SUPPLEMENTAL MATERIALS

Biomonitoring Human Exposure to Household Air Pollution and Association with Self-reported Health Symptoms – A Stove Intervention Study in Peru

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### Additional information on the personal CO measurements

As detailed in Commodore et al. (2013), time integrated 48-hour CO measurements were taken using passive CO diffusion tube, i.e. Dräger Diffusion Tube for Carbon Monoxide (Dräger Safety Inc., USA), with a range of 6-600 ppm-hour. CO tubes were broken at the start of the sampling period, placed within the sampling zone of the participants and covered with a tight fitting plastic cap at the end of the 48 hour sampling period. Upon return to the field study site, CO tubes were read independently by two of the authors (AAC and SMH) under a white, bright fluorescent lit laboratory room at a table with a white surface. The least squares regression technique developed by Smith et al (2010) for the Randomized Exposure Study of Pollution Indoors and Respiratory Effects (RESPIRE) was employed. The measured length of stain on the tube was converted to a cumulative exposure in ppm-hour and the latter was divided by the total sampling time to obtain CO personal exposures. The lower and upper limits of detection, derived from the least squares regression equations were 0.2 and 12.5 ppm respectively, with a 100% detection frequency in this study.

Three passive CO diffusion tubes were deployed in each household during the 48-hour sampling period to obtain 48-hour time integrated CO measurements in study kitchens and personal CO exposures of the mother and a child under the age of five. All CO exposure results were reported previously (Commodore et al. 2013). In this manuscript, 48-hour time integrated CO exposures of the mother—who were also the primary household cook—are used in the correlation analysis with the OH-PAH levels in urine samples collected from the same participants.

We employed a two-step quality assurance approach to confirm the precision and accuracy in CO tube measurements. First, we collected repeated same-day measurements in a small subset of households (26 households: 8 controls and 18 intervention; 22 mothers and 4 kitchens) over a one-month period (June to July 2009). We used SAS PROC MIXED to assess the intraclass correlation coefficient (0.74) and the natural log of the residual variance (estimate = 0.33, SE = 0.09). Second, we deployed real time CO measurements using the Dräger Pac III dataloggers fitted with CO-specific sensors and calibrated against standard CO span gas (Dräger Safety Inc., USA) concurrently with the CO tubes for 22 mothers and in 22 kitchens. Pearson correlation coefficients between the CO results from the real time CO measurements and passive CO sampling tubes were 0.94 and 0.82 for kitchen and mother respectively (n=22 each).

References***:***

Commodore AA, Hartinger SM, Lanata CF, Mausezahl D, Gil AI, Hall DB, et al. 2013. Carbon monoxide exposures and kitchen concentrations from cookstove-related woodsmoke in San Marcos, Peru. Int J Occup Environ Health 19: 43-54.

Smith KR, McCracken JP, Thompson L, Edwards R, Shields KN, Canuz E, et al. 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 Expos Sci Environ Epidemiol. 2010;20(5):406–16.

Table S1. Median and interquartile of urinary PAH metabolite concentrations (g/L) and creatinine-adjusted concentrations (g/g creatinine), stratified by stove type.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Urinary PAH metabolites** | | **Control** | | | |  | **Intervention** | | |
| **All control** | **Built-by-NGO** | **Self-improved** | **Traditional** |  | **All intervention** | **No-repair** | **Need-repair** |
|  | N | 179a | 23 | 35 | 121 |  | 155b | 99 | 56b |
| 1-naphthol  1-NAP | g/L | 16.1  (7.83-30.9) | 11.3  (5.8-18.6) | 11.2  (5-22.2) | 20.3  (9.2-36.7) |  | 15.5  (6.7-37.2) | 15.2  (6.6-34.7) | 17.3  (7.94-69.2) |
| g/g crea | 22  (13.6-45.9) | 16.9  (11.3-52.9) | 18.2  (10.6-32.8) | 25.9  (16.8-49.7) |  | 23.6  (12.7-49.9) | 23.1  (12.7-44.3) | 28.4  (13.5-75.1) |
| 2-naphthol  2-NAPc | g/L | 11.2  (6.14-19.9) | 10.1  (4.95-13.7) | 9.63  (3.86-14.5) | 12.5  (6.8-21.4) |  | 7.74  (4.72-13.3) | 7.2  (3.77-13.7) | 8.12  (5.69-12.7) |
| g/g crea | 16  (11.2-22.3) | 14.3  (11.2-24.2) | 13.5  (8.8-20.3) | 16.7  (12.2-22.8) |  | 13.2  (9.4-17.9) | 13.6  (9-17.9) | 13.1  (9.9-18.5) |
| 2-OH-fluorene  2-FLU | g/L | 1.73  (0.87-2.83) | 1.66  (0.84-2.63) | 1.73  (0.76-2.39) | 1.74  (0.98-3.12) |  | 1.43  (0.84-2.87) | 1.24  (0.72-3) | 1.5  (0.98-2.82) |
| g/g crea | 2.55  (1.75-3.8) | 2.71  (1.84-3.91) | 2.35  (1.68-3.61) | 2.57  (1.75-3.8) |  | 2.48  (1.72-3.74) | 2.45  (1.49-3.81) | 2.59  (1.88-3.65) |
| 3-OH-fluorene  3-FLU | g/L | 0.79  (0.43-1.19) | 0.66  (0.34-1.01) | 0.79  (0.27-1.01) | 0.82  (0.47-1.31) |  | 0.61  (0.38-0.99) | 0.61  (0.35-1.02) | 0.62  (0.43-0.98) |
| g/g crea | 1.05  (0.76-1.6) | 1.03  (0.78-1.49) | 1.03  (0.64-1.57) | 1.16  (0.79-1.62) |  | 1.04  (0.69-1.54) | 1.06  (0.63-1.61) | 1.01  (0.78-1.54) |
| 9-OH-fluorene  9-FLU | g/L | 1.65  (0.94-3.02) | 1.55  (1.09-2.28) | 1.92  (0.9-2.79) | 1.56  (0.94-3.14) |  | 1.48  (0.79-2.58) | 1.32  (0.67-2.53) | 1.59  (0.93-2.65) |
| g/g crea | 2.54  (1.6-3.94) | 2.5  (1.84-4.25) | 2.61  (1.86-4.3) | 2.54  (1.53-3.82) |  | 2.49  (1.57-4) | 2.42  (1.42-3.87) | 2.54  (1.75-4.04) |
| 1-OH-phenanthene 1-PHE | g/L | 1.1  (0.57-1.7) | 0.85  (0.58-1.34) | 1.1  (0.52-1.41) | 1.12  (0.59-1.77) |  | 0.91  (0.47-1.57) | 0.86  (0.43-1.6) | 1.02  (0.61-1.57) |
| g/g crea | 1.54  (1.04-2.33) | 1.64  (1.14-2.27) | 1.54  (0.99-2.33) | 1.53  (1.04-2.33) |  | 1.52  (1.04-2.1) | 1.52  (0.99-2.14) | 1.51  (1.13-2.02) |
| 2-OH-phenanthene  2-PHE | g/L | 0.7  (0.37-1.21) | 0.63  (0.48-1.04) | 0.61  (0.33-1) | 0.73  (0.38-1.29) |  | 0.62  (0.32-1.06) | 0.53  (0.28-1.06) | 0.73  (0.47-1.09) |
| g/g crea | 1.08  (0.68-1.66) | 1.2  (0.78-2.02) | 1.08  (0.68-1.59) | 1.06  (0.67-1.62) |  | 1.07  (0.66-1.46) | 1.05  (0.55-1.44) | 1.09  (0.8-1.46) |
| 3-OH-phenanthene 3-PHE | g/L | 0.76  (0.42-1.29) | 0.7  (0.35-1.12) | 0.78  (0.37-1.05) | 0.81  (0.43-1.43) |  | 0.71  (0.39-1.32) | 0.65  (0.33-1.33) | 0.78  (0.47-1.25) |
| g/g crea | 1.19  (0.78-1.73) | 1.21  (0.86-1.58) | 1.13  (0.8-1.65) | 1.2  (0.72-1.74) |  | 1.18  (0.79-1.69) | 1.14  (0.68-1.7) | 1.21  (0.83-1.69) |
| 4-OH-phenanthene 4-PHE | g/L | 0.18  (0.09-0.34) | 0.15  (0.09-0.26) | 0.14  (0.08-0.24) | 0.19  (0.09-0.37) |  | 0.17  (0.09-0.29) | 0.16  (0.08-0.29) | 0.18  (0.09-0.28) |
| g/g crea | 0.27  (0.16-0.42) | 0.22  (0.16-0.39) | 0.25  (0.16-0.36) | 0.27  (0.16-0.46) |  | 0.27  (0.16-0.44) | 0.28  (0.16-0.44) | 0.25  (0.17-0.46) |
| 1-OH-pyrene  1-PYR | g/L | 1.65  (0.87-2.96) | 1.72  (0.92-2.72) | 1.88  (0.77-2.6) | 1.59  (0.92-3.4) |  | 1.59  (0.92-2.79) | 1.29  (0.82-2.99) | 1.8  (1.08-2.59) |
| g/g crea | 2.65  (1.57-3.83) | 3.14  (1.89-4.42) | 2.3  (1.57-3.83) | 2.6  (1.53-3.65) |  | 2.63  (1.7-4.09) | 2.66  (1.63-4.52) | 2.52  (1.91-3.86) |

1. 1-NAP results in two participants in the control group were non-reportable because of chromatographic interference. Hence, the sample size for 1-NAP in the control group was 177.
2. Two urine samples in the need-repair intervention group did not have creatinine results. Hence, the number of samples with creatinine-adjusted concentrations was 153 and 54 for “all intervention” and “need repair” groups, respectively.
3. Urinary 2-naphthol concentrations (both un-adjusted and creatinine-adjusted) were significantly higher in the control group than in the intervention group, t-test, p<0.001

Table S2. Geometric means and 95% confidence intervals of urinary PAH metabolite concentrations (g/L) and creatinine-adjusted concentrations (g/g creatinine), stratified by stove type.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Urinary PAH metabolites** | | **Control** | | | |  | **Intervention** | | |
| **All control** | **Built-by-NGO** | **Self-improved** | **Traditional** |  | **All intervention** | **No-repair** | **Need-repair** |
| N |  | 179a | 23 | 35 | 121 |  | 155b | 99 | 56b |
| 1-OH-naphthalene  1-NAP | g/L | 17.6 [14.5,21.3] | 10.5 [6.7,16.5] | 11.5 [7.5,17.5] | 21.8 [17.2,27.7] |  | 18.9 [15.0,23.9] | 15.9 [12.2,20.7] | 25.9 [16.7,40.0] |
| g/g crea | 28.2 [23.8,33.3] | 19.6 [12.9,29.8] | 19.4 [13.7,27.6] | 33.6 [27.3,41.3] |  | 32.2 [26.1,39.7] | 28.1 [22,35.9] | 41.5 [28.2,61] |
| 2-OH-naphthalene  2-NAPc | g/L | 10.1 [8.98,11.4] | 8.87 [6.23,12.6] | 7.83 [5.88,10.4] | 11.2 [9.7,12.9] |  | 7.76 [6.82,8.84] | 7.24 [6.12,8.56] | 8.78 [7.15,10.8] |
| g/g crea | 16.5 [15,18] | 16.5 [11.9,22.9] | 14.1 [11.7,17] | 17.2 [15.4,19.2] |  | 13.4 [12.3,14.6] | 12.8 [11.5,14.2] | 14.5 [12.6,16.8] |
| 2-OH-fluorene  2-FLU | g/L | 1.6 [1.42,1.8] | 1.51 [1.08,2.13] | 1.4 [1.07,1.84] | 1.68 [1.45,1.95] |  | 1.47 [1.29,1.68] | 1.39 [1.16,1.66] | 1.63 [1.34,2] |
| g/g crea | 2.6 [2.37,2.85] | 2.82 [2.07,3.83] | 2.53 [2.06,3.09] | 2.58 [2.32,2.88] |  | 2.53 [2.31,2.76] | 2.45 [2.17,2.76] | 2.68 [2.36,3.04] |
| 3-OH-fluorene  3-FLU | g/L | 0.68 [0.6,0.76] | 0.57 [0.41,0.78] | 0.57 [0.43,0.74] | 0.74 [0.64,0.86] |  | 0.62 [0.54,0.7] | 0.58 [0.49,0.69] | 0.68 [0.56,0.83] |
| g/g crea | 1.1  [1,1.22] | 1.05 [0.75,1.48] | 1.02 [0.83,1.25] | 1.14 [1.01,1.28] |  | 1.06 [0.96,1.16] | 1.02 [0.9,1.16] | 1.13 [0.97,1.3] |
| 9-OH-fluorene  9-FLU | g/L | 1.59 [1.39,1.82] | 1.55 [1.07,2.25] | 1.47 [1.06,2.04] | 1.63 [1.38,1.92] |  | 1.45 [1.27,1.66] | 1.39 [1.16,1.67] | 1.56 [1.27,1.92] |
| g/g crea | 2.57 [2.31,2.87] | 2.89 [2.14,3.9] | 2.64 [2.01,3.45] | 2.5 [2.19,2.85] |  | 2.49 [2.25,2.76] | 2.45 [2.15,2.8] | 2.56 [2.19,2.99] |
| 1-OH-phenanthene 1-PHE | g/L | 0.98 [0.87,1.12] | 0.91 [0.66,1.27] | 0.9 [0.67,1.21] | 1.03 [0.88,1.2] |  | 0.89 [0.78,1.01] | 0.85 [0.71,1.01] | 0.97 [0.79,1.18] |
| g/g crea | 1.6 [1.45,1.77] | 1.7 [1.24,2.32] | 1.62 [1.28,2.05] | 1.58 [1.4,1.78] |  | 1.53 [1.39,1.68] | 1.5 [1.31,1.71] | 1.59 [1.39,1.82] |
| 2-OH-phenanthene  2-PHE | g/L | 0.67 [0.59,0.76] | 0.69 [0.5,0.95] | 0.59 [0.45,0.78] | 0.69 [0.59,0.81] |  | 0.6 [0.53,0.69] | 0.56 [0.47,0.67] | 0.69 [0.57,0.84] |
| g/g crea | 1.09 [0.99,1.21] | 1.28 [0.91,1.8] | 1.07 [0.85,1.34] | 1.06 [0.94,1.2] |  | 1.03 [0.93,1.14] | 0.98 [0.86,1.13] | 1.13 [0.97,1.31] |
| 3-OH-phenanthene 3-PHE | g/L | 0.72 [0.64,0.82] | 0.68 [0.49,0.93] | 0.62 [0.47,0.82] | 0.77 [0.66,0.9] |  | 0.69 [0.6,0.79] | 0.64 [0.54,0.77] | 0.78 [0.63,0.95] |
| g/g crea | 1.18 [1.06,1.31] | 1.26 [0.89,1.77] | 1.12 [0.9,1.39] | 1.18 [1.04,1.34] |  | 1.18 [1.07,1.3] | 1.13 [1,1.29] | 1.26 [1.09,1.47] |
| 4-OH-phenanthene 4-PHE | g/L | 0.17 [0.15,0.19] | 0.15 [0.1,0.22] | 0.13 [0.1,0.18] | 0.18 [0.15,0.22] |  | 0.15 [0.13,0.18] | 0.15 [0.12,0.18] | 0.17 [0.13,0.21] |
| g/g crea | 0.27 [0.24,0.3] | 0.27 [0.19,0.4] | 0.24 [0.19,0.3] | 0.28 [0.25,0.32] |  | 0.26 [0.24,0.29] | 0.26 [0.23,0.3] | 0.27 [0.23,0.32] |
| 1-OH-pyrene  1-PYR | g/L | 1.55 [1.35,1.78] | 1.62 [1.15,2.29] | 1.37 [0.98,1.9] | 1.6 [1.35,1.9] |  | 1.56 [1.36,1.78] | 1.46 [1.22,1.76] | 1.73 [1.42,2.11] |
| g/g crea | 2.52 [2.25,2.83] | 3.02 [2.14,4.27] | 2.46 [1.89,3.2] | 2.46 [2.13,2.83] |  | 2.66 [2.38,2.96] | 2.59 [2.24,2.99] | 2.79 [2.37,3.29] |

1. 1-NAP results in two participants in the control group were non-reportable because of chromatographic interference. Hence, the sample size for 1-NAP in the control group was 177.
2. Two urine samples in the need-repair intervention group did not have creatinine results. Hence, the number of samples with creatinine-adjusted concentrations was 153 and 54 for “all intervention” and “need repair” groups, respectively.
3. Urinary 2-naphthol concentrations (both un-adjusted and creatinine-adjusted) were significantly higher in the control group than in the intervention group, t-test, p<0.001

Table S3. Pearson correlation (r) between log-transformed creatinine-adjusted urinary OH-PAH concentrations and log-transformed personal CO exposures.

| **Log-Urinary PAH Biomarkers vs. Log-CO** | **r** | ***p-*value** | **Na** |
| --- | --- | --- | --- |
| log\_1\_NAP | 0.16 | 0.01 | 309 |
| log\_2\_NAP | 0.17 | <0.01 | 311 |
| log\_sum\_NAP | 0.17 | <0.01 | 309 |
| log\_2\_FLU | 0.18 | <0.01 | 311 |
| log\_3\_FLU | 0.15 | 0.01 | 311 |
| log\_9\_FLU | 0.24 | <0.01 | 311 |
| log\_sum\_FLU | 0.23 | <0.01 | 311 |
| log\_1\_PHE | 0.18 | <0.01 | 311 |
| log\_2\_PHE | 0.14 | 0.02 | 311 |
| log\_3\_PHE | 0.14 | 0.02 | 311 |
| log\_4\_PHE | 0.12 | 0.049 | 311 |
| log\_sum\_PHE | 0.16 | 0.01 | 311 |
| log\_1\_PYR | 0.13 | 0.03 | 311 |

a. A total of 313 participants (168 control and 145 intervention) had both urinary OH-PAH measurements and personal CO measurements. Of these, two urine samples did not have creatinine results, hence, did not have creatinine-adjusted OH-PAH concentrations. Among the 311 participants with creatinine-adjusted concentration, two did not have valid 1-NAP results due to analytical interferences. Therefore, the sample size for the correlational analysis was 309 for 1-NAP and summed NAP, and 311 for the remaining OH-PAHs.

Table S4. Median OH-PAHs (excluding 1-hydroxypyrene) concentrations (creatinine-adjusted, ug/g creatinine) in selected studies.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Population | 1-NAP | 2-NAP | 2-FLU | 3-FLU | 9-FLU | 1-PHE | 2-PHE | 3-PHE | 4-PHE | Reference |
| Non-smoking women using woodstoves, Peru | 22.5 | 14.4 | 2.52 | 1.05 | 2.5 | 1.54 | 1.07 | 1.18 | 0.27 | This study |
| *Other studies on populations exposed to wood smoke* | | | | | | | | | | |
| Non-smoking women with old woodstoves without chimney, Peru | 19.9 | 19.1 | 3.7 | 1.2 | 3.1 | 1.9 | 1.4 | 1.6 | 0.4 | Li et al., 2011 |
| Non-smoking women with chimney-equipped woodstove, Peru | 9.6 | 10 | 2.5 | 0.8 | 2.4 | 1.5 | 1 | 1.3 | 0.3 | Li et al., 2011 |
| Non-smoking women with indoor open-fire woodstoves, Afghanistan | n/a | n/a | n/a | n/a | n/a | 1.3a | 0.69a,b | 1.43a | 1.01a | Hemat et al, 2012 |
| Charcoal workers exposed to woodsmoke, Brazil | n/a | 9.14c | n/a | n/a | n/a | n/a | n/a | n/a | n/a | Kato et al. 2004 |
| *Reference levels from national surveys* | | | | | | | | | | |
| U.S. adult population (≥20 years) | 1.69 | 4.5 | 0.21 | 0.08 | 0.27 | 0.14 | 0.07 | 0.06 | 0.02 | CDC, 2015 |
| U.S. adult smokers | 14.3 | 1.37 | 0.76 | 0.66 | 0.21 | 0.13 | 0.16 | 0.05 | 0.27 | CDC, 2015 |
| Canadian population (3-79 years) | 1.3 | 3.5 | 0.21 | 0.07 | 0.15 | 0.14 | 0.06 | 0.08 | 0.02 | Health Canada, 2015 |
| German adult population (18-69 years) | n/a | n/a | n/a | n/a | n/a | 0.31 | 0.20b | 0.24 | n/a | Becker et al., 2003 |
| German adult smokers (18-69 years) | n/a | n/a | n/a | n/a | n/a | 0.42 | 0.23b | 0.36 | n/a | Becker et al., 2003 |

a. un-adjusted fresh weight concentrations (ug/L)

b. sum of 2- and 9-hydroxyphenanthrene

c. geometric mean

n/a: not available

Figure S1. Flow diagram for the parent c-RCT study and the current cross-sectional study on HAP exposure

