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Emissions Measurements from Household Solid Fuel Use in Haryana, India: Implications for Climate and Health Co-benefits

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ABSTRACT: A large concern with estimates of climate and health co-benefits of "clean" cookstoves from controlled emissions testing is whether results represent what actually happens in real homes during normal use. A growing body of evidence indicates that in-field emissions during daily cooking activities differ substantially from values obtained in laboratories, with correspondingly different estimates of co-benefits. We report $PM_{2.5}$ emission factors from uncontrolled cooking (n = 7) and minimally controlled cooking tests (n = 51) using traditional chulha and angithi stoves in village kitchens in Haryana, India. Minimally controlled cooking tests (n = 13) in a village kitchen with mixed dung and brushwood fuels were representative of uncontrolled field tests for fine particulate matter ($PM_{2.5}$), organic and elemental carbon ($p > 0.5$), but were substantially higher than previously published water boiling tests using dung or wood. When the fraction of nonrenewable biomass harvesting, elemental, and organic particulate emissions and modeled estimates of secondary organic aerosol (SOA) are included in 100 year global warming commitments (GWC_{100}), the chulha had a net cooling impact using mixed fuels typical of the region. Correlation between $PM_{2.5}$ emission factors and GWC ($R^2 = 0.99$) implies these stoves are climate neutral for primary $PM_{2.5}$ emissions of 8.8 ± 0.7 and 9.8 ± 0.9 g $PM_{2.5}$/kg dry fuel for GWC_{20} and GWC_{100}, respectively, which is close to the mean for biomass stoves in global emission inventories.

KEYWORDS: *Emission Factors, Global Warming Commitments, Solid Fuel, Biomass Cookstoves, India*

1. INTRODUCTION

Cleaner cookstoves can have direct health benefits through reductions in primary pollutant exposures in homes, through reduced downstream ambient pollution by preventing formation of secondary air pollutants including ozone and secondary organic aerosol, and through reductions in emissions affecting climate, including black carbon and short-lived climate forcing compounds.[1−3] Recent chemical characterization of emissions of particulate matter and 76 volatile organic compounds (VOCs)[2] from minimally controlled cooking tests in India demonstrated that use of dung patties leads to approximately three times more secondary organic aerosol and ozone formation compared to brushwood.[1] Because emissions of particulate and volatile species are dependent on combustion conditions, these data demonstrate the need to evaluate whether combustion conditions during either in-home or laboratory testing are representative of typical household cooking activities. They additionally highlight the need for methods that allow collection of household emissions measurements that are representative of combustion conditions during typical household cooking activities.[4]

Evaluating the climate and health benefits of cookstoves can help prioritize policies that maximize co-benefits for near-term climate, human health, agriculture, and the cryosphere.[5] In addition, climate finance, based on emission reduction credits, provides a mechanism to reduce up-front installation costs for clean cooking solutions, allowing them to be competitive with cost-effective health interventions.[6,7] While a number of studies have estimated climate and health implications of cookstoves,[8−11] they have been hampered by a lack of emissions data from stoves during normal usage. Furthermore, few

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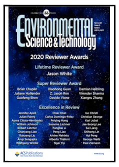

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detailed co-benefit analyses have been based on actual measurements of stove performance in-field.[12] A growing body of evidence has demonstrated substantial differences between laboratory testing and in-field observations.[13−18] There are therefore significant concerns whether climate and health co-benefits estimated from controlled emissions testing represent the reality in homes.

In this paper, we use Global Warming Potentials (GWP) to estimate climate forcing of emissions from uncontrolled and minimally controlled cooking tests in villages in Haryana, India using a full suite of climate forcing species including OC and secondary organic aerosol (SOA) and regional estimates of the fraction of nonrenewable harvesting of biomass. We show that CO_2-equivalent emissions from these stoves in India would be close to carbon neutral at both the 20- and 100-year time horizons on a g/kg dry fuel basis, which suggests that carbon offsets from cookstoves are much smaller than previously estimated.

2. METHODS

In the current study, emissions from uncontrolled testing in village homes were compared with emissions from minimally controlled testing in a village kitchen and emissions from laboratory based emissions testing in the literature. Cookstoves included chulhas (traditional Indian mud cookstoves used for cooking), angithis/haros (two names for similar traditional Indian mud cookstoves, used primarily to cook animal feed, differing only in that haros are fixed in place while angithis are portable), and the Philips HD4012 fan stoves (a modern, fan-driven, top-loading partal-gasifier stove). Table 1 lists an overview of all novel testing presented in this paper. Uncontrolled testing (n = 7 meals each for the chulha and Philips stove) was defined as measurement of emissions in situ in a village kitchen during daily cooking activities without prescribing quantity or amounts of foods to be cooked, amount of fuel, or fuel choice. As much as possible, the objective was to measure emissions during normal daily cooking behaviors. Minimally controlled cooking (n = 14 wood, 15 dung, and 12 mixed fuel meals) was defined as cooking a meal for a typical family size in the region with either rice or chapattis (a type of Indian flatbread) as the starch with dahl and market vegetables selected by the cook. An additional 10 minimally controlled cooking tests consisted of simmering animal fodder with angithis using dung fuel. In minimally controlled tests, fuels were preweighed and fuel types were prescribed, but the amount of fuel and ratios of mixed fuels were determined by the cook. In contrast, laboratory testing of similar stoves and fuels was defined as emissions tests using water boiling test protocols published in the literature.

Table 1. Emissions Measurements of Uncontrolled and Minimally Controlled Tasks

stove type	cooking task	fuel	minimally controlled (n)	uncontrolled (n)
chulha	meal	mixed	12	7[a]
	meal	brushwood	14	N/A
	meal	dung	15	N/A
angithi	animal fodder	dung	10	N/A
philips	meal	brushwood	N/A	7[b]

[a] Measured in five homes. [b] Measured in four homes.

For the uncontrolled in-home testing, village homes were identified in Manpur, Gehlab, Banchari, and Mitrol within the SOMAARTH demographic site.[19,20] SOMAARTH is the name of an International Clinical Epidemiology Network (INCLEN) Trust Demographic Development and Environmental Surveillance Site in Palwal district, Haryana, India encompassing 51 contiguous villages from three blocks with over 200 000 population. Minimally controlled testing was also done in SOMAARTH and was conducted in an outdoor kitchen in the village of Khatela, Palwal, Haryana, India (Supporting Information Figure S1). Palwal District has ∼170 000 homes in which 39% use wood as their primary cooking fuel, followed by dung (25%), and crop residues (7%).[21] In SOMAARTH, the percent of households using biomass as their primary fuel for cooking has been estimated at 96.6%.[22]

Uncontrolled Testing. Sampling occurred during both morning and evening cooking periods typical of this region. For the uncontrolled in-home testing fuel selection, meal-type, fuel loading, and fire-tending were determined by the individual cooks in each household. Fuel and food were not provided and homes were selected as a convenience sample in the village from households willing to participate.

Minimally Controlled Cooking. A local cook was hired to prepare a meal for four people based on the typical household size in the region with either rice or chapatti (an Indian flatbread) as starch, vegetables, and dahl based on market availability. Each meal was prepared by the same local cook who was instructed to use a specific fuel type from preweighed loads of fuel (dung or brushwood or both mixed together) and moisture content (wet or dry), but determined fuel loading and fire-tending according to her cooking preference. When fuels were mixed, the specific ratio of dung to brushwood was chosen by the cook. The cook was also instructed to cook typical daily village meals rather than food types cooked on special occasions. No other instructions regarding cooking were given to the cook in order to maximize the cook's ability to cook in their typical fashion.

Fuel Assessment. For both the minimally controlled and uncontrolled cooking tests, the total mass of each fuel type consumed was calculated by weighing the total fuel of each type before and after each cooking event, including residual unburned fuel from the stove, using a postal scale (model PE10, Pelouze, China). Fuel moisture was assessed using a 9 V digital moisture meter for both brushwood and dung patties (model: 50270, SONIN Inc., China). Moisture measurements for dung patties were adjusted in accordance with Gautam et al.[23]

Sampling and Analysis. For all testing, emissions were sampled and analyzed for CO_2, CO, and $PM_{2.5}$ using established methods.[24] In brief, three-pronged metal probes were hung above each stove and emissions were sampled using PCXR8 pumps (SKC Inc. Universal, PA). Simultaneous measurements were conducted in the kitchen yard for determination of background concentrations for subtraction during analysis. Flows were evaluated via a Mesalabs Defender 530 (BGI Mesa Laboratories, Lakewood, CO) during the in-home testing and a TSI 4140 flowmeter (TSI, Shoreview, MN) during controlled testing before and after each cooking event. Pumps were turned on before cooking began so that entire cooking events were captured and turned off when cooking was completed. Cooking was determined as finished when the cook indicated they were done and all unburned or smoldering fuel was weighed and then subtracted from the total fuel

Table 2. Geometric Mean MCEs and EF Consumption Rates for Uncontrolled Tests in Homes and Minimally Controlled Tests Using Mixed Fuels in the Chulha[a]

	uncontrolled ($n = 7$)	minimally controlled ($n = 13$)	difference in arithmetic mean	$P(T \leq t)$ two-tail
modified combustion efficiency (MCE)	89.2% (89.2%; 1.1%)	86.4% (86.4%; 2.5%)	2.80%	<0.01
$PM_{2.5}$ EF (g/kg dry fuel)	8.7 (11.0; 7.6)	12.3 (12.5; 2.5)[b]	−1.6	0.61
EC EF (g/kg dry fuel)	0.4 (0.6; 0.5)[b]	0.6 (0.7; 0.2)[b]	−0.1	0.69
OC EF (g/kg dry fuel)	3.9 (6.0; 5.7)[b]	5.6 (5.7; 0.9)[b]	0.3	0.91
consumption rate (dry fuel g/min)	22 (24; 9)	26 (26; 2)	−2	0.54

[a] Notes: Values are geometric mean (arithmetic means; arithmetic standard deviation). P values from Welch's two-sided t tests for differences in the arithmetic mean. [b] Sample size reduced by one due to a damaged filter.

loading. Johnson et al. reported less than a 1% difference between modified combustion efficiency (MCE, the ratio of emitted moles of CO_2 to CO_2 and CO) between sampling hoods and the three-pronged probes used in this study.[24] Similarly, Zhang et al. also reported no significant changes in emission ratios between flue gas and hood samples.[25] Concentrations of CO_2 and CO were analyzed for all samples using a TSI Q-Trak 7575 instrument (TSI, Shoreview, MN) and adjusted for background ambient concentrations.[26]

Size selection of aerosols to collect $PM_{2.5}$, EC, and OC was achieved using a SCC 1.062 (Triplex) personal sampling cyclone (Triplex, BGI Incorporated, Waltham, MA). Polytetrafluoroethylene (PTFE) filters with polymethylpentene (PMP) support rings (2.0 μm, 47 mm, SKC Inc., Fullerton, CA) were pre- and postweighed on a Cahn-28 electrobalance with a repeatability of ±1.0 μg after equilibrating for a minimum of 24 h in a humidity and temperature-controlled environment. Five field blanks were collected, by opening filters in the field site and resealing, which had an average mass difference of 0.4 ± 3.1 μg equivalent to less than 0.1% of average mass deposition of emissions samples and 0.2% of background samples. All sample filters, background, and emissions had a minimum of 109 μg of collected material, above the limit of detection for the method calculated at 9.3 μg or three times the standard deviation of the measurement of the field blanks. Quartz filters were collected and analyzed for EC and OC using a Sunset Laboratory OC/EC analyzer equipped for analysis using the thermal optical transmittance (TOT) method[27] with the temperature procedure reported in ref 28 and correcting for adsorption artifacts using a bottom filter.[29] A single set of filters were used for each cooking task.

Emission factors (EFs, g/kg Dry Fuel) for gases and $PM_{2.5}$ were determined using the carbon-balance method.[30] In brief, emission rates (ERs, mg/min or g/min) and EFs were determined by multiplying the carbon fraction of each pollutant emissions by the total emitted carbon during the burn. The carbon content of the fuel was taken to be 33.4% for buffalo dung and 45.4% for brushwood fuels based on Smith et al., (2000).[30] Carbon in ash was estimated as 2.9% and 80.9% of the mass of char for dung and brushwood, respectively.[30]

Climate impacts were estimated using 100 year global warming commitment potentials (GWC_{100}, see Supporting Information Table S1) as tCO_2e per kilogram of dry fuel incorporating the fraction of nonrenewable harvesting of fuels.[31] Species included in estimating climate impacts were CO_2, CO, EC, and OC emissions and SOA formed by reactions of volatile species in the atmosphere. In order to convert $PM_{2.5}$ emission factors from water boiling tests (WBTs)[30] into EC and OC, EF assumptions on the relationships between organic matter, organic carbon, elemental carbon, and $PM_{2.5}$ were utilized in a similar manner to that of Grieshop et al..[11] Elemental carbon was estimated as 21% of $PM_{2.5}$ mass, organic matter estimated as the remaining 79%, and organic carbon estimated as organic matter divided by 1.9 based on the values suggested for fireplace combustion of pine or oak in Roden et al.[32] The fraction of the fuel that is from nonrenewable biomass was assumed to be zero for dung and taken as 19% for wood fuels based on reported values for Haryana.[31] By assuming that organic matter is 1.9 times organic carbon, we may either over- or underestimate the contribution of organic carbon to GWP_{100}, as this relationship has been shown to vary between ∼1 and 3 depending on the source and age of the aerosol.[32] To estimate climate effects of SOA, SOA mass was estimated to be 1.64 (95% exact CI 1.52−1.76) times the mass of primary $PM_{2.5}$ based on recent U.S. Environmental Protection Agency Community Multiscale Air Quality (CMAQ) modeling estimates for these emission measurements in the Haryana region by Rooney et al.[33] SOA formation was estimated using CMAQ's secondary organic aerosol module, AERO6, incorporating select emission factors, gas-phase aging, semivolatile partitioning, as well as aerosol chemistry. SOA mass per primary $PM_{2.5}$ was based on a 4 km resolution run from September 7, 2015 to September 30, 2015 (UTC) at the SOMAARTH headquarters.[34] Additional information on equations used for climate impacts can be found in the Supporting Information with GWP values in Table S1.

Statistical analysis was performed with R version 3.3.1, and figures were produced in either Microsoft Excel 2010 or R version 3.3.1.

3. RESULTS AND DISCUSSION

Table 2 lists the geometric mean EFs for $PM_{2.5}$, EC, and OC in grams per kilogram of dry fuel and fuel consumption rates for the uncontrolled emissions tests in village kitchens ($n = 7$) and minimally controlled emissions tests in a village kitchen ($n = 12$) using mixed-fuels in the chulha. Use of mixed fuels is the typical practice in village homes in this region, but it complicates comparisons with controlled testing, as the majority of results from WBT tests typically use only one fuel type. Overall $PM_{2.5}$ emission rates from minimally controlled cooking were on the upper end of the range of uncontrolled emission factors, but no statistically significant differences ($p < 0.05$) were observed for $PM_{2.5}$, organic, or elemental carbon particulate EFs or fuel consumption rates (see Table S2) between the uncontrolled and minimally controlled testing).

While CO_2 emission factors in grams per kilogram of dry fuel were significantly higher in uncontrolled testing compared to minimally controlled cooking tests ($p < 0.01$), they were not significantly different on a carbon basis (g/kg carbon; see Table S2), due in part to differences in the ratio of dung/

Table 3. Differences in Particulate Emission Factors between Minimally Controlled Cooking, Uncontrolled Cooking, and Water Boiling Tests[a]

stove type	chulha	chulha	chulha	angithi/haro
fuel	cow dung	brushwood	mixed fuel	cow dung
minimally controlled cooking in Haryana (g $PM_{2.5}$/kg dry fuel)	18.2 ± 7.1 (n = 15)	6.3 ± 5.7 (n = 14)	12.3 ± 2.5 (n = 12)	32.3 ± 7.6 (n = 10)
uncontrolled cooking in homes (g $PM_{2.5}$/kg dry fuel)	N/A	N/A	8.7 ± 7.6 (n = 7)	N/A
India WBT from Smith et al.[30] (g TSP/kg dry fuel)	2.2 ± 0.3 (n = 3)	0.6 ± 0.1(n = 3)	N/A	0.5 ± 0.1 (n = 3)
factor difference between emission factors	8.3	10.5	0.7	64.6

[a] Notes: values as geometric mean ± standard deviation with n in parentheses. Mixed fuel tests were not conducted by Smith et al.[30] *N* was not sufficient in uncontrolled tests for single fuels to include.

brushwood in the mixed fuels. Because brushwood and dung have different carbon contents as a percent of dry weight, as the ratio of dung/brushwood changes, the amount of carbon per kilogram of dry fuel is also altered. Thus, relatively minor compositional changes in the ratio of dung: brushwood lead to differences in g/kg dry fuel not observed when analyzing on a per kilogram of carbon basis. Fuel consumption was also more highly variable in uncontrolled testing likely due to household size and specific cooking demands, which may also partially explain why the standard deviations for $PM_{2.5}$, EC, and OC emission factors and fuel consumption were higher in uncontrolled testing.

Differences between In-Home Measurements and WBT. Table 3 and Figure 1 show geometric mean $PM_{2.5}$ EFs during minimally controlled cooking tests in a village kitchen and uncontrolled cooking in homes. For comparison geometric mean EFs of total suspended particles (TSP) are shown for chulha and angithi/haro stoves using wood and cow dung measured during cold start WBTs in a simulated kitchen reported by Smith et al.[35] Use of mixed fuels or brushwood resulted in significantly lower $PM_{2.5}$ emissions compared to use of dung only. Average particulate emission factors from uncontrolled and minimally controlled cooking tests reported in Table 3 were similar to those reported by Weyant et al. for uncontrolled emissions measurements of chulhas in Maharashtra, India, using mixed dung and wood fuels during household cooking (n = 6), who reported $PM_{2.5}$ emission factors of 11.9 g/kg fuel, EC emissions of 0.9 g/kg, and OC emissions of 5.6 g/kg.[28] Similarly, average particulate emission factors and uncontrolled emissions measurements in Table 3 overlapped with those reported by Johnson et al. for 22 chulhas utilizing wood as their primary fuel during household cooking with an average $PM_{2.5}$ emission factor of 9.2 g/kg.[18]

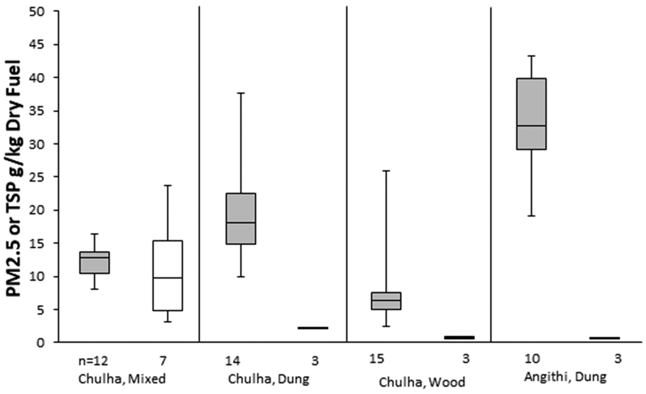

Figure 1. Particulate emission factors are $PM_{2.5}$ for minimally controlled cooking tests (gray), uncontrolled mixed-fuel cooking in homes (no color), and TSP for water boiling tests (black). Notes: Whiskers for WBT are not displayed as they are less than 0.1 gTSP/kg dry fuel.

$PM_{2.5}$ emissions reported in Table 3 from minimally controlled cooking using brushwood and cow dung in a chulha were approximately a factor of 9 higher than emissions of TSP per kilogram of dry fuel reported by Smith et al. during WBT.[35] Differences observed between minimally controlled testing and WBT are conservative because TSP includes both $PM_{2.5}$ and particles with larger aerodynamic diameters. Emissions of TSP have been previously reported to be 17% higher than emissions of $PM_{2.5}$ for firewood and 20% higher for dung cakes in traditional stoves.[36]

Pandey et al. also show an underestimation of $PM_{2.5}$ emission factors by WBT compared to prescribed cooking tests in a rural Indian home by a factor of 2−8.[16] Average reported in-field $PM_{2.5}$ EFs reported by Pandey et al. for fuel wood in Rajasthan and dung in Bihar were 10.5 (95% confidence interval 7.7−13.4) and 22.6 (14.9−32.9) g kg^{-1}; however, wood from Punjab had $PM_{2.5}$ EFs ranging from 3 to 15 g kg^{-1} (depending on combustion phase) and dung from Uttar Pradesh had $PM_{2.5}$ EFs ranging from 5 to 28 g kg^{-1}.[16] The low number of samples for each location ($n \leq 4$) precluded detecting any statistical differences between fuel wood types.[16] Weyant et al. also show substantial underestimations of emission factors for particulate matter in controlled laboratory tests of chulhas using WBT.[28] Similarly, both Johnson et al. and Roden et al. reported that laboratory WBTs were a factor of 2−4 lower than field measurements of traditional stoves for particulate matter emissions.[15,17] Although Table 3 shows larger differences in factors for the minimally controlled cooking than those of Johnson et al. and Roden et al., both report emission factors of >2 g $PM_{2.5}$/kg dry fuel for WBTs, which are larger than those reported by Smith et al. and would show differences of a similar magnitude (a factor of 2−4 lower) if compared to minimally controlled cooking. The magnitude of the differences between WBT and field tests, however, confirms that use of WBT tests are unlikely to lead to reliable estimates of actual emissions in the field for these stove and fuel combinations.

Emissions from the angithi/haro, which is typically used for slow simmering of milk or animal fodder using smoldering dung patties, were a factor 65 times higher in uncontrolled testing compared to WBTs, which suggests that the test protocol required to get this simmering stove to perform a WBT created highly uncharacteristic combustion conditions.

Table 4 shows a comparison of emissions from the wood-burning Philips stoves in the laboratory and from uncontrolled in-field testing. Emissions of $PM_{2.5}$ per kg dry fuel for the Philips stoves in the current study were substantially higher than those measured during laboratory tests of both wet and dry wood by Jetter et al.[37] Laboratory-based testing of

Table 4. Comparison of Average Emissions from the Philips Stove in Laboratory[37] and Uncontrolled Testing[a]

Philips stove	*n*	MCE	$PM_{2.5}$ EF	$PM_{2.5}$ ER	CO EF	CO ER
uncontrolled cooking (current study)	7	0.95	3.2	22.3	42.9	0.3
WBT simmer[20]	3	0.99/0.98	0.5/0.5	2.8/3.3	10.8/21.3	0.1/0.1
factor differences for simmering			6.6/6.4	7.9/6.7	4.0/2.0	4.5/2.0
p-values		0.015/0.033	0.019/0.019	0.031/0.034	0.024/0.081	0.008/0.038
WBT cold start[20]	3	0.99/0.98	0.5/1.4	7.3/19.3	10.4/25.8	0.2/0.4
factor differences for cold start			6.9/2.4	3.0/1.2	4.1/1.7	1.7/0.8
p-values		0.014/0.049	0.018/0.103	0.074/0.790	0.019/0.186	0.069/0.474
WBT hot start[20]	3	1.00/0.99	0.4/0.6	6.0/10.5	2.4/11.0	0.0/0.2
factor differences for hot start			9.0/5.2	3.7/2.1	17.9/3.9	6.7/1.5
p-values		0.007/0.013	0.016/0.027	0.057/0.243	0.007/0.021	0.004/0.134

[a] Note: Laboratory testing is listed as average values for triplicate (or more) measurements of dry wood/wet wood, with the wet wood value as the second entry. EFs are listed as arithmetic means in g/kg of dry fuel and ERs are listed in g/min for CO and mg/min for $PM_{2.5}$.

cookstoves utilizing the WBT employed three separate phases of testing; a cold start, a hot start, and a simmering phase (Water Boiling Test version 4.2.3). Emissions of $PM_{2.5}$ per kg of dry fuel were substantially higher in the current tests compared to the laboratory tests by factors of 2.4−9.0. Uncontrolled cooking tests and the wet wood WBTs had similar mean moisture contents (22.7% in the uncontrolled cooking versus 22.1 to 23% in the WBTs) although the variability in uncontrolled in-home testing was much larger as the standard error was 25.6% of the mean for uncontrolled cooking versus 3.1 to 12.3% for WBTs). ER and EF differences between uncontrolled cooking and laboratory testing were smallest for comparisons of the cold-start with wet wood (factors of 0.8−2.4), although significant differences in MCE were observed across all three phases of laboratory testing when comparing to the uncontrolled cooking via Welch's two-sided *t* tests ($p < 0.05$, shown in Figure S2).

Comparison to Laboratory Fuel-Burning. $PM_{2.5}$ emission factors from in-laboratory burning of fuel in noncooking settings by Saud et al. determined using a modified dilution sampler for dung cake and fuel-wood collected from Delhi, Uttar Pradesh, Punjab, Haryana, Uttarakhand, and Bihar of 16.3 ± 2.3 and 4.3 ± 1.1 g kg^{-1} for dung and fuel-wood, respectively,[38] were similar to uncontrolled field measurements in Haryana using the same fuels (18.2 ± 7.1 and 6.3 ± 5.7 g kg^{-1} for dung and fuel-wood, respectively) for the chulha, but were not reflective of the mixed fuel use typical of homes in the region and of emissions from Phillips and angithi stoves showing that fuel tests need to reflect the way in which the fuel is burned in real stoves.

Implications for Estimation of Environmental and Health Co-benefits. Although laboratory testing serves a critical function in evaluating stove design, the use of the results to draw wide conclusions about environmental and health co-benefits of cookstoves can provide misleading information on the relative benefits, as they do not reflect emissions from regular use in real homes. Minimally controlled cooking tests in our study villages resulted in emissions that were more reflective of actual usage in real homes than laboratory testing. Emission factors from minimally controlled cooking were close to those from uncontrolled tests in these villages and overlapping with those measured by Johnson et al.[18] Previous research has mostly indicated that emission factors for non-CO_2 species increase relative to CO_2 in cookstoves when fuel moisture is increased as a result of increased products of incomplete combustion,[39,40] although this effect is not universally true for all stove testing.[32,41] Selection of high and low moisture dung patties and/or brushwood for minimally controlled cooking tests, however, did not lead to significant difference in emissions rates, although verbal complaints about high moisture patties and compensatory behavior was expressed by the cook during cooking. Matching the moisture content of fuels for testing minimally controlled cooking tasks to those used on a regular basis for that cooking task would likely generate emissions estimates that more closely match those from uncontrolled cooking, and this shows promise for testing approaches that would provide more realistic estimates of climate and health co-benefits.

While the minimally controlled cooking tests in these villages in Haryana show promise in producing more representative emissions, there are a number of limitations. The sample size was limited in our study, a larger number of samples from more villages and a wider set of geographic locations in India and further afield would be required for wider applicability. In real homes, stove types, usage, and stove maintenance vary. Each of these parameters has significant impacts on combustion conditions, which in turn will change emissions. Use of minimally controlled cooking tasks does not inherently capture the wide range of stove types, maintenance, chimney heights, draft characteristics, and variations in operation and tending seen in homes. Further, consideration should also be made for the range of fuels used during different seasonal periods of the year and the degree of stove-stacking (use of multiple stoves and/or fuels in the same home for different cooking tasks) present in homes. Given the widespread presence of stove and fuel stacking in different parts of the world, estimating environmental and health implications of cookstoves by simply comparing results from water boiling tests from one stove to another assuming total replacement will lead to misplaced expectations for stove programs. In addition, incorporating stove stacking into current international emission guidelines for stoves, emission inventories and climate and health co-benefit estimates is a priority. Use of minimally controlled cooking tests allows for multiple stoves to be used according to user preferences and may generate more representative measurements of emissions in homes.

Geometric mean fuel consumption rates for the Philips stove during uncontrolled tests in these three villages in Haryana utilizing only brushwood were 6.9 ± 1.4 g/min, which were closer to those seen in the simmering phase of the water boiling test and were considerably lower than those seen in the cold start and hot start (5.7/6.4 for the simmering using dry/wet wood, 15.5/14.0 for the cold start, and 17.5/16.8 g/min for the hot start phase, respectively).[37] Thus, similar to cooking in Michoacan, Mexico,[14] the majority of cooking involved low-power tasks, and high-power tasks represent a small fraction of total stove usage. For the Phillips in this study, a burn cycle for dry brushwood where approximately 11% of the fuel was consumed in the cold-start phase and 89% in the simmering phase would achieve equivalent fuel-consumption rates to that seen during uncontrolled cooking, suggesting that task-based emission factors can provide more representative, realistic expectations of climate and health co-benefits for programs that provide alternative stoves.

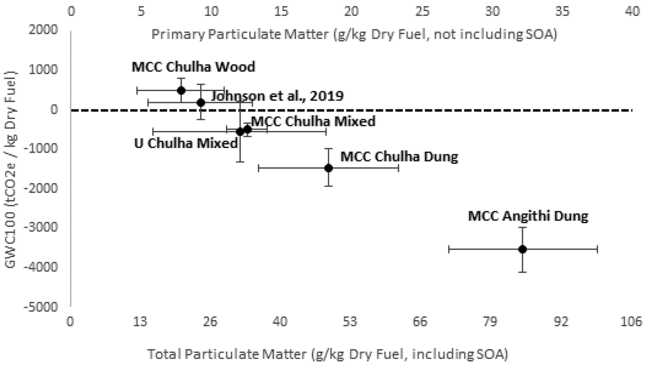

Figure 2. Primary and total particulate matter emissions and associated GWC_{100} for wood, dung, and mixed fuels in India. Particulate matter is expressed as both primary emissions only (top horizontal axis) and as total emissions including SOA mass (bottom horizontal axis). "U Chulha Mixed" refers to the uncontrolled testing, and "MCC" refer to minimally controlled cooking tests.

Climate Forcing Implications. Figure 2 shows primary and total particulate matter emissions and associated GWC_{100} for brushwood, dung, and mixed fuels in uncontrolled and minimally controlled cooking tests. For comparison, in-home emissions of 22 traditional Indian chulhas utilizing wood as their primary fuel reported by Johnson et al. are shown.[18] Two measurements for the minimally controlled cookstove angithi dung tests were not included in calculations due to damaged filters making EC/OC measurement impossible. The intercept at which stove emissions would be climate neutral was 9.8 ± 0.9 (95% approximate confidence interval 8.9−10.8) grams of $PM_{2.5}$ per kilogram of dry fuel measured as primary emissions directly from the stove, equivalent to 26 g/kg of $PM_{2.5}$ when including SOA formed by reaction in the atmosphere. Similarly, for GWC_{20}, cookstoves would be climate neutral at 8.8 ± 0.7 (95% approximate confidence interval 8.1−9.5) grams of primary $PM_{2.5}$ per kilogram of dry fuel (23 g/kg of $PM_{2.5}$ including SOA). In-field emission factors of traditional unvented biomass stoves from global cookstove inventories average around 7.4 g/kg dry fuel, with a typical range of 5 to 12 g/kg dry fuel,[42] which implies that traditional stoves using biomass fuels may be slightly warming or cooling with values close to neutral at both 20 and 100 year time horizons on a grams per kilogram of dry fuel basis. Estimation of overall climate impacts from dissemination of cookstoves with improved combustion would also need to account for any reductions in fuel consumption per cooking event, which would offset particulate emissions becoming more climate warming as OC is reduced. When a full suite of climate forcing species including OC and SOA and regional estimates of fraction of nonrenewable harvesting of biomass are incorporated, however, climate benefits based on these emissions measurements will be much smaller and less widespread and health benefits under similar ventilation conditions substantially larger than previously estimated.[5,10,11]

$PM_{2.5}$ emission factors and GWC_{100} are negatively correlated ($R^2 = 0.99$). Thus, as emissions of $PM_{2.5}$ decrease as a result of improvements in combustion, the stove emissions become more warming through reductions in the fraction of organic carbon emissions.

Climate forcing commitments from these stoves are dominated by contributions of short-lived climate forcing species OC and SOA (Figures S3 and S4). The intercept at which stove emissions would be climate neutral is largely dependent on current understanding and significant uncertainties surrounding the estimation of climate forcing for these species. While we use current understanding of the central estimates of the forcing of these species, improved understanding will likely improve estimates of the intercept and understanding of climate and health co-benefits from these stoves. Error bars for particulate matter and GWC_{100} shown in Figure 2 are the 95% confidence intervals for each sample but do not represent overall uncertainties in climate forcing estimates, as they do not include uncertainties associated with GWPs, fNRB, and the carbon content of the fuel. For climate forcing estimates, SOA was assumed to be OC and 164% of the primary $PM_{2.5}$ emissions' mass, based on SOA concentrations predicted by CMAQ simulations for September 2015 at SOMAARTH headquarters.[33] Estimates of warming or cooling from traditional stoves are not sensitive to the ratio of $PM_{2.5}$ to organic carbon, as previous uncontrolled measurements have found robust linear relationships between ratios of $PM_{2.5}$/OC, with slopes of 1.29 to 1.35 (R^2 between 0.74 and 0.99) for a variety of biomass stoves across 174 measurements representing a wide range of fuel types, stove types, flues, altitudes, and cooking locations.[43] Although there are issues with time horizons when using GWP to compare the effects of short-lived and long-lived atmospheric species on climate,[44−46] Figures S3 and S4 show contributions of each species to the GWC_{20} and GWC_{100}, respectively, demonstrating how warming commitment estimates from these stoves are dominated by contributions of short-lived climate forcing species OC and SOA. Inclusion of OC and SOA in methods to estimate carbon offsets is an important priority in estimating actual impacts of cookstoves on climate.

The fraction of nonrenewable biomass harvested and secondary organic aerosol generated in the atmosphere differs between agroclimatic regions, and thus, the relative impacts of stoves will vary across regions. Current estimates of the fraction of nonrenewable wood fuels have large geographic variations. For example in 2009, while India had seen a net gain in afforestation in recent years, 23.7% of India's wood fuel (range 17.3−32.4%) and 29.6% of Asia and Oceania's wood fuel were harvested unsustainably.[31] The intercept at which stove emissions are climate neutral is relatively insensitive to changes in nonrenewable biomass harvested largely because of the climate forcing contributions of SOA and OC and also because dung fuels are renewable from a harvesting perspective. Using Asia and Oceania's average fraction of nonrenewable biomass of 29.6% would raise the $PM_{2.5}$ intercept for climate-neutral emissions to 10.4 g/kg of dry fuel for a 100 year horizon and 8.9 g/kg for a 20 year horizon. Using this cutoff, some uncontrolled field tests of biomass burning stoves in Nepal, Cambodia, and Tibet would imply a net cooling.[28,47−49] The fraction of nonrenewable harvesting estimate of 29% for the region and 19% for Haryana result in only a modest difference in the intercept at which stove emissions are climate neutral. Assuming that SOA formation processes are similar between different regions, large fractions of global emissions would be close to climate neutral using these estimates. Clearly, however, in wood fuel harvesting hotspots, the intercept where primary emissions are warming would be higher, which highlights that the climate implications of stoves will depend on the specific communities in which the stoves are distributed. Similarly, where households use different mixtures of fuels, the intercept for climate neutral emissions will also vary from those presented here based on the specific fuel mixture present and fraction of renewable harvesting of each fuel. In spite of these limitations, these findings show that when SOA and other climate forcing particulate species are included in estimates, along with regional estimates of nonrenewable harvesting, emissions from traditional stoves using biomass fuels are likely to be much less climate warming than previously thought and some may be climate cooling. Although beyond the scope of the current paper, this has large implications both for methods to estimate carbon offsets and for the viability of climate offsets from solid biomass cookstoves, as improved combustion will lead to less $PM_{2.5}$ emissions primarily as a result of reduced OC. While these results cannot capture the full range of emissions, SOA formation conditions, and harvesting from different agroclimatic regions, these findings highlight the importance of calculating global warming from cookstoves including a full suite of climate forcing species including SOA formed after emission into the atmosphere and also including realistic estimates of the fraction of nonrenewable harvesting of biomass.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c05143.

> Picture of the Haryana study site; GWP values; calculation of GWC_{100} EFs; calculation of GWC_{20} EFs; comparison of select MCE values; additional mean EFs and ERs; stacked bar chart of the contribution of each species to total GWC_{20}; stacked bar chart of the contribution of each species to total GWC_{100} (PDF)

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Notes

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