



Article

# **Exposures to Carbon Monoxide in a Cookstove Intervention in Northern Ghana**

Ricardo Piedrahita <sup>1</sup>, Evan R. Coffey <sup>1,\*</sup>, Yolanda Hagar <sup>2</sup>, Ernest Kanyomse <sup>3</sup>, Christine Wiedinmyer <sup>4,5</sup>, Katherine L. Dickinson <sup>6</sup>, Abraham Oduro <sup>3</sup> and Michael P. Hannigan <sup>1</sup>

- Department of Mechanical Engineering, University of Colorado Boulder, 427 UCB, 1111 Engineering Drive, Boulder, CO 80309, USA
- Department of Applied Mathematics, University of Colorado Boulder, 526 UCB, Boulder, CO 80309-0526, USA
- Navrongo Health Research Center, Post Office Box 114, Upper East Region, Navrongo 03821, Ghana
- Cooperative Institute for Research in Environmental Sciences, Campus Box 216, University of Colorado Boulder, Boulder, CO 80309, USA
- Department of Environmental and Occupational Health, Colorado School of Public Health, 13001 E, 17th Place, Box B119, Aurora, CO 80045, USA
- National Center for Atmospheric Science, P.O. Box 3000, Boulder, CO 80307-3000, USA
- \* Correspondence: evan.coffey@colorado.edu; Tel.: +1-303-735-5045

Received: 30 May 2019; Accepted: 3 July 2019; Published: 16 July 2019



**Abstract:** Biomass burning for home energy use is a major environmental health concern. Improved cooking technologies could generate environmental health benefits, yet prior results regarding reduced personal exposure to air pollution are mixed. In this study, two improved stove types were distributed over four study groups in Northern Ghana. Participants wore real-time carbon monoxide (CO) monitors to measure the effect of the intervention on personal exposures. Relative to the control group (those using traditional stoves), there was a 30.3% reduction in CO exposures in the group given two Philips forced draft stoves (p = 0.08), 10.5% reduction in the group given two Gyapa stoves (locally made rocket stoves) (p = 0.62), and 10.2% reduction in the group given one of each (p = 0.61). Overall, CO exposure for participants was low given the prevalence of cooking over traditional three-stone fires, with 8.2% of daily samples exceeding WHO Tier-1 standards. We present quantification methods and performance of duplicate monitors. We analyzed the relationship between personal carbonaceous particulate matter less than 2.5 microns (PM<sub>2.5</sub>) and CO exposure for the dataset that included both measurements, finding a weak relationship likely due to the diversity of identified air pollution sources in the region and behavior variability.

Keywords: exposure; carbon monoxide; cooking; mixed effects models; micro-environment

# 1. Introduction

Carbon monoxide (CO) is commonly measured in cookstove and air pollution exposure studies due to its association with adverse health effects and birth outcomes such as low birth weight [1–3], the relatively low cost of real-time wearable monitors, logistical challenges of measuring other pollutants, and a (hypothesized) correlation with other co-emitted pollutants. These reasons, along with the well-established health effects associated with particulate matter less than 2.5 microns (PM<sub>2.5</sub>) [4], have led to the study and use of CO as a surrogate for PM<sub>2.5</sub>. Previous studies have found varied results for this relationship, with differences ascribed to variability in source types like fuels and stoves, behaviors, and home designs, among other characteristics [5–10].

Building on prior work, this paper seeks to characterize CO exposures, their determinants across households and across time, and their association with PM<sub>2.5</sub> exposures in the context of a

Atmosphere **2019**, 10, 402 2 of 13

cookstove intervention study in Northern Ghana. The REACCTING study (research of emissions, air quality, climate, and cooking technologies in Northern Ghana) was a 200 household cookstove intervention in the Kassena-Nankana districts of Ghana's Upper East region from November 2013 to December 2015 [11]. The 200 study households were divided into four groups, with one group given two locally made Gyapa rocket stoves, one group given two Philips HD4012 LS forced draft stoves, one group given one of each intervention stove, and the last group serving as control until the end of the study, when they were given their choice of two stoves. Details of the study design [11] and region [12] are described elsewhere. Piedrahita and colleagues [13] present personal, cooking area, and ambient carbonaceous PM<sub>2.5</sub> results from the study, while Piedrahita et al. (2016) and Dickinson et al. (2019) present stove usage results, and Piedrahita et al. (2019) reports on the use of time-activity monitors to further explain exposure as it relates to human behavior during and beyond cooking [14–16]. Findings from these publications support the idea that sources beyond residential cooking contribute to personal exposure and that cookstove-targeted efforts to reduce exposure represent a significant, yet limited, approach. For example, background CO concentrations in urban areas tend to be higher than rural ones [16], creating an elevated baseline concentration during urban cooking events that augment the direct impacts of the stove being used. Here, we present personal CO exposure results over the complete study period, focusing on the effects of stove group and other covariates on exposure. Further, we identify and characterize behavioral factors affecting exposures gleaned through the daily trends observed in CO concentrations in kitchen areas and exposure profiles. Finally, we report on correlations between personal CO and PM<sub>2.5</sub> exposures and reflect on reasons for their apparent divergence in this context.

## 2. Methods

To asses REACCTING intervention impacts on personal CO exposure, an air quality measurement campaign was performed. Personal CO exposure sampling was performed throughout the REACCTING study, from November 2013 through November 2015, with a total of 786 days of data collected (derived from 48 h samples) and passed through quality checks (Supplementary Information Section 2). A summary of the demographics of the people sampled for CO exposure is presented in Table 1. Lascar USB-CO300 and CO1000 monitors were used to measure 1 min CO concentrations and were typically worn on the mother and child of the study household, as well as other available and consenting family members. Sampling was usually carried out from Monday to Wednesday, and then Wednesday to Friday, at four households during each week, one from each arm of the intervention study. Adult study participants typically wore the monitors either on neck lanyards, or in a waist pack when additional monitoring equipment was included, such as PM<sub>2.5</sub> filter samplers. Children carried the Lascar monitors in the chest pockets of custom locally fabricated cotton t-shirts, or in backpacks when carrying other instruments (SI, Figure S1). Field enumerators asked the participants to wear the Lascar monitors as much as possible and instructed them to leave them within arms-reach if they could not wear them or while they were sleeping. A subset of samples included duplicate Lascar monitors placed side-by-side.

Atmosphere **2019**, 10, 402 3 of 13

		Control	Gyapa/Philips	Philips/Philips	Gyapa/Gyapa
Sample overview	Total days deployed *	256	239	278	210
	Non-flagged days deployed (retained for analysis)	207 (81% of total days deployed)	201 (84%)	212 (76%)	166 (79%)
	Duration hours (mean (SD))	23.1 (1.3)	23.1 (1.2)	23.1 (1.4)	23.3 (1.1)
	Days duplicate monitor were deployed	42	24	32	43
	Unique participants	62	73	68	64
Gender covariates	Primary cook Females (person-days)	113	97	105	111
	Non-primary cook females (person-days)	51	46	60	34
	Non-primary cook males (person-days)	43	58	47	21
	Female age in years (med, SD, min, max)	27.4, 18.7, 1.2, 75.4	33.4, 15.9, 2.4, 63.4	24.5, 16, 1.2, 53.4	30.4, 16.8, 2.1, 63.4
	Male age in years (med, SD, min, max)	4.2, 4.2, 1.3, 13.8	4.5, 17.1, 1.4, 61.4	6.4, 2.8, 1.1, 11.6	3.9, 2.8, 1.1, 10
SES ** (person-days)	Least poor	22	39	40	39
	Less poor	37	46	38	33
	Poor	63	41	65	45
	Poorer	22	45	28	40
	Poorest	63	30	41	9
Seasons *** (person-days)	Harmattan/bush burning	85	91	94	53
	Transition	20	20	16	18
	Light Rainy	14	26	30	32
	Heavy Rainy	60	34	45	32
	Hot dry	28	30	27	31

**Table 1.** Sample statistics for the personal CO exposure deployments throughout research of emissions, air quality, climate, and cooking technologies in Northern Ghana (REACCTING).

Bluetooth Beacons were deployed during a subset of samples to estimate distance between participants and cookstoves [17]. The variability in the distance measurement was used to estimate compliance; variability indicative of no motion of the participant suggests the devices were not being worn. Here, compliance was defined by thresholds determined by [17].

# 2.1. Calibration and Data Preparation Methods

CO monitor calibrations were performed as often as possible over the 2-year study period, averaging 1.5 calibrations per monitor (range 1–7, SD = 1.5) and Pearson's r of 0.99  $\pm$  0.06 from linear calibration functions (SI, Figure S3). In most cases (101/110), monitors were calibrated with NIST traceable CO gas concentration standards. Further, on a weekly basis, the monitors were co-located with the reference monitor (Thermo 48C, Thermo Scientific) located at the Navrongo Health Research Centre (NHRC) for field normalization (9/110 calibrations). In some instances, the ambient concentrations were too low to generate a satisfactory calibration curve in the concentration range desired for the personal exposure measurements; those normalizations were not applied.

Twenty percent (197) of the sampling days were removed due to CO sensor degradation and electronics concerns (flagged and removed sampling days are the difference between 'total days deployed' and 'non-flagged days deployed' in Table 1). Paired CO Lascar monitors were successfully deployed on 141 days, with duplicates showing a Pearson's r of 0.78 for the calibrated and 0.92 for the un-calibrated daily average concentrations (SI, Section 4). Lower correlation for the calibrated monitors can be attributed to the general observed trend of decreased sensitivity with Lascar age (SI, Figure S3). When some of the monitors were calibrated and others were not, the improved data quality of the calibrated monitors resulted in poorer correlation (SI, Figure S4). As the monitors were randomly distributed among stove groups and individuals, there is minimal risk of systematic bias, and the calibrated Lascars should provide more accurate results despite the lower correlation. Calibrations are also important to avoid bias related to the factory calibration of the monitors and to adjust for the

<sup>\*</sup> flagged days represent days where issues occurred and have been omitted from analysis. \*\* socioeconomic status as by Awini and colleagues. \*\*\* seasons: 'light rainy' (April–June), 'heavy rainy' (June–October), 'transition' (only October), 'Harmattan' (November to mid-February), and 'hot dry' (mid-February to April).

Atmosphere **2019**, 10, 402 4 of 13

Lascar change in sensitivity over time. When the monitors were first calibrated, the following linear regression form was used.

LascarSignal = 
$$p_1 + p_2$$
(ReferenceConcentration) +  $\varepsilon$ .

The average slope ( $p_2$ , sensitivity) was  $1.06 \pm 0.06$  and intercept ( $p_1$ ) was  $0.08 \pm 0.13$  ppm. When paired monitors were deployed and both were deemed free of failures, minute average concentrations from the two monitors were used in further analyses.

# 2.2. Personal Exposure Mixed Effects Model Specification

Mixed effects modeling was performed to ascertain the intervention effects while accounting for repeated measures [18,19], in an approach very similar to other work [13]. The Akaike information criterion (AIC) and adjusted  $R^2$  were used to select a parsimonious model and assess the fit after the most relevant variables were included (stove group, season, socioeconomic status (SES)). Using these model selection methods, the number of family members and age group variables were not found to have significant effects on predicted 24 h average CO exposures and did not improve the fit, so were excluded from the final model we present. The final model form regressed daily (24 h) average log-transformed CO exposure (ppm) against stove group, season, SES, and primary cook-by-gender (Model 1, see Equation (1)).

$$Log(Personal\ CO_{ij}) = \beta_0 + \beta_1(Stove\ group)_i + \beta_2(SES)_i + \beta_3(Primary\ cook^*Gender)_i + \beta_4(Season)_i + \alpha_i + e_{ij} \quad (1)$$

The equation above is slightly simplified;  $\beta_0$  is the model intercept and represents the overall expected log of CO exposure in ppm.  $\beta_1$  represents the 4 stove group categories, 'Control', 'Gyapa/Philips', 'Philips/Philips', and 'Gyapa/Gyapa' with 'Control' as the reference group.  $\beta_2$  represents five categories for socioeconomic status, 'poorest', 'very poor', 'poor', 'less poor', and 'least poor' [20], calculated for each household using data from a district-wide health and demographic surveillance survey [12].  $\beta_3$  represents three categories, 'female primary cook', 'female non-primary cook', and 'male non-primary cook', where primary household cook was identified in the baseline survey [11], and there were no male primary cooks.  $\beta_4$  represents the season categories, defined as 'light rainy' (April to June), 'heavy rainy' (June to October), 'transition' (only October), 'Harmattan' (windy with bush-burning, November to mid-February), and 'hot dry' (mid-February to April). The random effect  $\alpha_i$  accounts for the correlation within subjects due to repeated measures (i.e., daily exposure measurements made more than once for a given individual), and  $e_{ij}$  represents the variation from subject to subject. A small positive exposure concentration was added to all values to include exposures of zero ppm in the log-transformed model. A compound symmetry covariance structure was employed, using the assumption that any repeated measurements of participants had equal correlation regardless of the time between them [19]. Residual distributions approached normality, satisfying the assumptions required to employ this modeling technique. We applied Equation (1) separately to the calibrated and un-calibrated Lascar CO datasets to compare results and understand the importance of our calibrations over the study.

#### 3. Results

## 3.1. Personal CO Exposure Results

The results from Model 1 (Equation (1)) are presented here in terms of percent change relative to the reference categories built into the model, as well as expected concentration values (geometric means) at those categories (Table 2). The reference for Model 1 were those measurements taken from primary cooks (all female) in the control stove group also in the 'poorest' SES category during the Harmattan season. The reference group experienced a daily (24 h average) CO exposure concentration expected value of 0.55 ppm (95% CI 0.34–0.90). By holding all variables constant except for stove

Atmosphere **2019**, 10, 402 5 of 13

group, the effect of stove group is isolated relative to the control group. On average, the CO exposure of the Gyapa/Gyapa group was 10.5% lower than control group (0.49 ppm (0.32–0.77), p=0.62), the Philips/Philips group was 30.3% lower (0.39 ppm (0.26–0.58), p=0.08), and the Philips/Gyapa group was 10.2% lower (0.51 ppm (0.26–0.58), p=0.61). Males and females listed as non-primary cooks experienced 36.7% (p=0.02) and 33.2% (p=0.03) lower average CO exposures than female primary cooks, respectively. The 'light rainy' and 'heavy rainy' seasons had 59.0% and 15.5% higher expected exposures than the 'Harmattan' (p=0.02, p=0.38), while the 'hot dry' season was 30.1% lower (p=0.06) than the 'Harmattan' season. Relative to the 'poorest' SES group, on average, the 'poorer' group had 3.5% higher exposure levels, and the 'poor', 'less poor', and 'least poor' groups experienced 7.5%, 29.1%, and 38.6% lower exposure levels respectively ( $p \ge 0.05$ ). The between-subject variation was 0.80 (p < 0.01), and residual error variation was 1.44, giving an intra-class correlation coefficient (ICC) of 0.36. Summary statistics grouped by Model 1 covariates are presented in SI Table S1.

Using the mixed effects model described in Equation (1) on the un-calibrated exposure data demonstrated that the results were stable and consistent with the results presented in Table 2, though some covariates did shift out of significance (SI Section 5).

The average measured rate of compliance for the data subset that had Bluetooth Beacon measurements was 77.8% and can be expected to be a lower estimate of all the samples because the Beacons were deployed as part of a larger sampling pack, whereas most deployments included the less-intrusive Lascar USB-CO monitors only.

	Equation 1	% Change from Reference	Expected Exposure (ppm) (95% CI)	P Value
sdn	Control	Reference group	0.55 (0.34, 0.90)	
gro	Gyapa/Philips	-10.2	0.5 (0.33, 0.75)	0.61
Stove groups	Philips/Philips	-30.3	0.39 (0.26, 0.58)	0.08
St	Gyapa/Gyapa	-10.5	0.49 (0.32, 0.77)	0.62
er	Primary cook females	Reference group		
Gender	Non-primary cook females	-33.2	0.37 (0.26, 0.53)	0.03
	Non-primary cook males	-36.7	0.35 (0.24, 0.51)	0.02
	Least poor	-38.6	0.34 (0.21, 0.55)	0.05
	Less poor	-29.1	0.39 (0.24, 0.64)	0.17
SES	Poor	-7.5	0.51 (0.32, 0.82)	0.74
0,	Poorer	3.5	0.57 (0.37, 0.89)	0.88
	Poorest	Reference group		
	Harmattan	Reference group		
S	Transition	-14.5	0.47 (0.31, 0.73)	0.48
Seasons	Light rainy	59.0	0.88 (0.6, 1.28)	0.02
Se	Heavy rainy	15.5	0.64 (0.46, 0.88)	0.38
	Hot dry	-30.1	0.39 (0.27, 0.56)	0.06

Table 2. Personal CO 24 h average exposure Model 1 results and summary statistics.

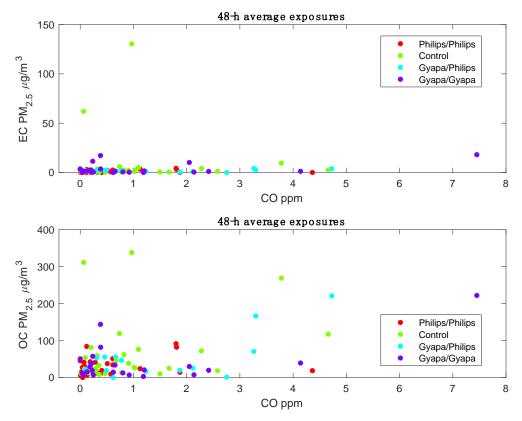
# 3.2. Relationship between 48 h Averaged CO and 48 h Cumulative Carbonaceous PM<sub>2.5</sub> Exposure

A subset of measured CO personal exposure measurements were made alongside personal exposure observations of carbonaceous (elemental carbon (EC) and organic carbon (OC))  $PM_{2.5}$  using pump and filters reported in detail elsewhere [13]. Linear regression modeling was employed to assess the relationship between the log of personal 48 h average EC and OC  $PM_{2.5}$  concentration and the log of 48 h average CO concentration with 108 personal exposure sample periods available for the comparison. Personal  $PM_{2.5}$  EC and OC were analyzed using the NIOSH 5040 method [21], as described by Piedrahita et al. [13]. A linear regression was employed to assess this relationship (Model 2, shown in Equation (2)).

$$Log(Personal PM2.5) = \beta_0 + \beta_1 * (log(CO)) + e$$
 (2)

Atmosphere **2019**, 10, 402 6 of 13

The correlation coefficient (R) of Model 2 (Equation (2)) was 0.07 for EC and 0.19 for OC (see Figure 1). (Supplementary Information, SI Section 7). The weak relationships are likely due to the variability in personal behaviors and the diversity of air pollution sources in the region [13,22]. The stronger relationship with OC may be due to the higher co-emission of CO than with EC in a typical biomass fire [23–25]. During the smoldering phase of biomass combustion, CO is more abundantly co-emitted with OC than with EC due to oxygen-deficient, lower temperature combustion where volatilized organics condense to form larger particles [26]. Additionally, the later smoldering phase of biomass combustion may be a higher source of CO exposure than other burning phases, due to the relative duration of the phase and behavioral factors (e.g., participants using smoldering fires to (pre) heat water for other tasks).



**Figure 1.** PM<sub>2.5</sub> elemental carbon (EC) and organic carbon (OC) concentration vs. CO personal exposure concentration (48 h averaging) from the dataset with concurrent measurements. Note the lack of correlation between pollutants (R = 0.07 for EC and R = 0.19 for OC), even when accounting for stove group.

#### 4. Discussion

# 4.1. Contributing Factors to Personal CO Exposure

Results from the mixed effects model (Equation (1)) using 24 h-averaged CO exposures showed reductions in CO exposure for all three intervention groups (though not statistically significant at p < 0.05), suggesting a reduction in air pollution exposure to CO due to the intervention. This result is supported by previous analysis from this study showing that carbonaceous PM<sub>2.5</sub> was lower for all intervention groups, though the reductions were less substantial for CO [13]. In that PM<sub>2.5</sub> analysis, OC exposure was found to be significantly lower in the three intervention groups by factors of 49.4% for the Philips/Philips group, 57.3% for the Gyapa/Gyapa group, and 63.2% for the Gyapa/Philips group [13]. This reduction in exposure to OC was observed despite high continued use of traditional three-stone fires (TSFs) in all of the intervention groups, especially the group given two Philips stoves,

Atmosphere **2019**, 10, 402 7 of 13

which here had the biggest decrease in CO exposure. The Gyapa/Gyapa and the Gyapa/Philips groups had the greatest replacement of TSFs, with TSF use was reduced by about 50% of days cooked as measured with stove use monitors (SUMs) and household surveys [14,15]. The result of larger average CO exposure reductions in the groups given the Philips stoves is further explained by results from stove emission testing that showed the Philips stove, operated by participants in the field, emitting roughly half as much CO on a dry fuel basis as a TSF, whereas the Gyapa stove CO emissions were no different [27].

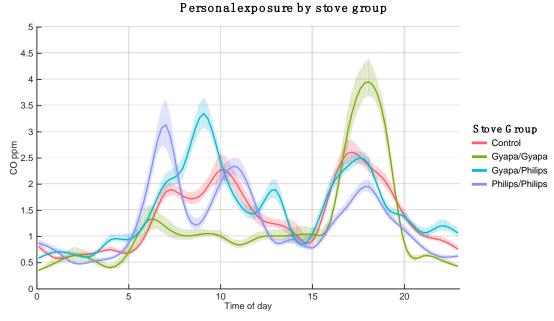
Although the PM<sub>2.5</sub> EC and OC results presented elsewhere [13] were from only the first year of the study, similar trends in seasonal effects were observed for the two-year CO data set. Compared to the 'Harmattan' season, CO exposures during the 'heavy rainy' and 'light rainy' seasons were higher by 18.3% (p = 0.30) and 59.0% (p = 0.02), while CO exposures during the 'transition' and 'hot dry' seasons were 33.2% (p = 0.03) and 14.0% (p = 0.49) lower, respectively. Lower carbonaceous PM<sub>2.5</sub> exposures were observed in all seasons relative to the 'Harmattan' season, but the biggest decreases in OC were during the 'light' and 'heavy' rainy seasons [13], in contrast to the results in the CO exposures. In-field stove emission testing during REACCTING found increased CO emissions from wood fuel at higher moisture contents, though not statistically significant at 0.05 [27]. Others have linked increased CO emissions with higher fuel moisture [28]; however, this trend does not appear to be shared with carbonaceous PM<sub>2.5</sub> at the fuel moisture levels observed here [29]. Dionisio and colleagues [7] also found increased CO exposure during the rainy season in The Gambia, when measuring exposure for children under age 5. The decrease in CO exposures during the 'transition' and 'hot dry' seasons observed in this study may be due to changes in fuel choices, behaviors during those seasons like cooking indoors or under a roof less frequently, as well as the relative importance of non-cooking pollution sources, such as regional biomass burning that occurs during these time periods [13].

The observed reductions in CO exposures may also have been dampened due to measurement uncertainty (0.14–0.15 ppm across stove groups) further discussed in Section 2 of the SI. As such, differences observed between stove groups may have been blunted. The root mean squared error (RMSE) of daily-average duplicate Lascar CO monitors was 1.16 ppm, higher than the expected exposure of any stove group (0.39–0.55 ppm). However, the large quantity of samples collected improves the statistical power of the analysis and allows the identification of CO exposure differences among groups and across explanatory variables such as season and demographics. Confidence in the results would be bolstered, and the relatively large uncertainty of single, 24 h average exposure estimates reduced by more frequent calibration and more precise instrumentation, which is recommended for future studies.

## 4.2. Observations from the Real-Time CO Exposure Time Series

Temporal patterns in the minute-resolution CO data can illuminate behavioral patterns, and changes therein, due to the intervention. Figure 2 shows CO exposures by time of day for each stove group smoothed using B-splines, which we use because of their efficient computation and calculation of confidence intervals [30,31]. All groups exhibited high exposure modes in the evening, peaking around 17:00–18:00 local time, while the Philips/Philips, Gyapa/Philips, and control groups had relatively large peak exposures at different times throughout the morning. The Gyapa/Gyapa group had the lowest morning exposure, but also the highest evening exposure, while both groups given a Philips stove had higher morning time peaks than evening peaks. Participants reported the Gyapa stove to be better at preparing TZ, the most popular dish in the region, than the Philips. TZ is a thick, starchy staple made from millet and is traditionally served with soup. This dish requires the cook to vigorously stir the contents of the pot, placing them directly over the stove for a significant period of time, exposing the cook to potentially high levels of CO and PM<sub>2.5</sub>. This specific cooking behavior may be more pronounced in the Gyapa/Gyapa group, explaining the large peak in evening CO exposure compared to cooks using the familiar TSF or preparing dishes more suitable to other stoves. These behavioral trends are difficult, if not impossible, to capture using surveys alone, which has motivated groups to monitor participant proximity to stoves using sensors [17]. The control group had nearly

equal CO exposure peaks in the morning and evening. The differences in morning exposures by stove group likely stem from changes in cooking behaviors, fuel type, or food preparation due to the stove intervention. The Philips stove was commonly fueled with woody biomass and/or charcoal whereas the Gyapa and TSF were exclusively fueled with woody biomass. The choice/availability of fuel at the time a meal was prepared can have impacts on emissions and resulting exposures. CO emissions from wood-fueled Philips stoves were on average half that of TSF, but charcoal-fueled Philips stoves emitted nearly 25% more CO than a TSF, per kilogram of fuel burned. The diurnal pattern is generally consistent with expected cooking patterns, although analysis of the stove use monitors (SUMs) showed the highest use for most stoves in the afternoons [14].



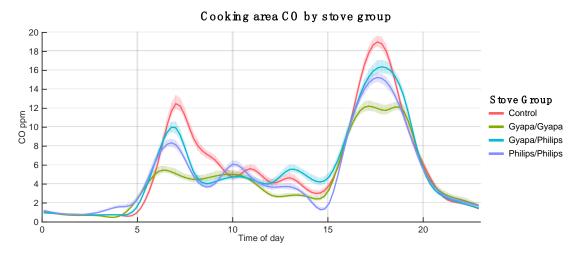
**Figure 2.** Spline-smoothed personal CO concentration by hour of day (0 = midnight) and stove group. The shaded bands represent 95% confidence intervals on the mean estimates.

Aggregated, smoothed time series were also plotted specifically for the primary cooks and for the study participants by gender. (SI Figures S7 and S8). Primary cook females had high morning and evening peaks, with the morning peaks coming around two hours earlier than male participants. This suggests differences in exposure sources, with those male exposures possibly incurred when away from home, and at work. Males under 5 had very similar exposure trends to the males over 5, and females under 5 had somewhat similar exposure trends to females over 5. These results suggest that gender roles are contributing to proximity to combustion sources and subsequent CO exposures as has been widely reported in other locations.

To further investigate personal exposure by stove group, we examined the time series of the CO concentrations measured in the cooking areas, which were available on a subset of the exposure monitoring periods (Figure 3). The cooking area CO concentration time series by stove group consistently showed morning and evening peaks, for all the stove groups, and largely matched the personal exposure trends for the earliest and latest peaks. The personal exposure trends showed more pronounced increases later in the morning, when the cooking area CO was not elevated, pointing to non-cooking CO exposure sources. Cooking area morning and afternoon CO peaks were also substantially higher than personal CO for all groups, especially for the control group, possibly indicative of changes in cooks' behaviors due to available stove and fuel types as discussed. Seasonal differences in the diurnal trends in cooking area CO concentrations were highly variable (SI Figure S6), possibly due to lower data density for some of the study groups. This, along with stove use, warrant further

Atmosphere **2019**, 10, 402 9 of 13

exploration as they also hold information about fuel and behavior change with season and location within the study region [17].



**Figure 3.** Spline-smoothed cooking area CO concentration by hour of day (0 = midnight) and stove group.

## 4.3. Comparisons with Previous Personal CO Exposure Results

Exposures in this study were generally low in comparison to past cookstove studies, given the extensive use of biomass fuels in the study households. In total, 8.4% of sample days exceeded the WHO guideline over the daily, 1 h maximum averaging period (35 mg/m<sup>3</sup>, 30.6 ppm), and less for the other guideline averaging periods [32] (SI Figure S2). Twenty-four-hour mean CO exposure for the control, Gyapa/Gyapa, Philips/Philips, and Gyapa/Philips groups were 1.05, 1.15, 0.90, and 0.94 ppm, respectively (SI Table S1). Key characteristics influencing exposure here include the prevalence of outdoor cooking, where pollutants can disperse at a faster rate, and low dwelling, vehicular, and industrial activity density. No primary cook reported smoking tobacco when asked in a survey. However, 30% of households reported having at least one tobacco smoker. Observationally, individuals smoking tobacco were elderly residents often smoking outdoors in common areas and are not expected to affect mean CO exposure differences among groups. In comparison, a study in Gambia that monitored the CO exposures of children under age 5 found mean 48 h exposures of 1.04 ppm (SD = 1.46 ppm) [7]. In rural Upper West Ghana, a study measuring the effects of a brick and mortar chimney stove intervention found that there was no significant difference in CO exposure compared to the control group, as measured during uncontrolled cooking tests [33]. Ochieng and colleagues [34] found median personal CO concentrations of primary cooks of 6.5 ppm in rural Kenyan homes using TSFs, and 4.4 ppm in homes using improved mud stoves. In Guatemala, in a colder climate with more indoor cooking, adult women cooks in the control group of the RESPIRE study experienced average 48 h exposures of 4.8 ppm (SD = 3.6), while the group with an improved plancha stove had an average exposure of 2.2 ppm (SD = 2.6). Also in the RESPIRE study, children under 18 months in the control group had average exposures of 2.8 ppm (SD = 2.5), while the intervention group averaged 1.5 ppm (SD = 1.9) [35]. Gautam and colleagues [36] measured daily CO exposure of primary cooks in Haryana, India using the Lascar-USB-CO finding 78% higher average CO exposures in households cooking with biomass  $(5.37 \pm 0.60 \text{ mg/m}^3)$  than households only using liquid propane gas  $(0.68 \pm 0.32 \text{ mg/m}^3)$ . Exposures measured in households that were categorized as 'no[t] cooking' were even lower  $(0.26 \pm 0.32 \text{ mg/m}^3)$ . Also, location of the kitchen (closed vs. open) was found to be a key determinant of exposure.

## 4.4. Relationship between Personal CO and Carbonaceous PM<sub>2.5</sub>

Strong relationships between CO and either PM<sub>2.5</sub> EC and PM<sub>2.5</sub> OC were not expected in this study region, due to the observed variability in cooking areas (often moved between indoors and outdoors), and the numerous additional contributing pollution source types observed both by observation and by source apportionment [13]. This is consistent with other studies. Roden and coworkers [37] directly measured emission factors from biomass cookstoves in laboratory and field settings, and found correlation coefficients between CO and PM<sub>2.5</sub> of 0.79 in-lab and 0.30 in-field. Other researchers [6] measured CO and PM<sub>2.5</sub> (N = 29) on children under age 5 in The Gambia, and found a correlation coefficient of -0.04. In Kenya, Ezzati et al. [38] found large variations in the strength of kitchen CO-PM<sub>10</sub> relationships depending on fuel and stove types. They further noted that the real-time correlation between CO and PM is weak and is thus a concern for the valid use of CO as a PM surrogate. McCracken and fellow researchers [8] observed a strong relationship between CO and PM<sub>2.5</sub> likely due to the relative contribution of cooking on personal exposure in the Guatemalan study region, consistency in fuels used, and the use of an indoor cooking area with a relatively tight ventilation envelope, that serves as the nucleus of the home. Carter et al. [5] reviewed studies comparing measured CO and PM<sub>2.5</sub> from indoor concentrations and personal exposures and found correlation coefficients (R) for personal exposure ranging from 0.22 to 0.97 (median = 0.53), and from 0.10 to 0.96 (median = 0.71) for indoor cooking area concentrations. That work discussed the sources of variability contributing to the quality of the relationship and noted that rigorous validation of the relationship should be performed if its widespread use is intended in a study. Our study confirms this inconsistency in the relationship between observed CO and PM<sub>2.5</sub>, suggesting that CO is not a good surrogate for PM<sub>2.5</sub> exposure in our study region.

#### 5. Conclusions

This work assessed the impacts of an improved cookstove intervention on personal CO exposure in a rural area of Northern Ghana over a two-year period. A combination of improved stoves was disseminated across three groups of the study and one group acted as a control receiving their preference of two stoves at the conclusion of the study. Lascar CO monitors were calibrated and deployed to measure personal CO exposure at 1 min temporal resolution. Mixed effects models were employed to better understand the drivers of CO exposure. Moreover, the relationship between CO and carbonaceous PM exposure was investigated. A decrease in CO exposure in the three intervention groups was identified, with the largest decrease (33%) seen in the Philips/Philips group (p = 0.08), the group receiving two of the cleaner-burning stove. Average daily CO exposure levels for all groups were quite low, averaging under 1.15 ppm for all arms of the intervention. Elevated 15 minute and one-hour average CO exposures indicative of cooking events were clearly observed in many exposure time series, especially for women that are primary cooks, but they were usually lower than the WHO Tier-1 guidelines of 35 and 100 mg/m<sup>3</sup>, respectively [32]. Seasonal impacts on daily CO exposure were likely related to fuel type availability/preference, weather conditions, and subsequent cooking behavior changes. Future work investigating links between cookstove use and exposure would benefit from time-resolved PM<sub>2.5</sub> measurements and more robust stove use information offering insights into activity-based exposure. A better understanding of these effects, along with the observed trend of decreasing CO with increasing SES, would provide valuable feedback for study participants. Diurnal effects on exposures and indoor concentrations by stove group showed clear differences that also warrant further investigation. Improved tools and methods to quantify the time people spend occupying spaces engaged in day-to-day activities (e.g., at home cooking, not cooking, and away from home) and the associated exposures during these times are essential to understanding the magnitudes of various source contributions to overall exposure. These tools and methods are in development and have been deployed in a subset of these study households and results from these measurements are published elsewhere [17]. The relationship between CO and carbonaceous PM<sub>2.5</sub> was weak, suggesting that CO should not be used as a surrogate for PM<sub>2.5</sub> exposure in the region, a finding also supported

by a source apportionment analysis of the organic component of  $PM_{2.5}$  from personal and cooking area samples from this study [13]. Future work will also address the link between cookstove use and CO exposure over time, as stove breakdowns over the two-year study were not uncommon and may have influenced inter-group comparisons.

Supplementary Materials: The following are available online at <a href="http://www.mdpi.com/2073-4433/10/7/402/s1">http://www.mdpi.com/2073-4433/10/7/402/s1</a>, Figure S1: Lascar CO monitors worn on lanyards around the neck or placed inside custom pockets on project t-shirts, Figure S2: Distributions of average CO exposure by time periods relevant to WHO tier-1 standards, Figure S3: Example calibration of a Lascar that returned from the field after deployment to Navrongo, Figure S4: Lascar USB-CO calibration and data quality time line. Some Lascar monitors like #1 and #3 never operated correctly and were returned to the manufacturer. In most cases, the monitors were non-operational upon their return to the CU Boulder Hannigan Lab, so a post-calibration could not be performed, Figure S5: Agreement of daily average Lascar USB-CO duplicates for both calibrated and raw values colored by days since first deployed, Figure S6: Personal exposure by season and stove group, smoothed using b-splines, Figure S7: B-spline smoothed personal CO exposure grouped by primary cook status gender group. '0' values are for non-primary cooks, and '1' is for the females listed as primary cooks. No males were listed as primary cooks, Figure S8: B-spline smoothed personal CO exposure grouped by age and gender group. '0' values are under 5, and '1's are over 5 years of age, Table S1: Descriptive 24 h average CO exposure statistics.

**Author Contributions:** Conceptualization, R.P., E.R.C., C.W., K.L.D., and M.P.H.; formal analysis, R.P., E.R.C., and Y.H.; methodology, R.P., E.R.C., and Y.H.; project administration, E.R.C., E.K., C.W., A.O., and M.P.H.; software, R.P. and E.R.C.; validation, R.P.; visualization, R.P.; writing—review and editing, R.P., E.R.C., Y.H., C.W., K.L.D., and M.P.H.

**Funding:** This publication was developed under Assistance Agreement No. 1211668 awarded by the U.S. Environmental Protection Agency. It has not been formally reviewed by EPA. The views expressed in this document are solely those of the authors and do not necessarily reflect those of the Agency. EPA does not endorse any products or commercial services mentioned in this publication. This work was also funded by a grant from the US National Science Foundation (Award #83542401).

**Ethical Considerations:** This work was reviewed and approved by the Institutional Review Boards at the University of Colorado Boulder, the National Center for Atmospheric Research, and the Navrongo Health Research Center, a part of the Ghana Health Service. The National Center for Atmospheric Research is operated by the University Corporation for Atmospheric Research under the sponsorship of the National Science Foundation.

Acknowledgments: Many thanks to the residents of the Kassena-Nankana districts of Ghana for their patience and participation. Thank you to all the staff and field workers at the Navrongo Health Research Center, for their hard work and dedication, without which this study would not have been possible. A special thanks goes to Natalie Banacos for her editorial administration. Publication of this article was funded by the University of Colorado Boulder Libraries Open Access Fund.

Conflicts of Interest: The authors declare no conflict of interest.

## References

- 1. Astrup, P. Some physiological and pathological effects of moderate carbon monoxide exposure. *Br. Med. J.* **1972**, *4*, 447–452. [CrossRef] [PubMed]
- 2. Longo, L.D. The biological effects of carbon monoxide on the pregnant woman, fetus, and newborn infant. *Am. J. Obstet. Gynecol.* **1977**, 129, 69–103. [CrossRef]
- 3. Smith, K.R.; Samet, J.M.; Romieu, I.; Bruce, N. Indoor air pollution in developing countries and acute lower respiratory infections in children. *Thorax* **2000**, *55*, 518–532. [CrossRef] [PubMed]
- 4. Naeher, L.P.; Brauer, M.; Lipsett, M.; Zelikoff, J.T.; Simpson, C.D.; Koenig, J.Q.; Smith, K.R. Woodsmoke Health Effects: A Review. *Inhal. Toxicol.* **2007**, *19*, 67–106. [CrossRef] [PubMed]
- 5. Carter, E.; Norris, C.; Dionisio, K.L.; Balakrishnan, K.; Checkley, W.; Clark, M.L.; Ghosh, S.; Jack, D.W.; Kinney, P.L.; Marshall, J.D.; et al. Assessing Exposure to Household Air Pollution: A Systematic Review and Pooled Analysis of Carbon Monoxide as a Surrogate Measure of Particulate Matter. *Environ. Health Perspect.* 2017, 125, 076002. [CrossRef] [PubMed]
- 6. Dionisio, K.L.; Howie, S.R.C.; Dominici, F.; Fornace, K.M.; Spengler, J.D.; Adegbola, R.A.; Ezzati, M. Household Concentrations and Exposure of Children to Particulate Matter from Biomass Fuels in The Gambia. *Environ. Sci. Technol.* **2012**, *46*, 3519–3527. [CrossRef] [PubMed]
- 7. Dionisio, K.L.; Howie, S.R.C.; Dominici, F.; Fornace, K.M.; Spengler, J.D.; Donkor, S.; Chimah, O.; Oluwalana, C.; Ideh, R.C.; Ebruke, B.; et al. The exposure of infants and children to carbon monoxide from

- biomass fuels in The Gambia: A measurement and modeling study. *J. Expo. Sci. Environ. Epidemiol.* **2012**, 22, 173–181. [CrossRef] [PubMed]
- 8. McCracken, J.P.; Schwartz, J.; Diaz, A.; Bruce, N.; Smith, K.R. Longitudinal Relationship between Personal CO and Personal PM2.5 among Women Cooking with Woodfired Cookstoves in Guatemala. *PLoS ONE* **2013**, *8*, e55670. [CrossRef] [PubMed]
- 9. Naeher, L.P.; Smith, K.R.; Leaderer, B.P.; Neufeld, L.M.; Mage, D.T. Carbon monoxide as a tracer for assessing exposures to particulate matter in wood and gas cookstove households of highland Guatemala. *Environ. Sci. Technol.* **2001**, *35*, 575–581. [CrossRef] [PubMed]
- 10. Northcross, A.; Chowdhury, Z.; McCracken, J.; Canuz, E.; Smith, K.R. Estimating personal PM2.5 exposures using CO measurements in Guatemalan households cooking with wood fuel. *J. Environ. Monit.* **2010**, *12*, 873. [CrossRef] [PubMed]
- 11. Dickinson, K.L.; Kanyomse, E.; Piedrahita, R.; Coffey, E.; Rivera, I.J.; Adoctor, J.; Alirigia, R.; Muvandimwe, D.; Dove, M.; Dukic, V.; et al. Research on emissions, air quality, climate, and cooking technologies in Northern Ghana (Reaccting): Study rationale and protocol. *BMC Public Health* **2015**, *15*, 126. [CrossRef] [PubMed]
- 12. Oduro, A.R.; Wak, G.; Azongo, D.; Debpuur, C.; Wontuo, P.; Kondayire, F.; Welaga, P.; Bawah, A.; Nazzar, A.; Williams, J.; et al. Profile of the Navrongo Health and Demographic Surveillance System. *Int. J. Epidemiol.* **2012**, *41*, 968–976. [CrossRef]
- 13. Piedrahita, R.; Kanyomse, E.; Coffey, E.; Xie, M.; Hagar, Y.; Alirigia, R.; Agyei, F.; Wiedinmyer, C.; Dickinson, K.L.; Oduro, A.; et al. Exposures to and origins of carbonaceous PM2.5 in a cookstove intervention in Northern Ghana. *Sci. Total Environ.* **2017**, *576*, 178–192. [CrossRef] [PubMed]
- 14. Piedrahita, R.; Dickinson, K.L.; Kanyomse, E.; Coffey, E.; Alirigia, R.; Hagar, Y.; Rivera, I.; Oduro, A.; Dukic, V.; Wiedinmyer, C.; et al. Assessment of cookstove stacking in Northern Ghana using surveys and stove use monitors. *Energy Sustain. Dev.* **2016**, *34*, *67*–76. [CrossRef]
- 15. Dickinson, K.L.; Piedrahita, R.; Coffey, E.R.; Kanyomse, E.; Alirigia, R.; Molnar, T.; Hagar, Y.; Hannigan, M.P.; Oduro, A.R.; Wiedinmyer, C. Adoption of improved biomass stoves and stove/fuel stacking in the REACCTING intervention study in Northern Ghana. *Energy Policy* **2019**, *130*, 361–374. [CrossRef]
- 16. Wiedinmyer, C.; Dickinson, K.; Piedrahita, R.; Kanyomse, E.; Coffey, E.; Hannigan, M.; Alirigia, R.; Oduro, A. Rural–urban differences in cooking practices and exposures in Northern Ghana. *Environ. Res. Lett.* **2017**, 12, 065009. [CrossRef]
- 17. Piedrahita, R.; Coffey, E.; Hagar, Y.; Kanyomse, E.; Verploeg, K.; Wiedinmyer, C.; Dickinson, K.L.; Oduro, A.R.; Hannigan, M.P. Attributing air pollution exposure to emission sources with proximity sensing. *Atmosphere* **2019**, *10*, 395. [CrossRef]
- 18. Burton, P.; Gurrin, L.; Sly, P. Tutorial in biostatistics. Extending the simple linear regression model to account for correlated responses: An introduction to generalized estimating equations and multi-level mixed modeling. *Stat. Med.* **1998**, *17*, 1261–1291. [CrossRef]
- 19. Peretz, C.; Goren, A.; Smid, T.; Kromhout, H. Application of Mixed-effects Models for Exposure Assessment. *Ann. Occup. Hyg.* **2002**, 46, 69–77.
- 20. Awini, E.; Mattah, P.; Sankoh, O.; Gyapong, M. Spatial variations in childhood mortalities at the Dodowa Health and Demographic Surveillance System site of the INDEPTH Network in Ghana. *Trop. Med. Int. Health* **2010**, *15*, 520–528. [CrossRef]
- 21. Birch, M.E. Monitoring of diesel particulate exhaust in the workplace. *NIOSH Man. Anal. Methods NMAM* **2003**, *2154*, 229–259.
- Ofosu, F.G.; Hopke, P.K.; Aboh, I.J.K.; Bamford, S.A. Biomass burning contribution to ambient air particulate levels at Navrongo in the Savannah zone of Ghana. *J. Air Waste Manag. Assoc.* 2013, 63, 1036–1045. [CrossRef] [PubMed]
- 23. Akagi, S.K.; Yokelson, R.J.; Wiedinmyer, C.; Alvarado, M.J.; Reid, J.S.; Karl, T.; Crounse, J.D.; Wennberg, P.O. Emission factors for open and domestic biomass burning for use in atmospheric models. *Atmos. Chem. Phys.* **2011**, *11*, 4039–4072. [CrossRef]
- 24. Andreae, M.O.; Merlet, P. Emission of trace gases and aerosols from biomass burning. *Glob. Biogeochem. Cycles* **2001**, *15*, 955–966. [CrossRef]
- 25. Patterson, E.M.; McMahon, C.K.; Ward, D.E. Absorption properties and graphitic carbon emission factors of forest fire aerosols. *Geophys. Res. Lett.* **1986**, *13*, 129–132. [CrossRef]

26. Atiku, F.A.; Mitchell, E.J.S.; Lea-Langton, A.R.; Jones, J.M.; Williams, A.; Bartle, K.D. The Impact of Fuel Properties on the Composition of Soot Produced by the Combustion of Residential Solid Fuels in a Domestic Stove. *Fuel Process. Technol.* **2016**, *151*, 117–125. [CrossRef]

- 27. Coffey, E.R.; Muvandimwe, D.; Hagar, Y.; Wiedinmyer, C.; Kanyomse, E.; Piedrahita, R.; Dickinson, K.L.; Oduro, A.; Hannigan, M.P. New Emission Factors and Efficiencies from in-Field Measurements of Traditional and Improved Cookstoves and Their Potential Implications. *Environ. Sci. Technol.* **2017**, *51*, 12508–12517. [CrossRef]
- 28. Bhattacharya, S.C.; Albina, D.O.; Myint Khaing, A. Effects of selected parameters on performance and emission of biomass-fired cookstoves. *Biomass Bioenergy* **2002**, *23*, 387–395. [CrossRef]
- 29. L'Orange, C.; DeFoort, M.; Willson, B. Influence of testing parameters on biomass stove performance and development of an improved testing protocol. *Energy Sustain. Dev.* **2012**, *16*, 3–12. [CrossRef]
- 30. Eilers, P.H.C.; Marx, B.D. Flexible smoothing with B-splines and penalties. *Stat. Sci.* **1996**, *11*, 89–121. [CrossRef]
- 31. Morel, P. Gramm: Grammar of graphics plotting for Matlab. JOSS 2018. [CrossRef]
- 32. Who Guidelines for Indoor Air Quality: Selected Pollutants; World Health Organization (Ed.) WHO: Copenhagen, Denmark, 2010; ISBN 978-92-890-0213-4.
- 33. Burwen, J.; Levine, D.I. A rapid assessment randomized-controlled trial of improved cookstoves in rural Ghana. *Energy Sustain. Dev.* **2012**, *16*, 328–338. [CrossRef]
- 34. Ochieng, C.A.; Vardoulakis, S.; Tonne, C. Are rocket mud stoves associated with lower indoor carbon monoxide and personal exposure in rural Kenya? *Indoor Air* **2013**, *23*, 14–24. [CrossRef] [PubMed]
- Smith, K.R.; Mccracken, J.P.; Thompson, L.; Edwards, R.; Shields, K.N.; Canuz, E.; Bruce, N. Personal child and mother carbon monoxide exposures and kitchen levels: Methods and results from a randomized trial of woodfired chimney cookstoves in Guatemala (RESPIRE). *J. Expo. Sci. Environ. Epidemiol.* 2009, 20, 406–416. [CrossRef] [PubMed]
- 36. Gautam, S.; Pillarisetti, A.; Yadav, A.; Singh, D.; Arora, N.; Smith, K. Daily average exposures to carbon monoxide from combustion of biomass fuels in rural households of Haryana, India. *Environ. Dev. Sustain.* **2018**, 1–9. [CrossRef]
- 37. Roden, C.A.; Bond, T.C.; Conway, S.; Osorto Pinel, A.B.; MacCarty, N.; Still, D. Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmos. Environ.* **2009**, *43*, 1170–1181. [CrossRef]
- 38. Ezzati, M.; Mbinda, B.M.; Kammen, D.M. Comparison of Emissions and Residential Exposure from Traditional and Improved Cookstoves in Kenya. *Environ. Sci. Technol.* **2000**, *34*, 578–583. [CrossRef]



© 2019 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).