

Longitudinal Relationship between Personal CO and Personal PM_{2.5} among Women Cooking with Woodfired Cookstoves in Guatemala

John P. McCracken¹, Joel Schwartz¹, Anaite Diaz², Nigel Bruce³, Kirk R. Smith^{4*}

1 Department of Environmental Health, Harvard School of Public Health, Boston, Massachusetts, United States of America, **2** Center for Health Studies, Universidad del Valle de Guatemala, Guatemala City, Guatemala, **3** Department of Public Health and Policy, University of Liverpool, Liverpool, United Kingdom, **4** School of Public Health, University of California, Berkeley, California, United States of America

Abstract

Household air pollution (HAP) due to solid fuel use is a major public health threat in low-income countries. Most health effects are thought to be related to exposure to the fine particulate matter (PM) component of HAP, but it is currently impractical to measure personal exposure to PM in large studies. Carbon monoxide (CO) has been shown in cross-sectional analyses to be a reliable surrogate for particles <2.5 μm in diameter (PM_{2.5}) in kitchens where wood-burning cookfires are a dominant source, but it is unknown whether a similar PM_{2.5}-CO relationship exists for personal exposures longitudinally. We repeatedly measured (216 measures, 116 women) 24-hour personal PM_{2.5} (median [IQR] = 0.11 [0.05, 0.21] mg/m³) and CO (median [IQR] = 1.18 [0.50, 2.37] mg/m³) among women cooking over open woodfires or chimney woodstoves in Guatemala. Pollution measures were natural-log transformed for analyses. In linear mixed effects models with random subject intercepts, we found that personal CO explained 78% of between-subject variance in personal PM_{2.5}. We did not see a difference in slope by stove type. This work provides evidence that in settings where there is a dominant source of biomass combustion, repeated measures of personal CO can be used as a reliable surrogate for an individual's PM_{2.5} exposure. This finding has important implications for the feasibility of reliably estimating long-term (months to years) PM_{2.5} exposure in large-scale epidemiological and intervention studies of HAP.

Citation: McCracken JP, Schwartz J, Diaz A, Bruce N, Smith KR (2013) Longitudinal Relationship between Personal CO and Personal PM_{2.5} among Women Cooking with Woodfired Cookstoves in Guatemala. PLoS ONE 8(2): e55670. doi:10.1371/journal.pone.0055670

Editor: Mehrdad Arjomandi, University of California San Francisco, United States of America

Received: August 3, 2012; **Accepted:** December 31, 2012; **Published:** February 26, 2013

Copyright: © 2013 McCracken et al. This is an open-access article distributed under the terms of the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited.

Funding: This work was supported by the National Institutes of Health (grants R01ES010178, P01-ES09825, T-32 ES07069-25, ES-0002, and ES015172), the World Health Organization, and the U.S. Environmental Protection Agency (grants R827353 and R832416). The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

Competing Interests: The authors have declared that no competing interests exist.

* E-mail: krksmith@berkeley.edu

Introduction

Household air pollution (HAP) from use of solid fuels is estimated to be a major risk factor for diseases, including acute respiratory, chronic respiratory, cancer, and cardiovascular outcomes [1,2]. Most of the epidemiological evidence for these relationships comes from studies using categorical exposure assignments based on stove and fuel types, which does not allow exposure-response analyses and limits comparability between studies in different settings. An ideal study design would include personal measures of exposure to the component of HAP that is causally related to the health effects being investigated. Fine particulate matter (PM_{2.5}) is often considered the best pollutant to measure for studies of health effects from combustion-generated pollutant mixtures, including HAP, secondhand tobacco smoke, and ambient air pollution [3,4]. Because of the size and weight of the monitoring equipment that has been available, personal PM measurements are generally burdensome and for infants infeasible, a particularly important limitation given the importance of quantifying the exposure-response relationship between HAP and pneumonia during infancy [5].

To overcome this problem, some HAP epidemiological studies have used area measurements of pollutant concentrations as

surrogates for personal exposures. Kitchen area measures have been found to be poor surrogates of personal exposures to HAP [6–8], which may be largely attributable to differences in people's time-location patterns and the wide variability across small distances within the household and over short time periods [9]. Indirect exposure assessment, using time-activity patterns combined with area measurements [9,10], may improve exposure assessment, but one study with simultaneous personal exposure measures indicated that this method has low validity [8].

An alternative approach to HAP exposure assessment is personal measurement of a surrogate pollutant for PM, such as carbon monoxide (CO), which is relatively easy and inexpensive to measure, for example with very small passive dosimeter tubes that can be attached to an infant's clothing. Both pollutants are products of incomplete combustion and are major components of biomass smoke [11]. Strong correlation has been found between CO and fine PM levels in kitchens where biomass fuels are used for cooking [12–14]. It has been unknown, however, whether the relationship between these pollutants in a fixed location can be extrapolated to personal exposures. Additionally, the aim of most HAP epidemiological studies is to investigate effects of long-term (several months to years) exposures, whereas the relationships

between CO and PM have previously been evaluated only in cross-sectional designs [12,14] or analyses [13].

The RESPIRE (Randomized Exposure Study of Pollution Indoors and Respiratory Effects) trial in Guatemala, the first randomized trial of an HAP exposure-reduction intervention, a chimney woodstove [5], for the prevention of pneumonia, included personal exposure measurements among a subset of women living in the study households. This short note presents a longitudinal analysis of the relationship between personal CO and PM among these women.

Methods

The study population and exposure assessment methodology have been described previously [15,16]. Briefly, women ≥ 38 years of age living in households participating in RESPIRE were recruited for a cardiovascular study. The study villages are located in the San Marcos department at approximately 2600 meters elevation above sea level. Smoking is uncommon, automobile traffic is low, and study households used only biomass fuels for cooking. The exposure assessment included a gravimetric (pump flow rate at 1.5 liters/minute, BGI Inc. sharp-cut cyclone inlet, 37 mm Teflon filter weighed before and after) measure of 24-hour personal exposure to particles with median aerodynamic diameter $< 2.5 \mu\text{m}$ (PM_{2.5}). Simultaneously, continuous measurement of personal CO was performed with the span-gas calibrated Hobo (Onset Inc.) passive electrochemical datalogger, with conversion of CO ppm values to mass concentration for comparison with the PM mass concentrations [17]:

$$\text{mg/m}^3 = (\text{ppmv}) \times (12.187) \times (\text{MW}) / (273.15 + C) \times (0.9877^A) \quad (1)$$

where the molecular weight (MW) of CO is 28.01,

C, the mean temperature at the site, is 12 deg celsius

A is the elevation of each house in 100 meters (range 2250–2960 m)

We analyze measures (up to three per subject) taken during the trial period, when the intervention group used the chimney stove and the control group used the open fire for cooking.

Pollution measures were right-skewed, so we applied a natural log transformation to the data before assessing the relationship between personal CO and personal PM_{2.5} by scatterplot, correlation coefficients, and regression models. We used linear mixed effects models with personal PM_{2.5} as the dependent variable and random subject intercepts to account for correlation among repeated measures within subjects and to estimate the within- and between-subjects variance components. The model residuals were consistent with being derived from a normal distribution. We compared the variance of the random subject intercept between models to measure the extent to which between-subjects differences in typical personal PM_{2.5} are explained by covariates (R^2_{between}). For example, we estimated the R^2_{between} for a model with CO as the independent variable by calculating the proportional reduction in the variance of the random subject intercept compared to the null model (no independent variable). The fixed effects in these models can be used to estimate personal PM_{2.5} based on covariates (stove, personal CO). To test for differences in the slope of PM_{2.5} on CO by stove type, we added a stove-by-CO interaction term. We tested for nonlinearity using a penalized spline for CO in a generalized additive mixed model (R software, GAMM function).

Protocols were approved by the Comité de Ética de la Universidad del Valle de Guatemala and the Harvard School of

Public Health, Office of Human Research Administration. Written consents were obtained from all participants.

Results

We obtained 216 simultaneous 24-hour measures of CO and PM_{2.5} among 116 women, 40 on one occasion, 52 on two occasions, and 24 on three occasions. The median (interquartile range) personal PM_{2.5} was 0.20 mg/m³ (0.11, 0.32) in the open fire group (67 women, n = 104) and 0.07 mg/m³ (0.04, 0.12) in the chimney stove group (49 women, n = 112), and personal CO was 2.02 mg/m³ (1.20, 3.35) in the open fire group and 0.63 mg/m³ (0.33, 1.22) in the chimney stove group. Figure 1 shows a direct relationship between the natural log-transformed values of personal CO and PM_{2.5} exposures. The Spearman rank correlation coefficient was 0.70 (p-value < 0.001) between these two pollutant exposures (see Table 1).

In linear mixed effects models, the variance of the random intercept decreased from 0.31 to 0.07 when CO was added as an independent variable, equivalent to an $R^2_{\text{between}} = 0.78$. A further reduction in random between-subject variability to 0.04 was achieved when stove type (chimney stove versus open fire) was added to the model ($R^2_{\text{between}} = 0.85$).

The estimated population-mean personal PM_{2.5} based on personal CO alone can be calculated with the following equation:

$$\text{PM}_{2.5} = e^{(-2.13 + 0.61 \cdot \ln(\text{CO}) - 0.36 \cdot \text{chimney})}, \quad (2)$$

where chimney = 1 for the chimney stove and chimney = 0 for open fire.

We did not find evidence of a difference in the slope by stove type (interaction p-value = 0.986), and we also did not find evidence of nonlinearity in these log-transformed data using generalized cross validation, which chose a spline with one degree of freedom (Figure 1).

Discussion

Absent or minimal assessment of exposure to combustion-generated PM has been a major weakness of most epidemiological

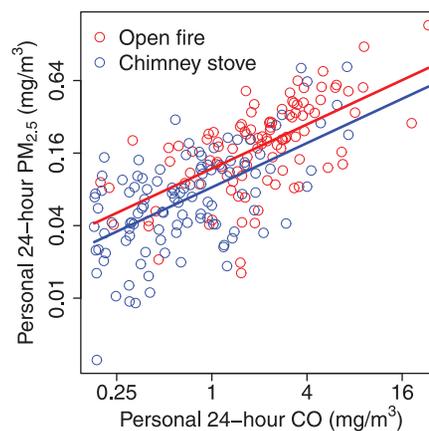


Figure 1. Scatter plot of simultaneous 24-hour personal fine particles (PM_{2.5}) and personal carbon monoxide (CO). Lines for each stove type (red for open fire, blue for chimney stove) using equation $\text{PM}_{2.5} = e^{(-2.13 + 0.61 \cdot \ln(\text{CO}) - 0.36 \cdot \text{chimney})}$ estimated from linear mixed effects regression model with natural log-transformed exposures (216 measurements among 116 women). doi:10.1371/journal.pone.0055670.g001

Table 1. Effect estimates (95% confidence intervals) and variance components from linear mixed effects models to predict natural log personal PM_{2.5} (216 24-hour exposure measures among 116 subjects).

Independent variables	Chimney stove Effect	CO slope (per log-unit)	Between-subject variance	Within-subject variance	R ² _{between}
Null			0.31	0.76	
CO		0.69 (0.59, 0.79)	0.07	0.48	0.78
Stove type	-1.00 (-1.25, -0.74)		0.07	0.75	0.77
Stove type and CO	-0.36 (-0.59, -0.12)	0.61 (0.50, 0.72)	0.05	0.48	0.85
Plus stove by CO interaction*	-0.36 (-0.60, -0.12)	0.61 (0.45, 0.77)	0.05	0.48	0.85

*Stove by CO interaction effect = -0.00 (-0.22, 0.22).
doi:10.1371/journal.pone.0055670.t001

studies of HAP in developing countries, particularly those with the additional challenges presented by assessing these exposures among infants. Previous studies have shown that CO is strongly correlated with PM_{2.5} in kitchens where there is a single major source of smoke, but it was unclear whether this relationship could be extrapolated to personal exposures. We performed a longitudinal analysis of personal exposures among women from households in the RESPIRE trial in Guatemala, and found a moderately, strong correlation between personal CO and personal PM_{2.5}. Repeated personal CO levels explain 78% of the between-subject variability in personal PM_{2.5}. The estimated slope for the relationship between log-transformed measures is a 61% increase in personal PM_{2.5} per 100% increase in personal CO.

Our results contrast with those from a study conducted among children <5 years of age in the Gambia by Dionisio et al [18], who did not find evidence of correlation between personal CO and personal PM_{2.5} ($r = -0.04$), but there are a number of potential explanations for the weak correlations. In that population, there were a mixture of wood and charcoal stoves in use, which have substantially different ratios of CO to PM_{2.5} in their emissions [19], which would reduce the local correlation between personal PM_{2.5} and personal CO [9]. Similarly, the peri-urban children in the Gambian study may have been exposed to high levels of traffic emissions, with an even greater difference in CO:PM_{2.5} in its emissions [20]. Moreover, the durations of personal PM_{2.5} (48 hours) and personal CO (72 hours) measures differed in The Gambian study, which is expected to lower the correlation because of day-to-day variability in exposure levels. Finally, it is possible that instrument measurement error may have led to underestimation of the true correlation. Duplicate measures of each pollutant in a subset of participants should be collected in future studies to account for measurement error. Alternatively, longitudinal analyses can be used to separate within-subject variation (both true changes over time and measurement error) from between-subjects variation, as herein presented.

The form of the relationship between personal CO and personal PM_{2.5} in our models suggests a smaller increment of PM_{2.5} per unit of CO at higher exposure levels. This contrasts with the constant slope on the linear scale reported previously for kitchen

concentrations in Guatemala [12–14], and there are several reasons why personal measurements may exhibit this relationship. Since combustion-generated PM_{2.5} is an irritant and the PM_{2.5}-CO emissions ratio varies throughout the solid fuel burn cycle [11], it is possible that avoidance of the discomfort of PM and associated irritating compounds in the smoke by the householders may decrease the PM_{2.5}-CO slope at higher exposure levels. In addition, particles tend to adhere to surfaces over time whereas CO does not. If people tend to be in the kitchen more after emissions have been exposed to surfaces around the household, this would also decrease the PM_{2.5}-CO slope. Finally, the PM_{2.5}-CO relationship may be different in microenvironments where people spend time other than the kitchen and the relative contribution of each microenvironment may vary by total exposure level.

Conclusions

Our findings demonstrate that personal CO, which is relatively inexpensive and easy to measure can be a reliable surrogate for personal PM_{2.5} in some settings. We emphasize that the association was observed among women living in Guatemalan villages with a single dominant source of combustion, but may be modified by time-activity patterns associated with demographic characteristics, and is unlikely to be generalizable to settings with mixtures of pollution source types.

Acknowledgments

We appreciate insights gained by discussions with Steven Chillrud, Alan Hubbard, Mark Nicas, and Charles Weschler. We thank the Randomized Exposure Study of Pollution Indoors and Respiratory Effects (RESPIRE) fieldworkers for their dedication to the project, and the Guatemalan Ministry of Health for their collaboration.

Author Contributions

Conceived and designed the experiments: JPM JS KRS NB. Performed the experiments: JPM AD. Analyzed the data: JPM KRS. Wrote the paper: JPM NB KRS.

References

- Smith KR, Mehta S (2003) The burden of disease from indoor air pollution in developing countries: comparison of estimates. *International journal of hygiene and environmental health* 206: 279–289.
- Balakrishnan K, Ramaswamy P, Sambandam S, Thangavel G, Ghosh S, et al. (2011) Air pollution from household solid fuel combustion in India: an overview of exposure and health related information to inform health research priorities. *Global health action* 4.
- Pope CA, 3rd, Burnett RT, Turner MC, Cohen A, Krewski D, et al. (2011) Lung cancer and cardiovascular disease mortality associated with ambient air pollution and cigarette smoke: shape of the exposure-response relationships. *Environmental health perspectives* 119: 1616–1621.
- Smith KR, Peel JL (2010) Mind the gap. *Environmental health perspectives* 118: 1643–1645.
- Smith KR, McCracken JP, Weber MW, Hubbard A, Jenny A, et al. (2011) Effect of reduction in household air pollution on childhood pneumonia in Guatemala (RESPIRE): a randomised controlled trial. *Lancet* 378: 1717–1726.
- Bruce N, McCracken J, Albalak R, Schei MA, Smith KR, et al. (2004) Impact of improved stoves, house construction and child location on levels of indoor air pollution exposure in young Guatemalan children. *Journal of exposure analysis and environmental epidemiology* 14 Suppl 1: S26–33.
- Baumgartner J, Schauer JJ, Ezzati M, Lu L, Cheng C, et al. (2011) Patterns and predictors of personal exposure to indoor air pollution from biomass combustion among women and children in rural China. *Indoor air* 21: 479–488.
- Cynthia AA, Edwards RD, Johnson M, Zuk M, Rojas L, et al. (2008) Reduction in personal exposures to particulate matter and carbon monoxide as a result of the installation of a Patsari improved cook stove in Michoacan Mexico. *Indoor air* 18: 93–105.
- Ezzati M, Saleh H, Kammen DM (2000) The contributions of emissions and spatial microenvironments to exposure to indoor air pollution from biomass combustion in Kenya. *Environmental health perspectives* 108: 833–839.
- Zuk M, Rojas L, Blanco S, Serrano P, Cruz J, et al. (2007) The impact of improved wood-burning stoves on fine particulate matter concentrations in rural Mexican homes. *Journal of exposure science & environmental epidemiology* 17: 224–232.
- Smith K (1987) *Biofuels, air pollution, and health: a global review*. New York: Plenum.
- Naeher LP, Smith KR, Leaderer BP, Neufeld L, Mage DT (2001) Carbon monoxide as a tracer for assessing exposures to particulate matter in wood and gas cookstove households of highland Guatemala. *Environmental science & technology* 35: 575–581.
- Northercross A, Chowdhury Z, McCracken J, Canuz E, Smith KR (2010) Estimating personal PM_{2.5} exposures using CO measurements in Guatemalan households cooking with wood fuel. *Journal of environmental monitoring* : JEM 12: 873–878.
- Naeher LP, Leaderer BP, Smith KR (2000) Particulate matter and carbon monoxide in highland Guatemala: indoor and outdoor levels from traditional and improved wood stoves and gas stoves. *Indoor air* 10: 200–205.
- McCracken JP, Smith KR, Diaz A, Mittleman MA, Schwartz J (2007) Chimney stove intervention to reduce long-term wood smoke exposure lowers blood pressure among Guatemalan women. *Environmental health perspectives* 115: 996–1001.
- McCracken J, Smith KR, Stone P, Diaz A, Arana B, et al. (2011) Intervention to lower household wood smoke exposure in Guatemala reduces ST-segment depression on electrocardiograms. *Environmental health perspectives* 119: 1562–1568.
- Seinfeld JH, Pandis SN (1998) *Atmospheric Chemistry and Physics*. New York: John Wiley & Sons. 1326 p.
- Dionisio KL, Howie SR, Dominici F, Fornace KM, Spengler JD, et al. (2012) Household concentrations and exposure of children to particulate matter from biomass fuels in The Gambia. *Environmental science & technology* 46: 3519–3527.
- Smith KR, Uma R, Kishore VVN, Zhang J, Joshi V, et al. (2000) Greenhouse implications of household stoves: an analysis for India. *Annual Review Energy Environment* 25: 741–763.
- Wallington TJ, Sullivan JL, Hurley MD (2008) Emissions of CO₂, CO, NO_x, HC, PM, HFC-134a, N₂O and CH₄ from the global light duty vehicle fleet. *Meteorologische Zeitschrift* 17: 109–116.