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Chemical Characterization and Source Apportionment of Household Fine Particulate Matter in Rural, Peri-urban, and Urban West Africa

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ABSTRACT: Household air pollution in sub-Saharan Africa and other developing regions is an important cause of disease burden. Little is known about the chemical composition and sources of household air pollution in sub-Saharan Africa, and how they differ between rural and urban homes. We analyzed the chemical composition and sources of fine particles ($PM_{2.5}$) in household cooking areas of multiple neighborhoods in Accra, Ghana, and in peri-urban (Banjul) and rural (Basse) areas in The Gambia. In Accra, biomass burning accounted for 39−62% of total $PM_{2.5}$ mass in the cooking area in different neighborhoods; the absolute contributions were 10−45 $\mu g/m^3$. Road dust and vehicle emissions comprised 12−33% of $PM_{2.5}$ mass. Solid waste burning was also a significant contributor to household $PM_{2.5}$ in a low-income neighborhood but not for those living in better-off areas. In Banjul and Basse, biomass burning was the single dominant source of cooking-area $PM_{2.5}$, accounting for 74−87% of its total mass; the relative and absolute contributions of biomass smoke to $PM_{2.5}$ mass were larger in households that used firewood than in those using charcoal, reaching as high as 463 $\mu g/m^3$ in Basse homes that used firewood for cooking. Our findings demonstrate the need for policies that enhance access to cleaner fuels in both rural and urban areas, and for controlling traffic emissions in cities in sub-Saharan Africa.

INTRODUCTION

Globally, around 2.8 billion people rely on solid fuels (biomass and coal) for their energy needs.[1] In sub-Saharan Africa, over 90% of the rural population and nearly three-quarters of the urban population use solid fuels as their primary cooking fuels.[1,2] Burning solid fuels in open stoves is associated with high levels of health damaging pollutants, including fine particles ($PM_{2.5}$; particulate matter with aerodynamic diameter ≤2.5 μm). Household air pollution (HAP) from solid fuel use was responsible for an estimated 3.5 million deaths and 4.3% of the global burden of disease in 2010.[3]

A number of studies have measured PM concentrations in rural households, with more recent studies also focusing on PM levels in urban households.[4−9] However, very few studies have analyzed the chemical composition of HAP and the contributions of different combustion and noncombustion sources in developing countries,[10−12] especially the differences in PM chemistry and sources between rural and urban households. In addition to household's own fuel, HAP in urban households is affected by the extent of biomass use in the neighborhood, and by traffic-related sources.[8] The relative roles of these sources may vary by the household and its community's socioeconomic characteristics.

In this paper, we analyze and report the chemical elemental composition of $PM_{2.5}$ in household cooking areas in two West

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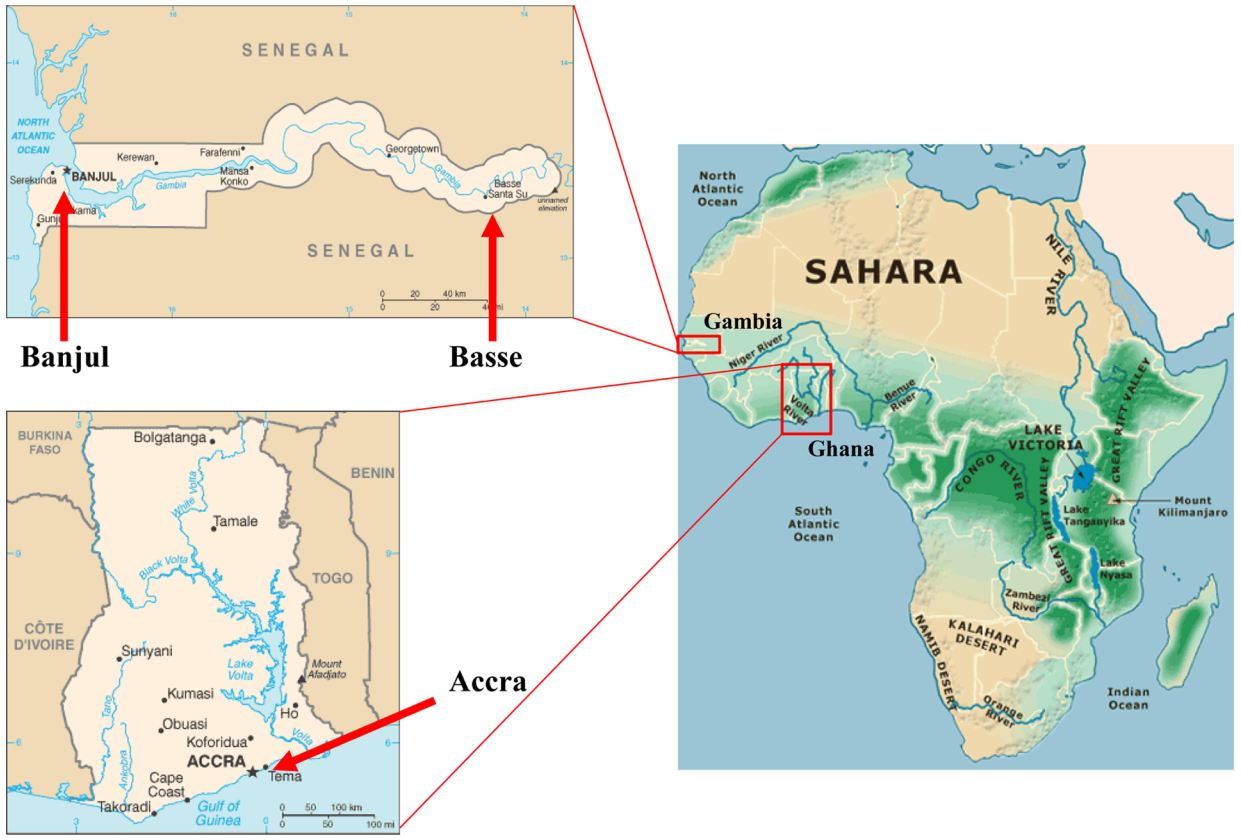

Figure 1. Location of study sites.

African countries using samples analyzed previously for mass concentration. We use elemental composition to identify, and quantify the contributions of, sources of household PM pollution. This study is among the first to present chemical composition and quantitative source apportionment of HAP in developing countries. It is unique in including data from both rural and urban area, and from neighborhoods of varying socioeconomic status, and is based on a significantly larger number of PM samples than previous studies.

MATERIALS AND METHODS

Study Locations. Data used are from Accra, Ghana (urban); a peri-urban area in Greater Banjul, The Gambia; and the Basse area (rural) in the Upper River Region of The Gambia (Figure 1). Accra is a growing urban area whose population increased from 2.9 million in 2000 to 4.0 million in 2010. The Accra study households were from four neighborhoods which lie on a line from the coast to the northern boundaries of the Accra Metropolitan Area (AMA), and have varying socioeconomic characteristics. In two low-income neighborhoods (Jamestown/Ushertown (JT), and Nima (NM)), cooking activities usually take place in an outdoor courtyard, with about 90% of homes using charcoal or firewood as their primary cooking fuel. Street food vendors are very common in JT and NM. In Asylum Down (AD) and East Legon (EL), two higher-income neighborhoods, many homes have modern indoor kitchens and use liquefied petroleum gas (LPG) to cook (Figure 2). Details on socio-demographic characteristics of the four neighborhoods are provided elsewhere.[8,13]

The Gambia is bordered on the North, East, and South by Senegal and has a short strip of Atlantic coastline. It had a

Figure 2. Images of typical cooking areas in Accra, Ghana, and The Gambia. (a) An outdoor cooking area in the courtyard of a compound house in Accra; (b) an indoor kitchen in Accra; (c) outside view of an enclosed cookhouse in The Gambia; and (d) inside view of an enclosed cookhouse in The Gambia.

population of 1.4 million in the 2003 national census, about one-fourth of which lived in the Greater Banjul area, including urban Banjul and less densely populated peri-urban settlements. In peri-urban Banjul most households use firewood or charcoal for cooking. The Basse region is located nearly 400 km east of Banjul and is predominantly rural (Figure 1). Firewood is the universal cooking fuel in Basse. In The Gambia, cooking is typically done in an enclosed cookhouse that belongs to a whole family compound, or in an attached cooking area with a roof (Figure 2), although some outdoor cooking also takes

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Table 1. Average Concentrations of Total $PM_{2.5}$ Mass and Associated Elemental Compositions in Household Cooking Areas

	neighborhood (number of samples)						
chemical species[a]	AD (n = 19)	EL (n = 15)	JT (n = 19)	NM (n = 24)	Banjul-charcoal[b] (n = 21)	Banjul-firewood[c] (n = 119)	Basse (n = 45)
total mass	33 ± 8	25 ± 9	74 ± 22	58 ± 31	134 ± 152	258 ± 208	551 ± 286
MgO	277 ± 165 (0.8%)[d]	78 ± 60 (0.3%)	317 ± 195 (0.4%)	362 ± 220 (0.6%)	518 ± 730 (0.4%)	1028 ± 1053 (0.4%)	3165 ± 3632 (0.6%)
Al_2O_3	2452 ± 1368 (7.4%)	436 ± 234 (1.7%)	3272 ± 1326 (4.4%)	3296 ± 1570 (5.7%)	3619 ± 3919 (2.7%)	4354 ± 3840 (1.7%)	10442 ± 7867 (1.9%)
SiO_2	6047 ± 3512 (18.3%)	1166 ± 626 (4.7%)	9133 ± 4014 (12.3%)	8252 ± 4350 (14.2%)	8175 ± 7959 (6.1%)	10416 ± 8222 (4.0%)	20514 ± 14524 (3.7%)
CaO	484 ± 209 (1.5%)	242 ± 123 (1.0%)	806 ± 382 (1.1%)	974 ± 713 (1.7%)	2304 ± 2089 (1.7%)	4516 ± 4057 (1.8%)	9615 ± 8543 (1.7%)
TiO_2	120 ± 65 (0.4%)	27 ± 15 (0.1%)	153 ± 62 (0.2%)	165 ± 82 (0.3%)	247 ± 278 (0.2%)	318 ± 283 (0.1%)	818 ± 625 (0.1%)
MnO	17 ± 6 (0.1%)	5 ± 1 (<0.1%)	26 ± 8 (<0.1%)	25 ± 12 (<0.1%)	25 ± 26 (<0.1%)	43 ± 40 (<0.1%)	320 ± 200 (0.1%)
Fe_2O_3	869 ± 430 (2.6%)	267 ± 121 (1.1%)	1179 ± 473 (1.6%)	1197 ± 574 (2.1%)	1421 ± 1431 (1.1%)	1617 ± 1376 (0.6%)	3153 ± 2201 (0.6%)
SO_4^{2-}	1845 ± 369 (5.6%)	2535 ± 621 (10.1%)	3888 ± 909 (5.3%)	2511 ± 885 (4.3%)	2088 ± 687 (1.6%)	2505 ± 1374 (1.0%)	3933 ± 2034 (0.7%)
Na	102 ± 105 (0.3%)	355 ± 112 (1.4%)	142 ± 163 (0.2%)	115 ± 167 (0.2%)	83 ± 174 (0.1%)	143 ± 315 (0.1%)	26 ± 124 (<0.1%)
K	1143 ± 492 (3.5%)	1024 ± 564 (4.1%)	3358 ± 1380 (4.5%)	3124 ± 2288 (5.4%)	2398 ± 2323(1.8%)	3113 ± 2287 (1.2%)	10746 ± 5912 (2.0%)
Cl	348 ± 340 (1.1%)	338 ± 262 (1.4%)	1354 ± 1490 (1.8%)	1525 ± 1657 (2.6%)	1757 ± 2110 (1.3%)	2402 ± 2371 (0.9%)	7899 ± 5997 (1.4%)
Zn	34 ± 15 (0.1%)	16 ± 5 (0.1%)	68 ± 35 (0.1%)	45 ± 26 (0.1%)	31 ± 20 (<0.1%)	46 ± 37 (<0.1%)	78 ± 37 (<0.1%)
Br	25 ± 11 (0.1%)	14 ± 4 (0.1%)	39 ± 22 (0.1%)	28 ± 14 (<0.1%)	15 ± 11 (<0.1%)	27 ± 24 (<0.1%)	67 ± 43 (<0.1%)
Pb	12 ± 4 (<0.1%)	7 ± 5 (<0.1%)	26 ± 13 (<0.1%)	55 ± 161 (0.1%)	10 ± 8 (<0.1%)	23 ± 95 (<0.1%)	24 ± 18 (<0.1%)
BC	7 ± 2 (21.2%)	4 ± 2 (16.0%)	11 ± 2 (14.9%)	8 ± 3 (13.8%)	11 ± 18 (8.2%)	26 ± 20 (10.1%)	41 ± 19 (20.3%)
% of total mass accounted	63%	42%	47%	51%	25%	22%	20%

[a]Units are in ng/m^3 except for total mass and BC, which are reported in μg/m^3. [b]Banjul households that use charcoal as primary cooking fuel. [c]Banjul households that use firewood as primary cooking fuel. [d]Mean ± standard deviation. Numbers in parentheses show the percent of total mass from each element or its oxides.

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place.[14] This is similar to rural Ghana, although charcoal is starting to be increasingly used in more affluent and accessible parts of rural Ghana.[15]

Heating is very rare in West Africa. Smoking prevalence was 5% in Accra study households and 33% in those in the Gambia, similar to national estimates reported by the WHO (7% and 31% of adult males, and 2% and 3% of adult females in Ghana and The Gambia respectively).[16] In both settings, people commonly smoke outside the house altogether, and especially outside the cookhouse where men rarely enter.

Study Design. We collected $PM_{2.5}$ samples in 80 homes in Accra between November 2006 and August 2007. The households were selected from those in the Women's Health Study of Accra,[17] whose participants were a random sample of all adult women in Accra, chosen through stratified socio-economic status (SES) and age-group sampling using the 2000 Population and Housing Census of Ghana as the sampling frame. Among this representative sample, we selected households that had more than two members and at varying distances from main roads. Details of household characteristics are described elsewhere.[8] In each household, we collected $PM_{2.5}$ samples over 48 h in the cooking area. Over the same 48 h period, we also collected ambient $PM_{2.5}$ samples at rooftop sites in the same neighborhoods as the study households. In The Gambia, we collected 72-h $PM_{2.5}$ samples in cooking areas of 203 homes (154 in Banjul and 49 in Basse) between August 2007 and June 2010. Study households were from an epidemiological study of pneumonia in young children between 2 and 59 months of age, including severe and nonsevere pneumonia cases and individually matched healthy controls recruited from each case's neighborhood. Household characteristics are described in detail elsewhere.[18] The number of valid samples is provided in Table 1.

PM Measurement and Analytical Methods. We placed monitors in the cooking area of each household at about 1 m above ground level and 1 m away from cooking stoves. Ambient PM monitors in Accra were located on rooftops of residences or businesses, at heights of 4 to 7 m above ground level.[13]

PM mass measurement methods have been described in detail previously.[8,18] In summary, $PM_{2.5}$ samples were collected on PTFE filters (Pall Life Sciences; Teflo, 0.2-μm pore size, 37-mm diameter), back-supported by a Whatman drain disc and placed inside a 37 mm SureSeal Air Monitoring Cassette (SKC Inc., Eighty Four, PA). In Accra, $PM_{2.5}$ samples were collected using either a Harvard impactor[19,20] or a personal exposure monitor (PEM) (BGI Inc., Waltham, MA) both with a D_{50} of 2.5 μm at 4 L per minute (LPM) ($\pm$10%). The Harvard impactor had two size-selective inlets in series, each with a preoiled impactor plate serving as the impaction surface to reduce the effects of particle bounce. The PEM had an internal level greased well serving as the impaction surface. In The Gambia, $PM_{2.5}$ samples were collected using a GK2.05SH (KTL) cyclone (BGI Inc., Waltham, MA), with a D_{50} of 2.5 μm at 3.5 LPM ($\pm$10%). In The Gambia, pumps were programmed to draw air for 1 out of every 6 min to conserve battery life. In both Ghana and The Gambia, air flow rates through the filters were measured at the beginning and end of each sampling period using a calibrated rotameter or digital mass flowmeter.

PM mass was measured on a Mettler Toledo MT5 microbalance at the Harvard School of Public Health Laboratory, after being conditioned in a temperature and humidity controlled environment (20.5 $\pm$ 0.2 °C, 39 $\pm$ 2% RH) for at least 24 h and statically discharged via a polonium source. In both pre- and post-weighing, filters were weighed twice; if these two masses were not within 5 μg of one another, they were weighed a third time. The mean of the two masses within 5 μg of one another was used for calculating concentrations. Final filter weights were adjusted using an air buoyancy correction.[21] Blanks and duplicate side-by-side measurements were collected in multiple homes and on rooftops. We excluded $PM_{2.5}$ samples for which the monitor operated for less than 85% of the designed measurement period or if there was evidence of broken connection in the air flow system. Details on quality control of measurements are provided elsewhere.[8,18]

The elemental concentrations of the samples were quantified by energy dispersive X-ray fluorescence (ED-XRF) using a Shimadzu EDX-700HS spectrometer (Shimadzu Corp., Japan) at the Institute of Astronomy, Geophysics and Atmospheric Science, University of Sao Paulo, Brazil. X-ray fluorescence analysis is a nondestructive analytical method often used for determining elemental concentrations for sodium (Na) through lead (Pb). Measured elements in our study included sodium (Na), magnesium (Mg), aluminum (Al), silicon (Si), sulfur (S), chlorine (Cl), potassium (K), calcium (Ca), titanium (Ti), vanadium(V), chromium (Cr), nickel (Ni), manganese (Mn), iron (Fe), zinc (Zn), bromine (Br), and lead (Pb). For reporting the chemical composition, we converted the major crustal elements to their most abundant oxide forms as most of earth's crust consists of oxide minerals. We also converted sulfur (S) to sulfate (SO_4^{2-}), the most common form of sulfur in the atmosphere. Other elements, whose forms in the atmosphere vary by source, are reported in their elemental form. Gallium (Ga), selenium (Se), rubidium (Rb), yttrium (Y), niobium (Nb), barium (Ba), and hafnium (Hf) were excluded from the analysis because a relatively large number of the measured concentrations were below the limit of detection.

Black carbon (BC) concentration was estimated using data on reflectance coefficients, measured by an EEL smoke stain reflectometer (Model 43D by Diffusion Systems Ltd., London UK). For 52 ambient samples, we also collected colocated PM samples on quartz fiber filters, used to directly measure BC concentrations. We used site-days with both direct measurement and data on reflectance coefficient to develop a regression equation that related the natural logarithm (ln) of BC concentration to the reflectance coefficient.[22] The regression equation was then used to estimate the BC concentration of the remaining samples whose reflectance, but not BC, had been measured.

Statistical Methods for Source Identification and Contributions. We used the Positive Matrix Factorization (PMF) receptor model (EPA PMF 3.0) to identify and quantify the sources of $PM_{2.5}$ in household cooking areas and neighborhood ambient sites. PMF is a multivariate receptor model that computes optimal source profiles and contributions by minimizing the sum of the squared residuals, weighted by uncertainty of elemental concentrations.[23] The PMF model relies on the premise that while the contributions of a source to particle pollution vary over time and space, source profiles themselves are relatively stable over time. Because source profiles may differ between study areas, we analyzed source contributions separately for each study neighborhood/area. In the analysis of the Accra data, we pooled household and ambient data in each study neighborhood to increase the number of samples. This stabilized the source profiles and

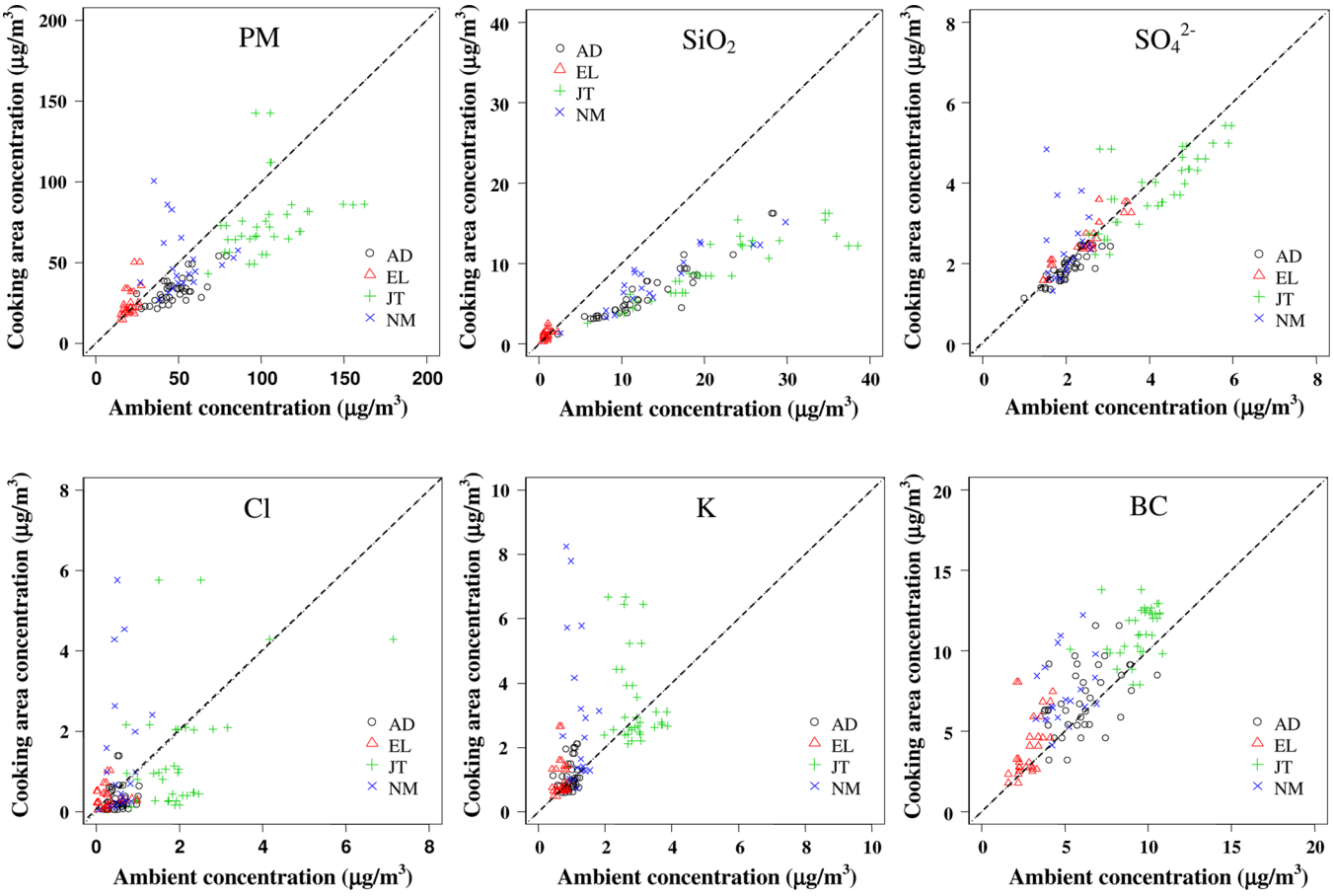

Figure 3. The relationship between cooking area and ambient $PM_{2.5}$ mass and selected elemental/oxide concentrations using simultaneously-collected household and ambient samples.

allowed us to compare source contributions at household and ambient sites.

RESULTS AND DISCUSSION

Total $PM_{2.5}$ Mass. $PM_{2.5}$ mass concentrations were described in detail previously.[8,18] In summary, in Accra, households in the two low-income neighborhoods had higher cooking-area $PM_{2.5}$ (74 ± 22 $\mu g/m^3$ in JT and 58 ± 31 $\mu g/m^3$ in NM) than households in the two wealthier neighborhoods (33 ± 8 $\mu g/m^3$ in AD and 25 ± 9 $\mu g/m^3$ in EL) (Table 1). Cooking-area $PM_{2.5}$ concentrations in Gambian homes were substantially and significantly higher (p-values <0.01) than those in Accra: 551 ± 286 $\mu g/m^3$ in rural Basse, and 258 ± 208 $\mu g/m^3$ for firewood-using households and 134 ± 152 $\mu g/m^3$ for charcoal-using households in peri-urban Banjul (Table 1).

Chemical Composition of Cooking-Area $PM_{2.5}$. In Accra, oxide minerals such as Al_2O_3 and SiO_2 were lower in homes in wealthier EL than in the other three neighborhoods (Table 1 and Figure 3), possibly because measurements in those three neighborhoods were done around the Harmattan period, when a large amount of Saharan dust is carried from the Sahara by trade winds and deposits in the city. In all neighborhoods, SiO_2 concentrations were lower in the cooking area than at the ambient site (Figure 3), hinting toward outdoor sources of SiO_2. Sulfate concentrations were mostly equal in household and ambient environments with a few exceptions in NM and JT. In contrast, the concentrations of K and BC were generally higher in the cooking area than in ambient air, indicating that K and BC may have originated from household activities, presumably cooking with biomass. There was no consistent household vs ambient pattern in Cl, although Cl concentrations tended to be higher at the JT ambient site, the neighborhood closest to the coast. Chlorine is a major component of fresh sea salt and, along with K and BC, is also associated with biomass burning.[24−26]

In The Gambia, all elements, except Na, had higher concentrations in Basse than in Banjul (Table 1). Banjul is on the coast and receives sea salt aerosols, which contain Na. In Banjul, total $PM_{2.5}$ mass, BC, and all measured elements were higher in households that used firewood than those cooking with charcoal (Table 1). BC was the component with the largest ratio of concentrations between households using firewood and charcoal.

Source Contributions to Cooking-Area $PM_{2.5}$. We identified 5−6 potential $PM_{2.5}$ sources for Accra homes and 4−5 potential sources for households in Banjul and Basse (Figures 4 and 5). In Accra, biomass burning, consisting of fresh biomass smoke and aged biomass particles, was the largest contributor to cooking area $PM_{2.5}$, accounting for 39−62% of total $PM_{2.5}$ mass in different neighborhoods. The absolute contribution of biomass burning was largest in JT (45 $\mu g/m^3$) and smallest in EL (10 $\mu g/m^3$). In comparison, biomass burning contributed between 7 $\mu g/m^3$ (AD) and 37 $\mu g/m^3$ (JT) to ambient samples collected over the same period as the household samples, accounting for 15−42% of total ambient $PM_{2.5}$ mass.

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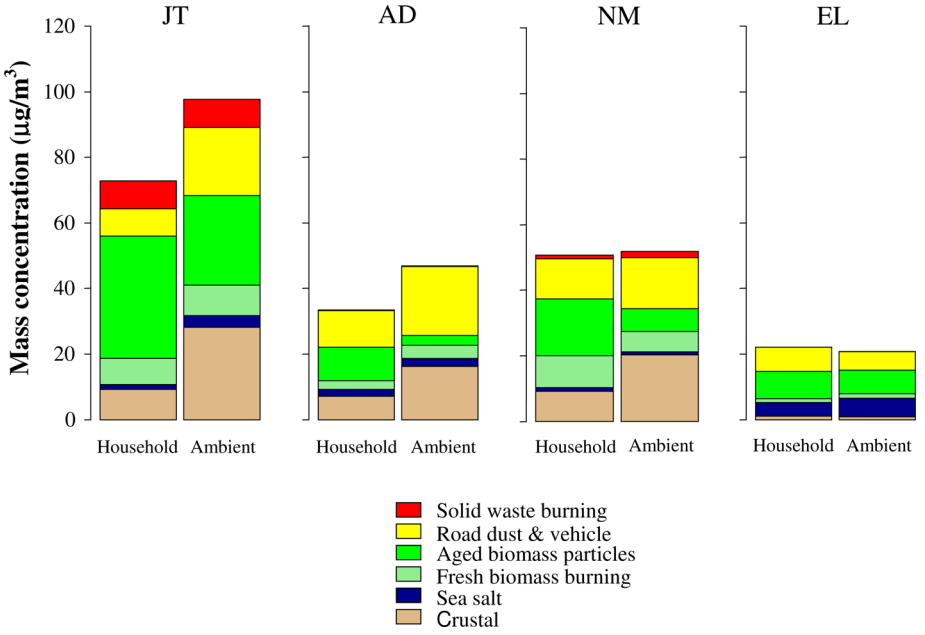

Figure 4. Average contributions of sources to $PM_{2.5}$ mass at ambient sites and the household cooking areas of four study neighborhoods in Accra, Ghana. Sea salt is characterized primarily by Na, Cl, and S (in aged sea salt, the Cl ion is replaced by SO_4^{2-} as a result of reaction with sulfuric acid [36]). Crustal sources are characterized by Al, Si, Mg, Ti, Mn, and Fe. Biomass smoke is primarily characterized by K, Cl, S, and BC.[25,26] Potassium in biomass smoke is emitted mostly as potassium salts such as KCl and K_2SO_4; with aging, most KCl particles are converted to K_2SO_4.[24,37] Road dust and traffic emission are characterized by Al, Si, Ca, Fe, Zn, and BC.[38,39] Solid waste burning is characterized by Br.[40] 95% confidence intervals (CIs) of total mass concentrations (in $\mu g/m^3$) at household sites were (64, 84) in JT, (29, 37) in AD, (45, 70) in NM, and (20, 30) in EL. 95% CIs of total mass concentrations at ambient sites were (94, 115) in JT, (45, 55) in AD, (46, 60) in NM, and (18, 23) in EL.

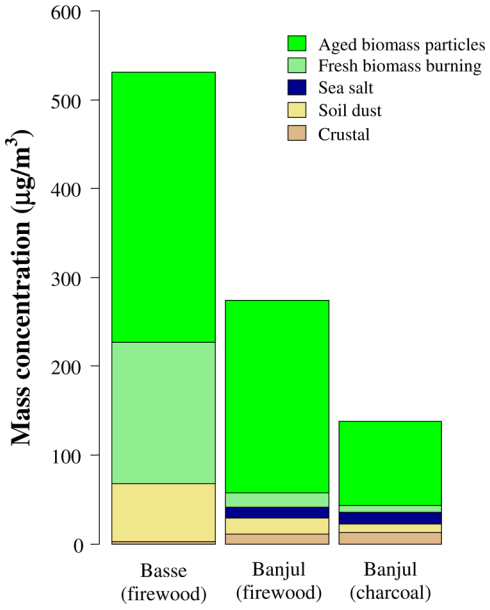

Figure 5. Average contributions of sources to $PM_{2.5}$ mass in household cooking areas in Banjul and Basse, The Gambia. For Banjul, results are stratified by primary household cooking fuel (firewood or charcoal). Soil dust is characterized by Al, Si, Mg, Ca, and Fe. Key features of the chemical profiles of other sources are provided in Figure 4 caption. 95% CIs of total mass concentrations (in $\mu g/m^3$) were (468, 635) at Basse homes, (237, 318) at Banjul homes using firewood, and (77, 202) at Banjul homes using charcoal.

Road dust and vehicle emissions comprised 12−33% of cooking-area $PM_{2.5}$ mass in different parts of Accra, possibly because many roads are unpaved and there is frequent traffic congestion throughout the city. Further, Accra homes are more open, with cracks in the walls and windows and doors that are often kept open, facilitating air exchange between household and ambient environments. The road dust contribution was larger at the ambient sites than in the households within the same neighborhood, except in EL, where the contributions were comparable (7 $\mu g/m^3$ for household samples and 6 $\mu g/m^3$ for ambient samples). The EL ambient site was farther from major and secondary roads than many of the study households.

EL household $PM_{2.5}$ had the lowest contribution from crustal dust (1 $\mu g/m^3$) but the highest contribution from sea salt aerosols (4 $\mu g/m^3$) (Figure 4). This result is not expected based on the location of this site alone (farthest from the ocean) and is likely due to the timing of measurements in different neighborhoods. EL measurement were done between July and August when prevailing winds blow more intensely from the south−southwest, from the Atlantic Ocean; measurements in other neighborhoods were mostly done around the Harmattan season, when a large amount of crust dust is brought from the Sahara Desert into the city by trade winds blowing from north to south. On average, crustal dust accounted for 13−21% of total cooking-area $PM_{2.5}$ mass in AD, JT, and NM. By contrast, crustal dust contributed 29−39% of ambient $PM_{2.5}$ mass. Solid waste burning was a significant source only in JT, contributing 12% (9 $\mu g/m^3$) of the total cooking-area $PM_{2.5}$ mass (Figure 4).

Cooking-area $PM_{2.5}$ in the Banjul and Basse areas of The Gambia was dominated by biomass burning particles, which accounted for 74−87% of total $PM_{2.5}$ mass (Figure 5). Biomass burning contributed more in Basse (463 $\mu g/m^3$), followed by households in Banjul that use firewood (233 $\mu g/m^3$). However, even in Banjul households that use charcoal, biomass smoke contributed 102 $\mu g/m^3$ of $PM_{2.5}$ mass, larger than total mass in any of the Accra sites. This occurred partly because The

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Gambian households tended to cook for longer hours than those in Accra, where purchasing street food for meals is common. It may also be partly due to differences in cooking area configuration—cooking occurs in an enclosed location in about 80% of The Gambia homes, and in open air in 54% of all Accra homes (86% of those that used biomass).[8,14] There were some sea salt aerosols (13 $\mu g/m^3$) in Banjul samples, but not in those from Basse, which is located hundreds of kilometers from the coast (Figure 1). Other sources in The Gambian homes were crustal material and soil dust, which together accounted for less than 15% of total $PM_{2.5}$ mass.

Implications of Findings on Source Contributions. Our findings on chemical composition and sources of household fine particles in West Africa highlight the important role of biomass fuels in both rural and urban homes. Biomass continues to be a common source of energy in sub-Saharan Africa, especially in low-income households and communities, for a number of reasons. The prices of cleaner fuels such as LPG and electricity may be too high for poorer households. Fuel switching requires strategies that improve affordability for the poor. Other important obstacles to fuel switching include the initial cost of a clean stove, uncertainty about regular supply, and uncertainty about future prices.[27] The initial cost of a clean stove requires household-level financial interventions, in the form of a one-time subsidy, or replacing lump-sum payment with gradual payments. Other obstacles go beyond individual households and require policies that improve energy infrastructure for whole communities and regions, and ensure that the poor are protected from macro-economic trends that affect energy prices and supply, as attempted for food and nutrition.

Our findings also highlight the importance of road dust and vehicle emissions as a pollution source in urban homes. In Accra, traffic-related emissions accounted for 12−33% (7−12 $\mu g/m^3$) of household $PM_{2.5}$ mass concentration, compared with only 3−8% (0.3−1.3 $\mu g/m^3$) in some studies in high-income countries.[28,29] The higher contribution of traffic-related emissions in Accra may be due to poor road and car conditions (mostly unpaved roads and a large number of imported used high-emission cars). If African cities follow the motorization trend of Asian and Middle Eastern megacities, then there will be even more traffic-related pollution. Curbing and reducing this important source requires more stringent vehicle emissions standards, paving road surfaces, and inevitably a pro-poor public transportation system, as implemented in cities like Bogota.[30]

Finally, there is some evidence from high-income countries that chemical characteristics of particles may be a determinant of toxicity above and beyond its mass and size distribution, although this question is the subject of ongoing research.[31−34] There is a need to study whether the differences in the chemical composition of PM across neighborhoods and areas affect their hazardous effects for human health.

■ STRENGTHS AND LIMITATIONS

To the best of our knowledge, this study is one of the first to investigate the chemical composition and sources of household $PM_{2.5}$ in developing countries, in particular in sub-Saharan Africa. Household cooking-area $PM_{2.5}$ was measured in poor and affluent urban neighborhoods, as well as in a peri-urban and a rural area. This data allowed us to examine to some extent how household air pollution levels, composition, and sources differ in relation to socioeconomic status. Moreover, in Accra, $PM_{2.5}$ was measured simultaneously in household cooking areas and at neighborhood ambient sites, which enabled us to compare source contributions inside and outside homes.

Similar to all field studies, our work is affected by some limitations. Logistical difficulties and time-intensive field work restricted our ability to conduct measurements simultaneously in all Accra study neighborhoods. This limited our ability to distinguish differences in source contributions across neighborhoods from those due to season. However, the comparison of elemental composition of ambient samples in this analysis with those from year-long measurement in all neighborhoods[22] shows that source contributions of biomass burning and traffic-related emission were relatively stable throughout the year. The most significant effect of lack of year-long household samples is likely to be on crustal and sea salt particles whose contributions vary by season. In addition, it would have been desirable to obtain repeated measurements to better examine day-to-day variation in elemental composition, but this was beyond our resources. Similarly, it would have been ideal to establish chemical composition and sources in the living area of each house. In terms of total mass, in Accra, $PM_{2.5}$ concentrations in cooking and living areas had similar levels and temporal patterns, perhaps because the living area is well-ventilated.[8] Limited data from The Gambia also shows correlated $PM_{2.5}$ between cooking area and children's sleeping and playing areas, with noticeably lower concentrations in the latter.[35]

Further limitations in this analysis include the reliance of source apportionment methods on relatively stable source profiles across samples. In reality, source profiles may differ somewhat, even within the same neighborhood. In addition, determining the number of sources and labeling PM sources in PMF analysis is based on both the goodness of model fit and the physical meaningfulness of the resolved factors. While this involves some subjectivity, we compared results from multiple trials with different numbers of factors to ensure that our conclusions are robust. Species with large uncertainty and a high percentage of data below the limit of detection were not included in the analysis. The Accra results would also have benefited from a larger number of samples. All of these limitations should inform the design of future research on the sources of household air pollution in cities in low- and middle-income countries.

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Notes

The authors declare no competing financial interest.

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