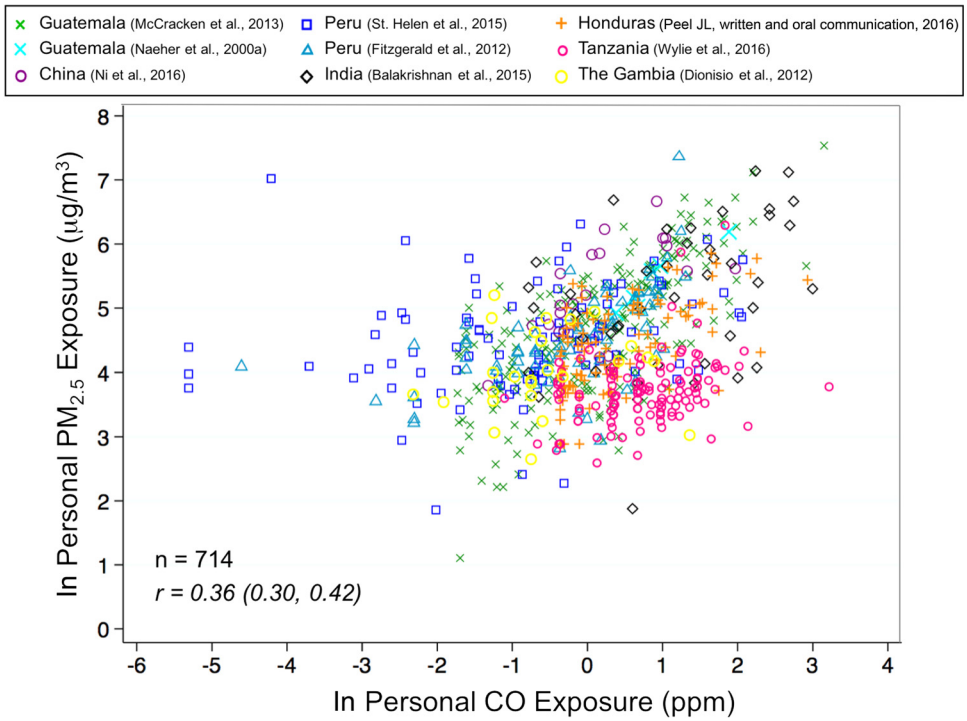

Figure 3. Natural log-transformed $PM_{2.5}$ personal exposures versus natural log-transformed CO personal exposures plotted for nine unique studies. The Spearman correlation ($\pm 95\%$ confidence intervals) for all observations ($n = 714$ pairs) is presented at the bottom left of the figure.

Associations between personal exposures to $PM_{2.5}$ and CO in the pooled analysis. Pollutant concentrations for personal exposure to $PM_{2.5}$ and CO were not normally distributed (right-skewed), and were natural log-transformed prior to evaluating their relationship using scatter plots (Figure 3) and locally weighted scatter plot smoothing and natural cubic spline models (see Figure S1a,b). Visual inspection of these plots indicated that the personal $\ln(PM_{2.5})$–$\ln(CO)$ relationship was approximately linear.

None of the univariate or multivariate linear regression models explained more than 50% of the variance in $\ln(PM_{2.5})$ exposure. The proportion of variation in $\ln(PM_{2.5})$ exposure explained by ln(CO) exposure was 13% with CO alone in the model and 19% in the model including fuel use, urbanicity, season, and CO method (Figure 4). Restricting the multivariate analysis to observations conducted in rural settings ($n = 478$) or during the heating season ($n = 453$) resulted in the highest explanation of variation in $\ln(PM_{2.5})$, namely 42% and 47%, respectively (Figure 4). Excluding two studies not meeting the U.S. EPA Quality Assurance Guidelines for gravimetric analysis did not substantially change our results ($n = 376$ pairs; $R^2 = 0.16$).

We observed significant differences in the $\ln(PM_{2.5})$–$\ln(CO)$ slope by fuel use, level of urbanicity, and season (all interaction

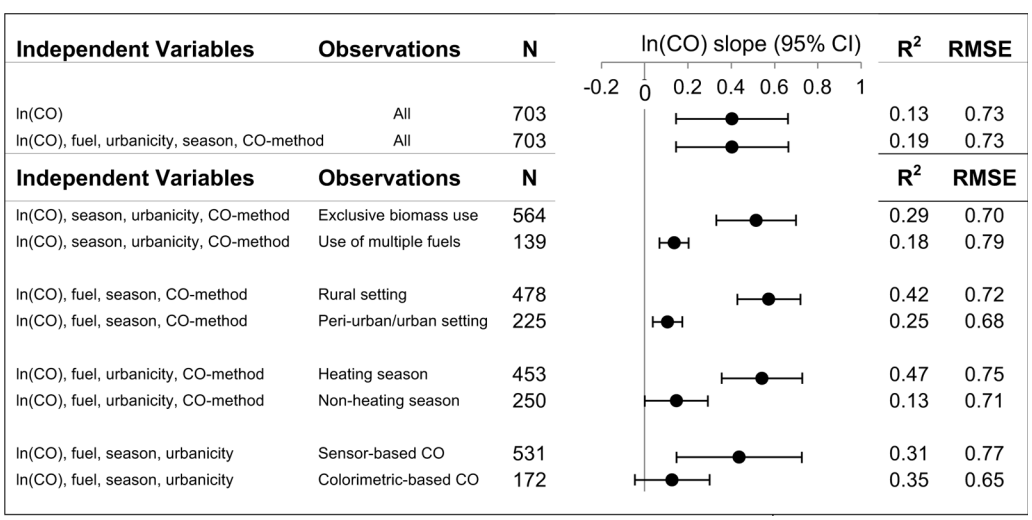

Figure 4. Comparison of estimates of the slope of $\ln(PM_{2.5})$ on ln(CO) ($\pm 95\%$ confidence intervals) for personal exposures using univariate and multivariate linear regression models for the full data set and stratified by subsets of the data. The R^2 values and root mean squared error (RMSE) for each model are reported to the right of the plotted ln(CO) slope. Note: CI, confidence interval; RMSE, root mean squared error.

p-values <0.02). The slope was three to five times greater for measurements with exclusive use of biomass fuel, in rural settings, and during the heating season (Figure 4). In one study, the $PM_{2.5}$–CO relationship also varied by whether the measurements were conducted pre- versus postintervention. In rural Peru (Fitzgerald et al. 2012), the slope of $\ln(PM_{2.5})$ on $\ln(CO)$ was 0.50 (95% CI: 0.30, 0.70; $n=41$) among participants cooking with open fires, which was twice that of the slope among participants using chimney stoves (0.22, 95% CI: 0.03, 0.42; $n=36$) in the same setting (interaction p-value <0.05).

Paired stationary concentrations of $PM_{2.5}$ and CO in cooking areas. Cooking area $PM_{2.5}$ and CO concentration means and ranges (see Table S7) and the strength of the $PM_{2.5}$–CO correlation varied by study ($n=18$ studies; median $=0.80$; range $=0.10$–0.92; IQR $=0.57-0.82$). After combining the paired cooking area $PM_{2.5}$ and CO concentrations from these studies, the $PM_{2.5}$–CO correlation was $r=0.46$ ($n=1{,}336$; 95% CI: 0.42–0.50) but improved to $r=0.74$ ($n=981$; 95% CI: $0.71-0.76$) after removing 350 observations (26% of all observations) from a study in India with a $PM_{2.5}$–CO correlation of $r=0.10$ (Balakrishnan et al. 2013). The CO measurements in the India data set had minimal variability (range: $0.2-11.0$ ppm; IQR: 0.3–3.0 ppm), whereas the PM measurements ranged from 25 to 8,820 $\mu g/m^3$. As the low CO variability could be attributable to instrument failure, these data were excluded from subsequent analyses. Of the remaining 981 observations, biomass was the primary cooking fuel for 82% ($n=807$) of observations, followed by LPG (12%), dung (4%), kerosene (0.5%), coal (1.4%), and electricity (0.1%). Over 86% ($n=847$) of observations were conducted in rural settings, and 64% ($n=625$) took place during the nonheating season.

Associations between cooking area concentrations of $PM_{2.5}$ and CO in the pooled analysis. A natural cubic spline model of $\ln(PM_{2.5})$ and $\ln(CO)$ with three knots was consistent with a linear relationship (see Figure S2). The proportion of variation in $\ln(PM_{2.5})$ concentrations explained by $\ln(CO)$ concentrations was 48% in both the univariate and the multivariate models, which included fuel use, setting, season, and CO method (Figure 5). The $\ln(PM_{2.5})$–$\ln(CO)$ slope for cooking area measurements was twice as large in homes exclusively using biomass fuels compared with homes using multiple fuels (interaction p-value <0.001) and in rural compared with peri-urban settings (interaction p-value <0.001) (see Figure S3). The slope for cooking area measurements did not significantly differ by season (interaction p-value $=0.18$) or CO method (interaction p-value $=0.73$).

Comparison of personal exposure and cooking area $PM_{2.5}$–CO correlations and slopes. For five studies (Dionisio et al. 2012; Fitzgerald et al. 2012; Ni et al. 2016; Peel JL, written and oral communication, 2016; St. Helen et al. 2015), personal exposure and cooking area $PM_{2.5}$ and CO measurements were collected concurrently (Figure 5). With the exception of a study in China (Ni et al. 2016), the R^2 value was considerably higher for cooking area measurements than for personal exposures in the same study, suggesting that studies planning to use personal CO exposures as a surrogate for personal $PM_{2.5}$ exposures would benefit from prior validation studies of personal exposure measurements, rather than cooking area measurements alone. In four of the five studies shown in Figure 5, the slope of $\ln(PM_{2.5})$ on $\ln(CO)$ concentrations in cooking areas was two to eight times steeper than the slope of $\ln(PM_{2.5})$ on $\ln(CO)$ exposures in the same study, which further suggests that use of cooking area concentrations to develop a model to estimate personal $PM_{2.5}$ from measurements of personal CO exposure could lead to biased estimates.

Conducting our multivariate models with untransformed CO concentrations and exposures did not change our overall results (see Tables S8 and S9). In a subset of studies, adding covariates at the individual level in the models led to modest changes (3–19%) in the explanation of variation in ln(PM) explained by ln(CO) in the fully adjusted model relative to the univariate model (see Table S10). Removing observations where dung ($n=49$ kitchens) or wood-charcoal ($n=42$ exposures) or coal ($n=13$ exposures) was used as the primary fuel with biomass did not appreciably change our results (data not shown). The variance inflation factors for our independent variables did not exceed 2.5, indicating a lack of multicollinearity.

Discussion

Our results suggest that exposure to CO is not a consistently valid surrogate of exposure to $PM_{2.5}$ in settings with household air pollution, as indicated by low-to-moderate personal $PM_{2.5}$–CO correlations [range: 0.22 ($n=29$)–0.97 ($n=12$); median $=0.57$]. None of the multivariate regression models explained >50% of the variation in personal $PM_{2.5}$ exposures. Further, the personal $PM_{2.5}$–CO relationship was not transportable across different energy-use and environmental settings, suggesting that, if personal CO exposure is pursued as a surrogate measure of personal $PM_{2.5}$ exposure, a separate $PM_{2.5}$–CO validation may be needed for each unique study setting and, within studies, potentially each season or pre- versus post-stove/fuel intervention.

We found a stronger correlation between personal $PM_{2.5}$ and CO exposures among exclusive biomass users relative to mixed fuel users ($R^2=0.29$ vs. 0.18), supporting previous studies (Naeher et al. 2000a; Naeher et al. 2000b; McCracken et al. 2013). This finding is consistent with results from stove emission tests in laboratory and field settings. For example, Jetter et al. (2012) observed a higher coefficient of determination for $PM_{2.5}$ versus CO emissions from biomass stoves compared with nonbiomass stoves during Water Boiling Tests (see Figure S4). This study and others describe fundamental sources of variability in combustion conditions and energy-use behaviors that limit the strength and consistency of the correlation we may expect for $PM_{2.5}$ and CO exposures and concentrations in real-world settings with biomass combustion (Zhang et al. 2000; Roden et al. 2009; Shen et al. 2010; Chen et al. 2012; Shen et al. 2013). With widespread use of multiple stoves and fuels (i.e., stove stacking), exclusive biomass use is increasingly less common (Masera and Navia 1997; Masera et al. 2000; Ruiz-Mercado et al. 2011, Rehfuess et al. 2014; Ni et al. 2016;) and may reduce the number of settings in which validation studies will demonstrate CO to be a valid PM surrogate.

We observed a stronger personal $PM_{2.5}$–CO relationship for measurements conducted in rural versus peri-urban settings ($R^2=0.42$ vs. 0.25; interaction p-value <0.001), likely because densely populated peri-urban neighborhoods may have more community (i.e., solid waste burning) and regional pollution. At the same time, a stronger $PM_{2.5}$–CO relationship for personal exposure measurements conducted in the heating season relative to the nonheating season ($R^2=0.47$ vs. 0.13; interaction p-value <0.001) may reflect the greater proportion of time people spend indoors next to the fire, which is also where stationary indoor monitors are usually located. Notably, the R^2 for the personal $PM_{2.5}$–CO exposure relationship in the heating season is almost identical to the cooking area relationship (0.47 vs. 0.46). Separately, we found that the personal $PM_{2.5}$–CO relationship was modified significantly by season (interaction p-value $=$ 0.02), but the cooking area $PM_{2.5}$–CO relationship was not (interaction p-value $=0.34$). This finding supports recent studies showing that personal exposures are impacted by other (i.e. noncooking) air pollution sources (Baumgartner et al. 2014;

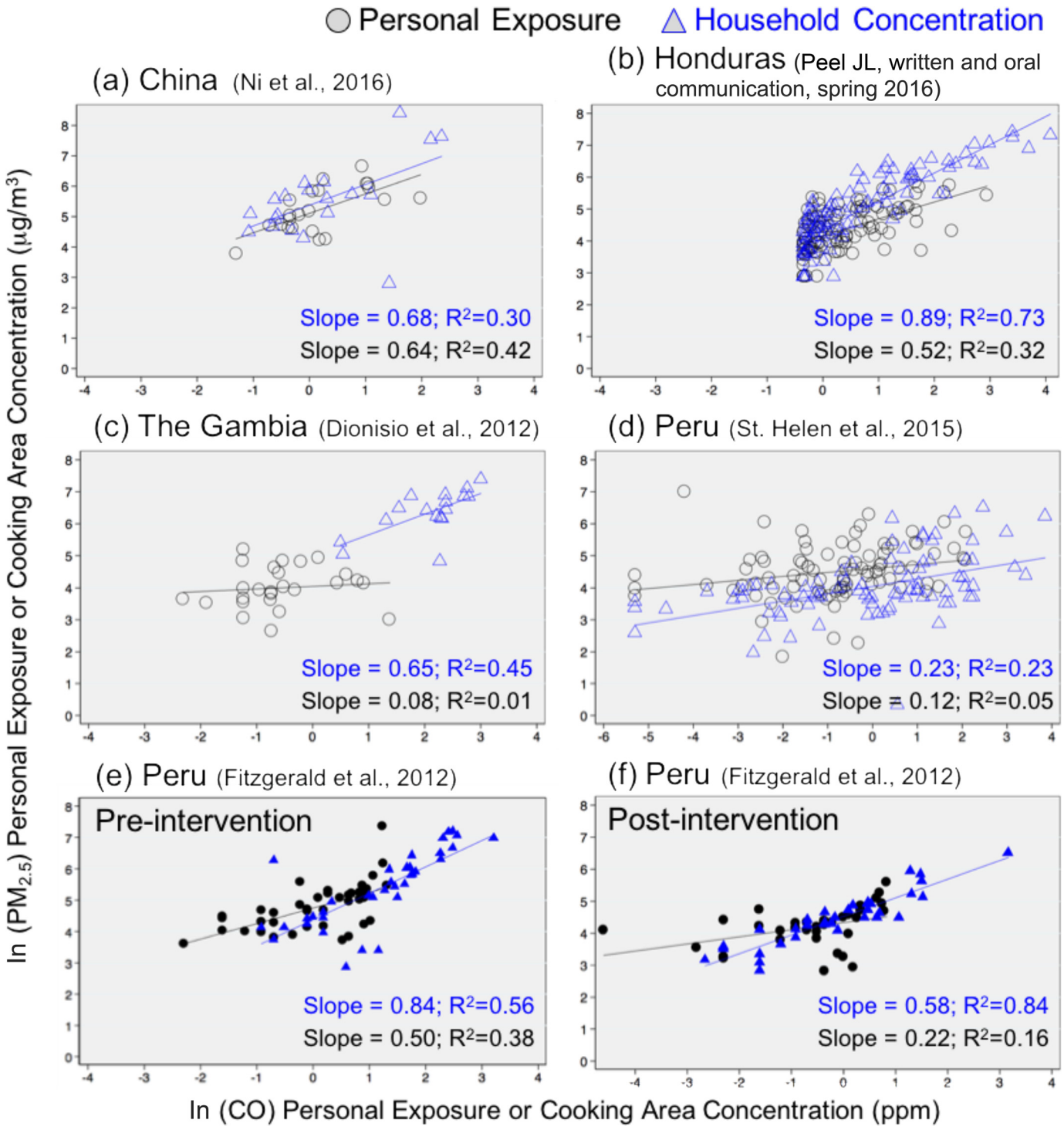

Figure 5. Paired personal and cooking area $PM_{2.5}$ and CO (24- or 48-hr integrated concentrations) for (*a*) China (Ni et al. 2016), (*b*) Honduras (Peel JL, written and oral communication, spring 2016), (*c*) The Gambia (Dionisio et al. 2012), (*d*) Peru (St. Helen et al. 2015), and Peru (*e*) pre- and (*f*) postintervention (Fitzgerald et al. 2012). The R^2 and slope of the $\ln(PM_{2.5})$-$\ln(CO)$ relationship is shown for cooking area measurements (blue) and personal exposures (black).

Huang et al. 2015; Secrest et al. 2017). These other air pollution sources impact noncooking area measurements and weaken the basis for transportability of the cooking area $PM_{2.5}$–CO relationship to personal exposures. Using a cooking area $PM_{2.5}$–CO relationship to estimate personal $PM_{2.5}$ from measurements of personal CO exposure may yield inaccurate results, as our graphical comparison of these two relationships from the same studies suggests (Figure 5).

The $PM_{2.5}$–CO exposure relationships may also vary by age or gender. Although 24-hr CO and $PM_{2.5}$ exposures were measured in participants ranging from 18 months to 90 years of age, most exclusively measured adult women's exposures, highlighting the limited data on exposures in infants and young children and the need to reduce technological barriers to measuring their exposure. No studies evaluated this relationship in men, though it is unlikely that the $PM_{2.5}$–CO relationship would be stronger in

men who, in many settings, are less likely to be the primary cooks and more likely to spend time outside of the home.

The type and range of pollutants of interest may vary depending on the health endpoint. Though the exact PM components responsible for its health impacts are unclear, there is strong and consistent evidence that both short- and long-term exposures to $PM_{2.5}$ are associated with a range of clinical health outcomes in adults and children (WHO 2007; Chen et al. 2008; U.S. EPA 2009; Brook et al. 2010), including a number of PM exposure–response studies conducted in settings of biomass burning (Ezzati and Kammen 2001; Smith et al. 2011; Baumgartner et al. 2011; Norris et al. 2016). The evidence base for direct health impacts of CO exposure, beyond acute poisoning, is less strong. Animal studies indicate that fetal carboxyhemoglobin levels equilibrate with maternal levels (Longo 1977), and that very high maternal CO exposures are associated with adverse pregnancy outcomes, including pregnancy loss and low birth weight (Astrup et al. 1972; Garvey and Longo 1978). In epidemiologic studies, exposure to low-to-moderate CO concentrations in pregnant women was associated with reduced fetal growth in some studies (Ritz and Yu 1999; Ha et al. 2001; Gouveia et al. 2004; Salam et al. 2005) but not others (Alderman et al. 1987; Koren et al. 1991; Chen et al. 2002). Notably, a recent study that measured both personal $PM_{2.5}$ and personal CO exposure in pregnant Tanzanian women cooking with biomass stoves found that only $PM_{2.5}$ exposure was associated with adverse birth outcomes (Wylie et al. 2016), supporting a similar finding among pregnant women in the urban United States (Parker et al 2005).

A strength of our pooled analysis is the inclusion of multiple independent variables that have been shown to influence the $PM_{2.5}$–CO relationship. Still, it is possible that inclusion of other individual- or study-level variables could further improve the ability of CO to predict PM exposures or concentrations. For example, we did not have access to detailed socio-demographic data for study participants and could not include variables for altitude, monitor placement, or stove type because these variables were collinear with study, measurement type, and fuel type, respectively, and were thus excluded from the models.

Though our systematic review revealed inconsistencies in the reporting of quality assurance and quality control protocols for $PM_{2.5}$ and CO measurements, which are subject to systematic error and may introduce bias, we recognize that investigators have to balance the scientific, logistical, technical, and cost trade-offs in selecting an exposure metric for their study. Standardized and transparent reporting could improve the comparability of pollution measurements collected across diverse settings. Such reporting would include, for example, filter handling, collection, and transport; field and lab blank correction; duplicate precision estimates, if possible; method detection limits and instrument sensitivity; instrument flow rate precision estimates; and traceability and calibration levels of gas standards used to calibrate CO sensors. Then, the trade-offs in pollutant selection, study design, and measurement precision, stability, practicality, and number—inherent to efforts to reduce exposure misclassification for long-term air pollution exposures (McCracken et al. 2009; Pillarisetti 2016)—could be evaluated more consistently across settings. Further, improving our collective assessment of these trade-offs would bring to light more suitable and effective approaches and technologies to measure exposure, especially among young children and infants, for whom we have the least information on PM exposure (Balakrishnan et al. 2011; Clark et al. 2013).

Conclusions

Our systematic review and pooled analysis suggest that personal CO exposures are a poor surrogate of measured personal PM exposures, even when biomass is exclusively burned. Our conclusions support those reached in recent studies reporting low $PM_{2.5}$–CO correlations for cooking area concentrations (Klasen et al. 2015; Bartington et al. 2016). The relationship between cooking area $PM_{2.5}$ and CO concentrations in this review was stronger than for personal exposures, potentially due to the closer proximity of stationary monitors to the solid fuel emission source, but still the variation in ln(CO) did not explain more than 48% of the variation in $\ln(PM_{2.5})$. Based on the evidence presented in this analysis, to use CO exposure as a surrogate for PM exposure would require repeated validation studies, especially if study conditions change over time. Lowering the barriers to $PM_{2.5}$ exposure assessment, particularly for infants and young children, is an important direction for future research. Recent developments in portable lightweight $PM_{2.5}$ monitors that are virtually silent and low-profile (Birch et al. 2015; Volckens et al. 2017) could expand the feasibility of PM exposure assessment to different populations and settings. Given that $PM_{2.5}$ is likely one of the important drivers of the health effects associated with air pollution exposure, further research and development is needed to minimize $PM_{2.5}$ measurement error, to reduce the logistical and technological challenges of large-scale PM exposure assessment, and to identify better surrogate measures of $PM_{2.5}$ exposure and dose, potentially including internal biomarkers, for epidemiologic and intervention studies involving household air pollution.

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