Household concentrations and exposure of children to particulate matter from biomass fuels in The Gambia

Kathie L Dionisio (DSc)[1,2], Stephen RC Howie (MD)[3], Francesca Dominici (PhD)[4], Kimberly M Fornace (BA)[5], John D Spengler (PhD)[2], Richard A Adegbola (PhD)[3,6], Majid Ezzati (PhD)[7,8*]

1 Department of Global Health and Population, Harvard School of Public Health, Boston, MA, USA
2 Department of Environmental Health, Harvard School of Public Health, Boston, MA, USA
3 Child Survival Theme, Medical Research Council, The Gambia Unit, Fajara, The Gambia
4 Department of Biostatistics, Harvard School of Public Health, Boston, MA, USA
5 Veterinary Epidemiology and Public Health Group, Royal Veterinary College, London, UK
6 Bill & Melinda Gates Foundation, Seattle, WA, USA
7 MRC-HPA Center for Environment and Health, Department of Epidemiology and Biostatistics, Imperial College London, London, UK
8 Department of Epidemiology and Biostatistics, School of Public Health, Imperial College London, London, UK

*Address correspondence to: Majid Ezzati; MRC-HPA Centre for Environment and Health, Department of Epidemiology and Biostatistics, Imperial College London, Norfolk Place, London W2 1PG, UK
Tel: +44 (0)20 7594 0767
Fax: +44 (0)20 7594 3456
E-mail: majid.ezzati@imperial.ac.uk

Number of pages (including cover page): 8

Number of figures: 9

Number of tables: 1

Measurement Methods

Personal and cookhouse CO

Children's CO exposure and cookhouse CO concentration were measured using Drager CO 50/a-D Diffusion Tubes (Drager Safety AG & Co. KGaA, Luebeck, Germany) with a detection range of 50-600 ppm-hr. Measurement methods for children's CO exposure are described in detail elsewhere [1]. In the cookhouse, tubes were placed inside a Drager CO tube holder, and attached to the underside of the shelf on the wooden stand. In both cases, CO color change levels were checked and recorded at 24-h intervals. At each reading, the tube's color change was measured to the nearest millimeter using a metric ruler; millimeter measurements were then converted to ppm-hrs as described elsewhere [1]. Measurement quality control details are described elsewhere [1].

Cookhouse $PM_{2.5}$

We measured integrated $PM_{2.5}$ gravimetrically. A PTFE filter with ring (Pall Life Sciences, Teflo, 0.2 µm pre size, 37 mm diameter) with underlying support pad was placed inside a 37 mm SureSeal Air Monitoring Cassette (SKC Inc., Eighty Four, PA) and connected to a GK2.05SH (KTL) cyclone (BGI Inc., Waltham, MA), with a D_{50} of 2.5 µm (aerodynamic diameter) at 3.5 lpm (±10%). Tygon PVC tubing was used to connect the cyclone to a PCXR8 Universal Sampling Pump (SKC Inc., Eighty Four, PA) drawing air at 3.5 lpm. To conserve battery life, we programmed pumps to draw air for 1 out of every 6 minutes. Flow rates were checked at the beginning and end of each sampling period using a calibrated rotameter or digital mass flowmeter. The mean absolute difference of pre- and post-measurement weights of 31 blank samples was 4.6 µg, equivalent to 1.82 µg/m^3 using target sample volume of 2.52 m^3. The LOD for filter weights (calculated as three times the standard deviation of the mean absolute difference of blanks) was 11.6 µg, with the lowest filter weight being 3 times the LOD. In 10 randomly selected cookhouses, two separate cyclones were co-located as duplicate samples. Mean difference and mean absolute difference between duplicate samples were 3 μg/m^3 (0.8% of the average of duplicate samples) and 26 μg/m^3 (6.8%) respectively. All filters were conditioned in a temperature- and humidity-controlled environment, and were weighed pre- and post-sampling at the Harvard School of Public Health Laboratory. Detailed weighing procedures are provided elsewhere [2].

We measured continuous $PM_{2.5}$ in the cookhouse using DustTrak Model 8520 monitors (TSI Inc., Shoreview, MN). $PM_{2.5}$ concentration was measured every second, averaged and recorded at 1-minute intervals. DustTraks were operated at a flow rate of 0.8 lpm. DustTraks were operated at a flow rate of 0.8 lpm because two upstream external mini-Personal Exposure Monitors (mini-PEMs) (Harvard School of Public Health, Boston, MA) [3] were used as size selective inlets for $PM_{2.5}$. In one mini-PEM, polyurethane foam (PUF) served as the impaction surface, in the second mini-PEM, a level greased well served as the impaction surface. DustTraks were calibrated to a zero filter prior to each sampling session. Following previous studies, continuous $PM_{2.5}$ measurements were corrected using the ratio of the co-located integrated (gravimetric) $PM_{2.5}$ measurement to the average of continuous measurements over the same time period [2, 4]. This correction accounts for error in the light scattering technique of measurement used in the DustTrak machine.

Personal $PM_{2.5}$ exposure

We gravimetrically measured children's $PM_{2.5}$ exposure using Personal Exposure Monitors (PEMs) (Harvard School of Public Health, Boston, MA) [3] with a D_{50} of 2.5 μm (aerodynamic diameter) at 1.8 lpm (±10%) and an internal, level, greased impaction surface. External elutriators were connected to the PEM device. Inside the PEMs, PTFE filters with ring (Pall Life Sciences, Teflo, 0.2 μm pore size, 37 mm diameter) were back-supported by Whatman drain discs. PEMs were connected by Tygon PVC tubing to a Casella Apex Lite or Casella Tuff personal sampling pump (Casella USA, Amherst, NH) drawing air at 1.8 lpm. Flow rate was checked at the beginning and end of each sampling period using a calibrated rotameter or digital mass flowmeter.

The assembled and tested PEM and pump were placed inside the backpack with the elutriator protruding from the backpack. Mothers were asked to have the child wear the backpack during all waking hours, except during bathing. While sleeping, the mother was asked to place the backpack beside the sleeping child.

Supplementary Figure 1: Histogram of weights from AIC and BIC, where:

$$\text{AIC weight}_i = \frac{e^{-\frac{1}{2}*AIC_i}}{\sum_{j=1}^{56} e^{-\frac{1}{2}*AIC_j}}; \qquad \text{BIC weight}_i = \frac{e^{-\frac{1}{2}*BIC_i}}{\sum_{j=1}^{56} e^{-\frac{1}{2}*BIC_j}}$$

Supplementary Figure 2: Mean minute-by-minute corrected continuous cookhouse $PM_{2.5}$.

a) all study households

b) by study site

c) by measurement season

d) by type of fuel used most for cooking.

Mean of correction factors for samples with both continuous and gravimetric data was 0.54±0.70.

Supplementary Figure 3: Children's measured CO and predicted $PM_{2.5}$ exposures (n = 1,266), overlaid on the $PM_{2.5}$-CO relationship (n = 213) in the cookhouse. Note that both axes are on the log scale.

Supplementary Figure 4: Distribution of annual personal $PM_{2.5}$ exposure estimated from annual CO exposure and cookhouse $PM_{2.5}$-CO relationship.

Supplementary Figure 5: Cookhouse $PM_{2.5}$-CO relationship compared to measured child $PM_{2.5}$-CO relationship. Note that both axes are on the log scale.

Note: Linear relationships are shown because we could not estimate the non-linear PM-CO relationship for personal exposure due to relatively small number of measurements. The relationship does not change when restricted to firewood users only, possibly due to the small number of charcoal users in this study subset (1 for child measurements and 20 for cookhouse measurements).

Supplementary Figure 6: Relationship between directly-measured child personal $PM_{2.5}$ exposure and measured cookhouse $PM_{2.5}$ concentrations.

Supplementary Figure 7: Three scenarios of the relationship between $PM_{2.5}$ exposure estimated using two indirect methods: using measured CO exposure and cookhouse CO-$PM_{2.5}$ relationship, and using time-location-activity budgets and cookhouse $PM_{2.5}$ concentrations. See Methods for details on exposure estimation.

a) Scenario 1: $I_{i,j} = 1$

b) Scenario 2: $I_{i,j} = 1$ if the child is ≤ 4 meters from the stove; $I_{i,j} = 0.5$ if the child is > 4 meters from the stove

c) Scenario 3: $I_{i,j} = 1$ if the child is ≤ 4 meters from the stove; $I_{i,j} = 0$ if the child is > 4 meters from the stove

Supplementary Figure 8: Relationship between directly-measured $PM_{2.5}$ exposure and exposure estimated indirectly by applying cookhouse $PM_{2.5}$-CO relationship to CO exposure over the same period, without the single child whose household fuel was charcoal. For this sensitivity analysis, the $PM_{2.5}$-CO relationship was estimated among wood users only.

Supplementary Figure 9: Flow chart of number of measurements. Numbers of valid measurements used in the analysis are reported. See text for total number of measurements.

Table S1: Number of measurements of personal and cookhouse $PM_{2.5}$ and CO.

		Number of children	Number of measurements [a]
Personal exposure	$PM_{2.5}$	31	31
	CO	1181	2263
		Number of households	Number of measurements [a]
Cookhouse	Integrated $PM_{2.5}$	203	219
	Continuous $PM_{2.5}$	116	124
	CO	322	356

[a] Number of valid measurements is reported. See text for total number of measurements.

References

1. Dionisio, K. L.; Howie, S. R.; Dominici, F.; Fornace, K. M.; Spengler, J. D.; Donkor, S.; Chimah, O.; Oluwalana, C.; Ideh, R. C.; Ebruke, B.; Adegbola, R.; Ezzati, M., The exposure of infants and children to carbon monoxide from biomass fuels in The Gambia: A measurement and modeling study. *Accepted manuscript, Journal of Exposure Science and Environmental Epidemiology* **2011**.

2. Dionisio, K. L.; Howie, S.; Fornace, K. M.; Chimah, O.; Adegbola, R. A.; Ezzati, M., Measuring the exposure of infants and children to indoor air pollution from biomass fuels in The Gambia. *Indoor Air* **2008,** *18*, (4), 317-27.

3. Demokritou, P.; Kavouras, I. G.; Ferguson, S. T.; Koutrakis, P., Development and Laboratory Performance Evaluation of a Personal Multipollutant Sampler for Simultaneous Measurements of Particulate and Gaseous Pollutants. *Aerosol Science and Technology* **2001,** *35*, 741-752.

4. Dionisio, K. L.; Arku, R. E.; Hughes, A. F.; Vallarino, J.; Carmichael, H.; Spengler, J. D.; Agyei-Mensah, S.; Ezzati, M., Air Pollution in Accra Neighborhoods: Spatial, Socioeconomic, and Temporal Patterns. *Environmental Science and Technology* **2010,** *44*, (7), 2270-2276.