Carbon Monoxide As a Tracer for Assessing Exposures to Particulate Matter in Wood and Gas Cookstove Households of Highland Guatemala

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Kitchen-area 22-h gravimetric PM2.5 and passive diffusion stain-tube carbon monoxide (CO) concentrations were measured in homes with open fire and improved wood cookstoves in two studies. In the first study (Guat-2), which also studied homes with gas cookstoves, three samples were collected per stove condition from each of three test houses. In the second study (Guat-3), one sample was collected per house from 15 open fire and 25 improved-stove houses. CO personal samples were also taken for mother and child in both studies. Spearman correlation coefficients (R) between kitchen-area CO and PM2.5 levels in homes using open fires or improved wood cookstoves were high ranging from 0.92 (Guat-2) to 0.94 (Guat-3), as were those between the personal samples for mother and child ranging from 0.85 (Guat-3) to 0.96 (Guat-2). In general, the correlations were lower for less-polluted conditions. The study found that CO is a good proxy for PM2.5 in homes using open fires or planchas (improved wood cookstove with chimney) but not under gas stove use conditions. It also determined that mother personal CO is a good proxy for child’s (under 2 years of age) personal CO and that area CO measurements are not strongly representative of personal CO measurements. These results generally support the use of Draeger CO passive diffusion tubes as a proxy for PM2.5 in such cases where a single type of emission source is the predominant source for CO and PM2.5.

Introduction

It is estimated that around 50% of the world’s population, and as much as 90% of households in some developing countries, rely on biomass fuels (wood, dung, and crop residues) for household cooking and sometimes heating (1, 2). In much of the Third World, these fuels are typically burned in simple open stoves. Such small-scale combustion of biomass fuels results in indoor air pollution levels, including respirable particulate pollution, among the highest ever measured (3, 4); thus, on a worldwide basis, it is these populations that have the greatest exposures (5).

Numerous urban studies in industrialized countries have shown associations between particulate air pollution and acute and chronic respiratory morbidity and mortality in children and adults (6). Although exposure characteristics vary tremendously from the urban developed world to rural areas in the developing world (e.g., particulate composition, exposure circumstances, demographics, and underlying health status), it is hypothesized that high exposure to contaminants from biomass fuel combustion is a risk factor for low birth weight (7), acute respiratory infections (ARI) (8), chronic obstructive pulmonary disease, and other health problems in the developing world. In its 1993 report, Investing in Health, the World Bank estimates that indoor air pollution is responsible for almost 50% of the burden of total disease resulting from poor household environments in developing countries (9).

Further studies from rural areas in developing countries are necessary to clarify the association, including the exposure–response relationship between particulate pollution from biomass fuel cooking and health. In 1992, a committee organized by the World Health Organization (WHO) began to examine the feasibility of carrying out controlled intervention studies designed to assess the effects on key child and adult respiratory health outcomes of a measured reduction in exposure. This has formed part of a review, carried out by the WHO, of potential ARI prevention strategies that would complement case management (10). Pursuant to a set of epidemiological studies to determine the risk-reduction potentials of various interventions (e.g., including fuel substitution, stove alteration, ventilation provision, and behavioral modification), the WHO sponsored several pilot studies in the western highlands of Guatemala (11–14). Building on the research of Smith and colleagues (14), we contributed to this preliminary work through three exposure assessment-related studies in the western highlands of Guatemala in 1993–1994. The objective of the first study, reported elsewhere (15), was to determine if villages in the proposed region had high enough indoor pollution levels from indigenous stoves emissions to make it a viable area for an intervention study to investigate the association between biomass fuel combustion and health effects in adults and children. This study provided a useful picture of the pollution levels coming from a range of cooking stoves in various levels of disrepair, a representation of how outdoor particle mass and CO vary from high- versus low-density villages, and demonstrated that the region has adequate exposure scenarios to be a good site for an intervention study. The objective of the second study (16) was to determine the effectiveness of a range of cooking stoves that are indigenous to the proposed study region in reducing indoor exposures to air pollution. This study found that reductions in indoor levels achieved by bottled gas (LPG) stoves and well-operating planchas (indigenous wood-fueled cookstove with chimney) are apparently to 15–30% of concentrations observed from open fire PM2.5 (particles nominally less than 10 μm aerodynamic diameter [AD]) and to 10–20% of open fire PM2.5 (particles nominally less than 2.5 μm AD), indicating that a reliable, statistically significant (p < 0.05), and meaningful
The objective of the third study, presented herein, was to explore the efficacy of CO as a proxy for PM_{2.5} to determine how personal mother and child CO exposure measurements relate to area PM_{2.5} and CO, and to determine how personal mother CO relates to personal child CO. On the basis of available knowledge of fuel combustion, air pollution, and health (5, 8), particulate exposures would arguably provide the best indicator of pollutant risks from fuel combustion for planned studies of childhood ARI. Unfortunately, the large-scale, long duration, and difficult logistics (e.g., sampling pumps and particulate sampling apparatus cannot be easily attached to infants for personal sampling) of such studies make problematic and expensive any widespread use of the usual particulate monitoring equipment. Thus, we designed studies to determine whether it might be possible to use simple Draeger CO passive diffusion tubes as a reliable surrogate for particulate exposures. These devices are considerably easier to use in these circumstances, although not having the accuracy of other methods (e.g., battery-operated Draeger CO electrochemical sensor with datalogger). Published evidence however shows that the Draeger CO passive diffusion tubes are reasonably reliable (17, 18), including for the elevated CO levels observed in this study environment, and that CO and particulate emissions are well-correlated in wood-smoke combustion environments (19).

**Experimental Section**

**Methods.** The studies were conducted in rural villages in the Quetzaltenango (also known as Xela) region of the western highlands of Guatemala (altitude range 2500–2800 m). The first part of this study (a test house study, hereafter referred to as Guat-2) was done in the fall of 1993, and the second (a cross-sectional study, hereafter referred to as Guat-3) was done in the summer of 1994, both during the rainy season which runs from May to November in this region. Most families in the study region burn wood on open fires, with around 10–20% using wood-burning stoves with chimneys such as the plancha and the lorena and a few using gas stoves. The Lorenas are taller and more massive than the planchas and typically have three burners in a circle atop a mud-based unit with no brick, steel, or tile components such as the plancha. Most of the Lorenas were constructed over a decade ago, are in poor condition (e.g., cracked stove body and top), and function more or less as an open fire stove as opposed to a functional wood stove with a chimney like the newer planchas. The houses are generally made of adobe and wood, sometimes with only one room that serves as kitchen, main living area, and bedroom. In the primarily Mayan population of this region, the women typically carry children under two years of age on their backs during much of the day, which is believed to result in high exposure for these children when the mother is cooking.

In Guat-2, kitchen, bedroom, and outdoor measurements were made in the same three homes using first no stove (background), and then sequentially the three cookstoves—open woodfire, bottled gas (LPG), and plancha. Two of the homes were chosen from the town of Concepcion Chiquirichapa, a village with a comparatively high housing density (i.e., semi-urban) for the region (approximately 17 houses/ha) (15). In this town, one home (H1) was selected with the kitchen and the sleeping quarters in the same room, and one (H2) was selected with the kitchen and the sleeping quarters in separate rooms. The third test house (L2) was selected from Buena Vista, a town with a comparatively low housing density (i.e., rural) for the region (approximately 0.2 houses/ha) (15) and had a separate kitchen and sleeping quarters.

The test homes in Guat-2 were selected under the following criteria: (i) no smokers lived in the home; (ii) a mother lived in the home; (iii) a child under the age of 24 months lived in the home; (iv) the homeowners used an open fire as their only means of cooking; (v) the home met the specified urban vs rural and one room or multiple room dwelling criteria; (vi) the homeowners were willing to cooperate with all the components of the study (e.g., area and personal air monitoring, temporary use of a gas stove, and installation and use of a plancha); and (vii) the home had electricity. We selected homes according to criteria (e.g., presence/absence of smokers, housing density, etc.) aimed at identifying homes representative of the region. Considering the home selection criteria used combined with the homogeneous nature of the homes in this region, as observed by Smith et al. (14) and Naheer et al. (15), we are confident that the three homes selected were sufficiently representative of the region to achieve the objectives of this study. The householders were trained by a representative from the Institute of Nutrition of Central America and Panama in the use of the improved stoves; the training required was minimal.

Since the overall purpose was to understand changes in total exposure, which could then be related to health effects, essentially all monitoring was done on a near-24-h basis minus the approximately 2 h required each day to change filters, download data, calibrate pumps, and otherwise maintain the equipment. Thus, the actual monitoring time for each sample day was roughly 22 h. The specific scheduling of the 2-h maintenance time varied daily depending on the technician’s schedule but was generally scheduled during noncooking times between meals.

Each cooking condition in each test home was monitored for three separate 22-h periods at two (H1; kitchen/bedroom and outside) or three (L2, H2; kitchen, bedroom, and outside) locations. At each location there was placed a sampling pack containing (i) an SKC Universal Flow Sample pump running at 2.0 L/min attached to a filter cassette for integrated total suspended particulates (TSP); (ii) a similar pump running at 4.0 L/min attached to a PAM impactor for integrated particles nominally less than 10 μm AD (PM_{10}); (iii) another pump running at 3.5 L/min attached to an MIE cyclone (respirable cyclone precollection and cyclone precollector (DR–RCP10), Dorr-Oliver 10-mm Nylon cyclone and fittings with a 3.5-μm particle cut point) and filter cassette for integrated particles nominally less than 2.5 μm AD (PM_{2.5}); (iv) an infrared-scattering Miniram with Langen datalogger for continuous PM_{2.5} inserted between the cyclone and filter of iii; (v) a battery-operated Draeger CO electrochemical sensor with datalogger for continuous CO; and (vi) a Draeger CO passive diffusion (color stain) tube for integrated CO. The continuous PM_{2.5} data are not presented herein. All particle collection was on 37-mm Teflon-coated glass fiber Pallflex filters. The air sampling packs were suspended from the ceiling at roughly 1 m from the nearest wall and 1.3 m off the ground and were attached to 110 V household current.

In homes L2 and H2, a pack was placed in the bedroom, kitchen, and outside of the house; in home H1, a pack was placed in the bedroom/kitchen (same room) and outside of the house. For all of the houses, the packs in the kitchen were placed 1.3 m off the ground and within 0.5 m of the stove, close to where the woman of the home would typically spend most of her cooking time, while the packs placed outside of the house were located roughly 10 m away from the room containing the cooking stove.

In addition to the area monitoring for PM_{2.5} and CO, a Draeger CO tube was worn by the mother and one child (<24 months age) in each test house. The personal Draeger CO passive diffusion tubes were attached to the clothing in...
the chest area of the mother and child. The area (i.e., kitchen, bedroom, and outside) and personal (i.e., mother and child) Draeger CO passive diffusion tube data and the area pump and filter PM2.5 and PM10 data are presented herein. The TSP and continuous CO data are presented elsewhere (16).

Guat-3 was designed to further explore the relationship between personal (mother and child) 24-h integrated CO and kitchen-area 24-h integrated PM2.5 measurements. The correlation between kitchen 24-h integrated CO and 24-h integrated PM2.5 was also investigated, as was the correlation between mother and child 24-h integrated CO. Area (i.e., kitchen) and personal (i.e., mother and child) 24-h integrated CO and area PM2.5 were collected using the same monitoring equipment and methodology (i.e., Draeger CO passive diffusion tubes, SKC pump, PM2.5 cyclone, and Teflon filter) described above for Guat-2. Due to the scheduling logistics of Guat-3, the 24-h measurements represent a true 24-h, as opposed to the 22-h measurements conducted in Guat-2.

Twenty-five households with open fires and an equal number with planchas were selected using the following criteria: (i) a plancha or an open fire was the sole stove type used in the home; (ii) a mother and a child less than 24 months of age lived in the home; and (iii) no cigarette smoking occurred in the home. However, data from only 15 of the open fire households were used in this analysis because of various pump and filter related problems experienced in the field.

Equipment Handling and Quality Control. Standard methods were used to determine air pollution concentrations from the measurements made and to maintain quality control. Pre- and post-weighting occurred in climate-controlled laboratory facilities at the Harvard School of Public Health. The filters were conditioned for 48 h for both weighings. Two lab filter blanks were collected at the time of the preweighing, and 12 field filter blanks were collected, one per week for the duration of the study; particulate mass values for the study samples were adjusted for lab and field filter blank values. Filters were placed in individually labeled filter sampling cassettes immediately following the preweighing and removed at the time of the post-weighing. Support pads were used. Since little to no variation was seen in the pump flow rate in the early stages of this study, all pumps were calibrated every 2–4 days with a bubble tube. The Draeger CO passive diffusion tubes were read onsite immediately following the sampling period or covered with an air-tight cap and read the same day upon return to the field base.

Results and Discussion

Here we report the correlations between stain-tube CO levels and measured PM2.5 levels for the test-house (Guat-2) study and the cross-sectional (Guat-3) study. Raw data from Guat-2 (including CO, PM2.5, PM10, and TSP) and Guat-3 (including CO and PM2.5) are provided in Tables 1 and 2, respectively.

The Spearman correlation coefficient (R) between CO and PM2.5 for all of the Guat-2 locations (i.e., kitchen, bedroom, outside areas), households (i.e., H1, H2, L2), and stoves (i.e., open fire, plancha, gas stove) reveals a reasonably strong correlation (R = 0.77; p < 0.001; n = 68). Kitchen measurements only (i.e., excluding those from the bedroom and outside areas) produce a stronger correlation (R = 0.89; p < 0.0001; n = 26) (Figure 1). More directly relevant to the objectives of the current study is the correlation between CO and PM2.5 in the kitchens of homes using open fires (R = 0.50; p = 0.17; n = 9) or planchas (R = 0.90; p = 0.003; n = 8). As one would expect, the correlation is strengthened if the open fire and plancha data are pooled (R = 0.92; p < 0.0001; n = 17) (Figure 2). If the most extreme data points (PM2.5 > 800 μg/m3) are removed from the analysis, the correlation remains largely similar (R = 0.90; p < 0.0001; n = 15), although the regression equation changes from [PM2.5 (μg/m3) = CO (ppm) × 84.7 + 9.2] to [PM2.5 (μg/m3) = CO (ppm) × 59.2 + 62.5].

While the correlation for CO versus PM2.5 at the area monitoring sites is strong, especially when there is a strong source of smoke (e.g., strong indoor emissions source like open fire or plancha), the correlation between mother and child personal CO and kitchen area PM2.5 is not. For example, open fire and plancha data from Guat-2 reveals a Spearman correlation coefficient (R) of 0.65 (p = 0.044; n = 10) between personal CO and kitchen area PM2.5, and 0.8 for the mother and 0.74 (p = 0.034; n = 10) for the child. The correlation between mother personal CO and child personal CO is convincingly strong. For example, open fire and plancha data from Guat-2 reveals strong correlations coefficients (R) of 0.95 (p = 0.052; n = 4) and 0.95 (p = 0.0013; n = 7), respectively, while R = 0.96 (p < 0.0001; n = 11) when the open fire and plancha data are pooled (Figure 3). If the most extreme data points (CO > 10 ppm) are removed from the analysis, the correlation remains largely similar (R = 0.95; p < 0.0001; n = 10), while the regression equation changes from [child CO (ppm) = mother CO (ppm) × 1.004 – 0.157] to [child CO (ppm) = mother CO (ppm) × 0.918 + 0.006].

For Guat-3, the kitchen CO versus kitchen PM2.5 correlations were stronger for plancha data (R = 0.89; p < 0.0001; n = 25) than for open fire data (R = 0.70; p = 0.004; n = 15) and strongest when both open fire and plancha data were pooled (R = 0.94; p < 0.0001; n = 40) (Figure 4). If the most extreme data points (PM2.5 > 800 μg/m3) are removed from the analysis, the correlation remains largely similar (R = 0.93; p < 0.0001; n = 36), while the regression equation changes from [PM2.5 (μg/m3) = CO (ppm) × 75.3 – 15.4] to [PM2.5 (μg/m3) = CO (ppm) × 65.1 + 42.8].
The correlation between kitchen area PM2.5 and personal CO observed in Guat-3 is similar to that found in Guat-2. When the open fire and plancha data are pooled, the correlation (R) between kitchen area PM2.5 and mother personal CO is 0.63 (p < 0.0001; n = 39) and the correlation with child personal CO is 0.56 (p < 0.0001; n = 41). When the open fire and plancha data are pooled, a strong correlation between mother personal CO and child personal CO exists (R = 0.85; p < 0.0001; n = 50) (Figure 5). If the most extreme data points (CO > 10 ppm) are removed from the analysis, the correlation remains largely similar (R = 0.84; p < 0.0001; n = 49), while the regression equation changes from [child CO (ppm) = mother CO (ppm) × 0.894 + 0.066] to [child CO (ppm) = mother CO (ppm) × 0.831 + 0.165]. The strong correlation between mother personal CO and child personal CO does not break down if the open fire (R = 0.85; p < 0.0001; n = 25) or plancha data (R = 0.68; p < 0.0002; n = 25) are looked at separately.

Discussion

The results of Guat-2 and Guat-3 show strong correlations between daily average area concentrations of PM2.5 and CO while clarifying some of the limitations of the ability for CO to serve as a proxy for PM2.5 exposures. For example, although the correlation between CO and PM2.5 was high and convincing in homes where open fires or some planchas TABLE 2. Kitchen CO and PM2.5 and Mother and Child Personal CO from Guat–3

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FIGURE 1. Guat-2: Carbon monoxide vs PM2.5 in all houses for all stove conditions; kitchen measurements only. R = 0.89; p = 0.0001; n = 26.

The correlation between kitchen area PM2.5 with personal CO observed in Guat-3 is similar to that found in Guat-2. When the open fire and plancha data are pooled, the correlation (R) between kitchen area PM2.5 and mother personal CO is 0.63 (p < 0.0001; n = 39) and the correlation (R) with child personal CO is 0.56 (p < 0.0001; n = 41). When the open fire and plancha data are pooled, a strong correlation between mother personal CO and child personal CO exists (R = 0.85; p < 0.0001; n = 50) (Figure 5). If the most extreme data points (CO > 10 ppm) are removed from the analysis, the correlation remains largely similar (R = 0.84; p < 0.0001; n = 49), while the regression equation changes from [child CO (ppm) = mother CO (ppm) × 0.894 + 0.066] to [child CO (ppm) = mother CO (ppm) × 0.831 + 0.165]. The strong correlation between mother personal CO and child personal CO does not break down if the open fire (R = 0.85; p < 0.0001; n = 25) or plancha data (R = 0.68; p < 0.0002; n = 25) are looked at separately.

Discussion

The results of Guat-2 and Guat-3 show strong correlations between daily average area concentrations of PM2.5 and CO while clarifying some of the limitations of the ability for CO to serve as a proxy for PM2.5 exposures. For example, although the correlation between CO and PM2.5 was high and convincing in homes where open fires or some planchas...
(those with high air pollution levels) were used, the correlation largely broke down in homes where either no stove was used or gas stoves were used (i.e., where concentrations were low as compared to open fire conditions). There are several possibilities why some planchas pollute more than others (wood type or moisture content, cooking practices, etc.). One primary possibility, supported by our data and other studies in this series (14, 15), is that some planchas function less efficiently than others, often due to age-induced damage like cracking in the metal top pot holes or in the brick body of the unit.

Unlike the strong correlations observed between PM$_{2.5}$ and CO using time-integrated area measurements, the correlations between personal (mother and child) CO and area PM$_{2.5}$ were only moderately correlated. This can be partly explained by the fact that mothers and children spend only part of the day in the kitchen and, more importantly, that the part of the day that people spend in the kitchen may vary in duration and time relative to high pollution periods; thus, the personal and area measurements are not fully comparable. Another component of the explanation, however, relates to the temporal pattern of the wood burn cycle. It has been shown in the literature that the production ratios of particulates and CO are highly correlated over the entire wood burn cycle, but that there are significant variations at different times during the burn (20, 21). Thus, if people routinely do not stay near the fire for the whole burn cycle, CO may not be as good an indicator of actual particulate exposure as it seems from the 22–24-h correlations, which include only complete burn cycles.

These findings are consistent with those of Ezzati et al. (21), who looked closely at the potential of using CO as a proxy for suspended particulate concentrations. They concluded that, given the recent commercial availability of small-size monitoring devices for suspended particulate matter (PM) and the critical importance of the episodic nature of exposure to indoor smoke, the usefulness of CO as a means for detailed assessment of exposure to suspended particulate matter is limited. A few points made in the Ezzati et al. (21) paper warrant comment here. First, Ezzati et al. (21) note the importance of considering type of fuel and stove when using CO as a proxy for PM. We too find that fuel is important, but not so much the stove, and find that the same CO:PM relationship cannot be extended from wood to gas stove emissions. It is also true that 24-h CO means do not necessarily indicate contaminant peak levels, which may possibly be a factor in ill health. Nevertheless, measurements of mean levels have historically proved remarkably effective as indicators for pinning down and protecting against health effects of air pollution across a range of pollutants, popula-
tions, and health endpoints. In addition, it is probable that most interventions to control means will also control peaks, although perhaps not to the same degree; this is clearly an important area for future research. Finally, although devices are available for highly accurate real-time measurement of CO and PM, they are currently much too expensive (in capital and labor costs) for use in many of the developing country situations where problems are worst; accuracy sought should depend on the application being considered and the resources available. Indeed, our work and that of Ezzati et al. point to the need to develop inexpensive devices of this sort that may not be as accurate as those developed for industrial country applications but can be applied practically in the relatively high-concentration conditions common in developing country households.

Relevant to future biomass-fueled cooking stove and ARI epidemiology studies, results from the current study suggest three issues of importance. First, although CO is a reasonable strong proxy for PM$_{2.5}$ exposure in homes using open fires or planchas, its use as a proxy is not valid under gas stove use or similarly clean burning conditions. This suggests that CO may serve as a proxy for PM$_{2.5}$ only under certain conditions but that alternative PM$_{2.5}$ measures must be used under cleaner burning conditions. Second, the mother personal CO is strongly correlated with child’s (under 2 years of age) personal CO, suggesting that mother personal CO can be used as a proxy for child’s CO. Third, since area CO measurements are not strongly representative of personal CO measurements, emphasis should be placed on collecting personal CO measurements instead of area measurements. Overall, these results generally support the use of Draeger CO passive diffusion tubes as a proxy for PM$_{2.5}$ in such cases where a single type of emission source is the predominant source for CO and PM$_{2.5}$.

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