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A laboratory comparison of emission factors, number size distributions and morphology of ultrafine particles from eleven different household cookstove-fuel systems

Guofeng SHEN[£], Chethan K. Gaddam[†], Seth M. Ebersviller^{\$}, Randy L. Vander Wal[†], Craig Williams^φ, Jerroll W. Faircloth^{||}, James J. Jetter^{†,*}, and Michael D. Hays[‡]

[£].Oak Ridge Institute for Science and Education (ORISE) Postdoctoral Fellow at U.S. Environmental Protection Agency, Office of Research and Development, 109 T.W. Alexander Drive, Research Triangle Park, NC 27709, USA

^{\$}.University of Findlay, 1000 North Main Street, Findlay, Ohio 45840, USA

[†].John and Willie Leone Family Department of Energy and Mineral Engineering and the EMS Energy Institute, Penn State University, University Park, PA 16802, USA

^φ.CSS-Dynamac Inc., 1910 Sedwick Road, Durham, NC 27713, USA

^{||}.Jacobs Technology Inc., 600 William Northern Boulevard, Tullahoma, TN 37388, USA

[‡].U.S. Environmental Protection Agency, Office of Research and Development, 109 T.W. Alexander Drive, Research Triangle Park, NC 27709, USA

Abstract

Ultrafine particle (UFP) emissions and particle number size distributions (PNSD) are critical in the evaluation of air pollution impacts; however, data on UFP number emissions from cookstoves, which are a major source of many pollutants, are limited. In this study, eleven fuel-stove combinations covering a variety of fuels and different stoves are investigated for UFP emissions and PNSD. Combustion of LPG and alcohol ($\sim 10^{11}$ particles per useful energy delivered, particles/MJ_d), and kerosene ($\sim 10^{13}$ particles/MJ_d) produced emissions that were lower by 2–3 orders of magnitude than solid fuels (10^{14} – 10^{15} particles/MJ_d). Three different PNSD types; unimodal distributions with peaks ~ 30 – 40 nm, unimodal distributions with peaks < 30 nm, and bimodal distributions; were observed as the result of both fuel and stove effects. The fractions of particles smaller than 30 nm (F_{30}) varied among the tested systems, ranging from 13% to 88%. The burning of LPG and alcohol had the lowest PM_{2.5} mass emissions, UFP number emissions and F_{30} (13–21% for LPG and 35–41% for alcohol). Emissions of PM_{2.5} and UFP from kerosene were also low compared with solid fuel burning, but had a relatively high F_{30} value of approximately 73–80%.

*Corresponding author: James J. Jetter, U.S. Environmental Protection Agency, Tel: 919-541-4830; Fax: 919-541-2157; Jetter.jim@epa.gov.

The authors declare no competing financial interest.

Supporting information

Supporting materials associated with the manuscript are available free of charge via the internet:

Figures comparing UFP number emissions, PNSDs, GMD values, and F_{30} values; TEM images; and detailed information on cookstoves and fuels

TOC

1. Introduction

Solid fuels for household cooking and heating are used by nearly 3 billion people globally. Exposure to household air pollution (HAP) due to solid fuel combustion poses a serious environmental health risk, causing ~4.0 million premature deaths annually.^{1,2} Domestic fuel burning is also a concern for climate.^{3, 4} Estimates show that residential fuel combustion emissions exert a global annual mean direct radiative effect between -66 and $+21$ mW/m², and a global annual mean aerosol indirect effect between -52 and -16 mW/m².³

Domestic heating and cooking emissions comprise multiple products of incomplete combustion (PIC), such as CO, PM_{2.5} (particulate matter with aerodynamic diameter 2.5 μ m), black carbon, and ultrafine particles (UFPs, particles with diameter less than 100 nm).^{2, 5-7} Residential stoves are a major anthropogenic source of primary particle number emissions, contributing 17% to the global average, and are principal sources in Africa and many developing nations like China and India.⁸ Particle number size distributions (PNSDs) are important input to climate models, and particle size is also a key factor for determining how deeply particles penetrate the lung and consequently influence health. In fact, some studies show negative health effects to be more closely associated with particle number concentration (PNC) than with PM mass concentration.⁹⁻¹¹ Moreover, epidemiological evidence indicates a strong relationship between UFP exposure and asthma and cardiovascular disease.^{9, 12-16} Thus, there is currently substantial interest in the UFP fraction.

However, studies on UFP emissions and number size distributions from residential solid fuel combustion are relatively limited. Most available studies of this sort examine wood (logs and pellets) burning in heaters or boilers.¹⁷⁻²² Residential wood heating stoves are usually sealed and have air controls. Particles emitted from wood heating stoves were found to have compact structures with fractal-like dimensions near three. Operating conditions and fuel and stove types, can also strongly affect UFP emissions and size distributions from these stoves.²² This data gap has contributed to uncertainties in air quality modeling studies that consider particle number emissions.^{8, 23} Available studies do show that particle size distributions and emissions from cookstoves are significantly affected by fuel and stove type

and burning conditions.^{24–29} For example, Just *et al.*²⁴ found that particle emissions from wood burning in improved rocket and gasifier stoves were 40–70% lower than those from a three-stone wood fire, and the number mode diameter was also smaller (24, 35 and 61 nm for the gasifier, improved rocket and three-stone stoves, respectively). L'Orange *et al.*²⁸ showed that particle size distributions are not strongly dependent on stove construction material or stove wall temperature, but are affected by the cooking pot temperature possibly due to the physical interaction between the emission plume and the pot.

A multitude of fuels and stove types are used worldwide. The main objective of this study is to quantify and compare UFP emissions and PNSDs for a variety of household fuel and cookstove types. Fuels tested include gas (liquefied petroleum gas, LPG), liquid (alcohol and kerosene), charcoal, wood pellets and wood sticks (at high- and low-moisture levels). Wood-fueled stove technologies included a 3-stone fire, a natural-draft stove (NDS) and a forced-draft stove (FDS). This study provides information on UFP emissions and number size distributions in cookstove emissions that are critical for human health and regional climate change studies.

2. Methods

Cookstove testing system, fuel and stoves.

Combustion emission tests were performed at the U.S. Environmental Protection Agency (EPA), Research Triangle Park, NC, Cookstove Test Facility which is designed for testing thermal efficiency and pollutant emissions for a wide variety of fuels and stoves with or without chimneys. The Water Boiling Test (WBT) protocol (Version 4.2.2), including a cold-start high-power test phase, a hot-start high-power phase, and a low-power phase, was used for evaluating cookstove performance. Laboratory testing that follows the international WBT protocol^{30,31} is not aimed at duplicating all field conditions, but instead provides the controlled conditions necessary for making meaningful comparisons. Cookstove emissions were collected with a stainless steel hood connected to a dilution tunnel, from which gaseous and PM pollutants were sampled. Emissions were quantified using the total-capture constant-flow method, similar to EPA Method 5G,³² in which the dilution ratio varies with the emission rate. A second stage of dilution (~1:10) provided by a dilution sampling system (DSS)^{33,34} was used for particle size measurements. Concentrations measured in the DSS were used to generate PNSD figures with $dN/d\log(D_p)$ profiles.

UFPs were measured from the DSS using a Scanning Mobility Particle Sizer (SMPS) Model 3936 consisting of a Model 3080 Electrostatic Classifier with a Model 3081 Long Differential Mobility Analyzer (L-DMA), and a Model 3010S Condensation Particle Counter (CPC), (TSI, Inc.; Shoreview, MN, USA). A particle mobility diameter range of 14.6–661 nm was measured. The SMPS provided a full scan every 150 s (up scan 120 s and down scan 30 s), and some short-duration emission events may have been missed. Carbon monoxide (CO) and carbon dioxide (CO₂) were monitored continuously with nondispersive infrared analyzers, and methane (CH₄) and total hydrocarbons (THC) were monitored with flame ionization detector analyzers (Models 200, 300-HFID and 300M-HFID, California Analytical; Orange, CA). Data were recorded automatically every 10 seconds. PM_{2.5} was also sampled isokinetically on polytetrafluoroethylene (PTFE) membrane and quartz-fiber

filters positioned downstream of PM_{2.5} cyclones (URG, Chapel Hill, NC). PTFE filters were used to obtain PM_{2.5} mass emissions through gravimetric analysis with a Model MC5 microbalance (Sartorius, Gottingen, Germany). Background air was measured with all monitoring instruments for at least 10 minutes before and after each test. Background concentration of PM_{2.5} mass was measured by sampling background air for 30 minutes without fuel burning. More detailed information about the facility, sampling and quality control can be found in Jetter et al.³⁰ A total of eleven fuel-stove combinations, with three test replicates, were evaluated (Table 1). Detailed information on fuels and stoves is provided in Supporting Information. Fuel properties (proximate analysis for solid fuels, elemental analysis and lower heating values for all fuels tested) are listed in Table S1. Fuel energy was calculated from fuel mass measured before and after each testing phase, and the values were used in the calculation of emission factors.

High-Resolution Transmission Electron Microscopy (HR-TEM).

Particle morphology was studied using transmission electron microscopy to produce bright field images using a JEOL EM-2010F instrument with field-emission electron source.^{35,36} After samples were collected on quartz filters, a punch was removed and extracted ultrasonically in ethanol (15 min). The extract was then placed on a TEM grid by drop deposition using a glass pipette. Deposited samples were allowed to dry completely prior to being loaded into the instrument. This process ensures that the aggregate particles are dislodged from the filter without changing aggregate morphology.^{35,36} Samples captured directly on TEM grids are clearly preferred, but there is considerable precedence for filter collection followed by dispersal upon a TEM grid for observation. Adherence to sonication procedures and detailed examination of particle-particle connections gives confidence to observations of individual aggregates. Although aggregates are observed as a two-dimensional projection, imaging a high number of aggregates sufficiently randomizes for this factor to provide a statistical representation. Depending on the soot samples and analysis, the applied magnifications varied between 40k and 500k.

Data analysis.

UFP number emission factors (EFs) are quantified from measured UFP number concentrations, calculated total volume of emission exhaust, mass of fuel consumed, and useful cooking energy delivered. EFs are presented as total UFP number per fuel mass (particles/kg), fuel energy (particles/MJ) and useful energy delivered (particles/MJ_d). Useful energy delivered is the energy absorbed by the water in the pot including the energy that raises water temperature and that evaporates water from the pot during testing. Stove thermal efficiency is calculated as the ratio of useful energy delivered to the total energy of fuel consumed.

3. Results and Discussion

3.1 Emission factors

Difference among testing phases—UFP emission factors on the basis of fuel mass, fuel energy, useful energy delivered, and time (emission rate) for the tested fuel/stove groups during the three testing phases are summarized in Table 2. For solid-fuel stoves, mean values

for UFP number emissions tend to be higher in the cold-start, but there is no statistical difference between the cold-start and hot-start high-power phases ($p > 0.05$, Figure S1). This is also the case for EFs measured for most fuel/stove combinations at high- and low-power phases, with the exceptions of [Kero-WS] and [Char-NDS] ($p < 0.05$, Figure S2). For [Kero-WS], the low-power phase emits ~2.5 times more UFPs than the high-power phase. Cooking power for the [Kero-WS] is controlled by adjusting the height of the wick. At low-power, combustion is improved, as indicated by the higher MCE, with a resulting shift of the particle size distribution toward the ultrafine region and with a greater number of UFP emitted. In contrast, [Char-NDS] emissions are about one order of magnitude lower in the low-power phase than in the high-power phase, due to combustion of volatiles during high-power and combustion of mainly elemental carbon during low-power.

Emission rates (Figure S3) show no significant differences among the three testing phases for each fuel-stove; except for [Char-NDS], for which the UFP emission rates during the cold- and hot-start high-power phases were about two orders of magnitude higher than those during the low-power phase. Generally, the UFP emission rates are $\sim 10^{11}$ particles/hour for [LPG-OBS] and [Alc-EBS], $\sim 10^{14}$ particles/hour for [Kero-WS] and [Pell-NDS], and $\sim 10^{15}$ particles/hour for [Char-NDS], [RiH-FDS] and wood combustion. [LMW-3SF] shows the highest emission rate at nearly 10^{16} particles/hour.

Difference among fuel/stove combinations—UFP EFs on the basis of fuel mass (Table 2) are $\sim 10^{12}$ particles/kg for [LPG-OBS] and [Alc-EBS], and $\sim 10^{15}$ particles/kg for the other fuel/stove systems. On the basis of fuel energy, UFP EFs range from 1.0×10^{11} particles/MJ to 4.3×10^{14} particles/MJ. Significant differences ($p < 0.001$) are observed among the tested fuel/stove groups. [LPG-OBS] and [Alc-EBS] ($\sim 10^{10}$ – 10^{11} particles/MJ) had the lowest emissions, followed by [Kero-WS] ($\sim 10^{13}$ particles/MJ). UFP emissions from the combustion of solid fuels (charcoal, rice hulls, wood and pellets) ($\sim 10^{14}$ particles/MJ) are about 1–3 orders of magnitude greater than the emissions for gas or liquid fuels. Among the solid fuels tested, [Pell-NDS] and [Char-NDS] emit less compared with [RiH-FDS] and wood combustion. UFP emission from [RiH-FDS] is even higher than that from the [LMW-3SF] on a fuel energy basis. The difference in UFP emissions between high- and low-moisture wood burned in the same stove ([LMW-NDS1] vs [HMW-NDS1]) is significant, with much higher emissions for the former. UFP emissions from burning low-moisture wood in the forced-draft stove ([LMW-FDS] and the two natural draft stoves ([LMW-NDS1] and [LMW-NDS2]) show no significant difference.

Thermal efficiency (Table 2) is generally greater for non-solid fuels ([LPG-OBS] 55–61%, [Alc-EBS] 54–58% and [Kero-WS] 40–43%), followed by processed solid fuels ([Pell-NDS] 48–58%, [RiH-FDS] 40–54%, [Char-NDS] 30–42%), compared with wood combustion in the FDS (36–41%) and NDS (24–37%), with [LMW-3SF] as the lowest (~15%). Correspondingly, emissions of UFPs per useful energy delivered are lowest for [LPG-OBS] and [Alc-EBS] ($\sim 10^{11}$ particles/MJ_d), and [Kero-WS] ($\sim 10^{13}$ particles/MJ_d), followed by relatively low emissions from [Pell-NDS] and [Char-NDS] ($\sim 10^{14}$ particles/MJ_d). Though [RiH-FDS] has similar or slightly higher UFP emissions on a fuel energy basis compared with wood combustion, the useful energy based emissions for [RiH-FDS] are lower than

those from wood combustion, primarily due to higher thermal efficiency. [LMW-3SF] has the highest UFP emissions on a useful energy delivered basis.

For the UFP number emissions during the low-power phase (Table 2), the differences among tested fuel-stove groups are significant. The [LPG-OBS] and [Alc-EBS] ($\sim 10^{10}$ particles/MJ) produce the lowest emissions, followed by [Char-NDS] and [Kero-WS] ($\sim 10^{13}$ particles/MJ). The [LMW-3SF] produces the highest emissions. The remaining fuel-stove types generate emissions of $\sim 10^{14}$ particles/MJ.

Emissions trends with respect to fuel type are consistent with fuel molecular structure. LPG with its high content of light hydrocarbons and lack of aromatics inherently produces less soot. Oxygenated liquid fuels generally burn cleaner than hydrocarbon analogues given the embedded oxygen – accounting for the decreased mass and number concentrations from the alcohol relative to the kerosene. In the simple kerosene-fueled cookstove however, this advantage is partially mitigated by the use of a wick rather than spray injectors as used in engines. Still, oxygenated fuels will produce fewer emissions given that the oxygen content of fuel enables more complete combustion, lowering the fuel-to-air equivalence ratio. The physical state of the fuel can also influence emissions. Liquid fuels vaporize quickly and hence are easier to mix with air, compared to solid fuels. This benefit was found to apply to biofuels, e.g. biodiesel.³⁵ Comparatively raw or pelletized wood with lignin content contains the aromatic precursor compounds that accelerate soot formation. Beyond these general observations it is difficult to relate the measured mass or number emissions to a particular fuel given the interplay among fuel components, fuel properties, stove types, and convection design.

3.2 Particle Number Size Distributions (PNSDs)

PNSD replicates.—Although UFP number emissions varied in repeat trials, with coefficients of variation (COVs) ranging from 5–60% for solid fuels, 10–80% for liquid kerosene and alcohol, and 80–140% for LPG, the PNSDs are generally similar among repeated tests (Figure S4). Lower particle counts measured with the SMPS for [LPG-OBS] and [Alc-EBS] likely contributed to the relatively higher standard deviations observed. PNSDs for the cold- and hot-start high-power phases (Figure S5), and between high-power and low-power phases are similar for most tested combinations, with the exception of [Kero-WS] and [Char-NDS]. For [Kero-WS], a peak is observed at ~ 18 nm with a second abundance of particles at 80–90 nm during the high-power phase, while the low-power phase produces a single dominant peak at ~ 23 nm (Figure S6). For [Char-NDS], the peak is around 30–35 nm during the high-power phases, while the low-power phase produces a higher number of smaller particles (Figure S6). As mentioned above, for [Char-NDS], combustion of volatiles occurs during high-power, while during low-power, mainly combustion of elemental carbon occurs, resulting in low emissions of total UFPs and a shift in the PNSD toward the ultrafine region. For [Kero-WS], improved combustion in the low power phase resulted in a favorable formation of finer particles.

PNSDs for tested fuel-stove combinations—Figure 1 compares the PNSDs for tested fuel-stove combinations during the cold-start high-power phase. The profiles for [Alc-EBS]

and [LPG-OBS] were not as smooth as the others, mainly due to low particle counts measured with the SMPS. Generally, these PNSDs can be classified into three distinct groups: (A) a unimodal distribution with a ~30–40 nm nucleation mode peak ([Char-NDS], [LMW-NDS1], [LMW-3SF], and [HMW-NDS1]); (B) a unimodal nuclei mode distribution with maxima < 20 nm ([Pell-NDS], [Alc-EBS], [Kero-WS], [RiH-FDS] and [LMW-FDS]); and (C) a bimodal distribution with a major nucleation mode peak at ~20 nm and a smaller second peak at ~80–100 nm ([LPG-OBS] and [LMW-NDS2]).

PNSD geometric mean diameter (GMD)—Figure S7 shows the phase-based, average GMD for the PNSDs for the present study. [LPG-OBS] produces the highest GMD (~70 nm). For [Alc-EBS] and wood combustion, the GMD is generally ~40–50 nm with the exception of [LMW-FDS] which is ~24 nm. Relatively low GMD values are also observed in emissions from [Kero-WS] and [RiH-FDS] (~26–28 nm). GMD values and PNSD profiles are affected by a variety of factors including, but not limited to, combustion conditions, fuel properties, stove design, dilution and cooling, particle concentrations, temperature, and relative humidity.^{19, 25–29, 36–38} For example, the low GMD values observed for [LMW-FDS] compared with [LMW-NDS1] and [LMW-NDS2] are likely due to relatively efficient, high-temperature burning that favors the formation of smaller particles at lower concentrations thus reducing condensation and coagulation which contribute to growth. More independent spheres were observed for the wood burning in a forced-draft gasifier stove in comparison with emissions from wood combustion in natural-draft rocket stoves and a three-stone fire.²⁴ The [LPG-OBS] and [Alc-EBS] produced low particle concentrations that reduced the SMPS signal thereby increasing uncertainty in the GMD determination. Further testing may be performed in the future in an effort to determine the variables that contributed most substantially to these PNSDs.

The GMD values for high- and low-power phase emissions are generally similar, except the notable difference for [Pell-NDS], for which the GMD values are 41 and 59 nm in cold- and hot-start high power phases, respectively, but 26 nm in the low-power phase. As shown in Figure S6, a small peak in the 120–150 nm size range is more visible during the high-power hot start phase than during the other phases. The size distribution in the range of 40–660 nm appears very similar between the cold start and simmer phases, but the peak in the finest size range is higher during the low-power simmer phase.

The geometric standard deviation (GSD) values for all tested combinations are 1.6–2.4, 1.5–2.6 and 1.3–2.4 nm during the three phases, suggesting a polydisperse distribution mode. Note that the actual GMDs may be lower than the values reported here due to a truncated distribution in the measured size range in the SMPS.

GMD for volume (mass) distribution—The GMD values for particle volume (or particle mass) are estimated assuming sphericity and a density of 1.8 g/cm³.²² GMD values are generally lower for [Kero-WS] (62–177 nm in different testing phases) and [Char-NDS] (66–157 nm), than [HMW-NDS1] (180–220 nm). Greater particle volume GMD values are observed in emissions from [Pell-NDS] (334–388 nm) and [RiH-FDS] (350–396 nm), followed by [LMW-NDS2] (315–348 nm), [LMW-FDS] (315–380 nm), [LPG-OBS] (237–268 nm) and [Alc-EBS] (220–290 nm).

Fractions of particles smaller than 30 nm—We enlist the approach of Just et al., (2013) to determine the particle number fraction with $d_a \leq 30$ nm (F_{30}) due to their potential adverse human health impacts.^{24, 39} Figure S8 compares the F_{30} values for the eleven fuel-stove combinations. Relatively high F_{30} values are observed for [LMW-FDS] (~80%), [Kero-WS] (73–80%) and [RiH-FDS] (62–72%). The F_{30} values for the LMW—representing two natural-draft stoves and a 3-stone fire—are 40–50%. Lower F_{30} values are observed for [LPG-OBS] (13–21%), [HMW-NDS1] (23–30%) and [Alc-EBS] (35–40%). For [Char-NDS], F_{30} values are roughly 40–50% during the high-power phase, increasing to 88% during the low-power phase, due to a change in PNSD between these two phases, as mentioned above.

Differences between startup and steady state operations.—Emissions often vary over the combustion cycle, e.g. ignition, fuel addition, and burnout phases. Changes in PNSDs over time are shown in Figure S9 for select tests. Presently, the PNSDs obtained during startup and at steady-state are compared. The startup is defined here as the first 2.5 min (1 scan), and the steady state is from 10 min after ignition to the test end. Results are presented in Figure S10 for all fuel-stove combinations and in Figure 2 for select combinations. Generally, the startup produces larger particles than the steady state for [LMW-3SF], [LMW-NDS1], [LMW-NDS2], [HMW-NDS1], [Char-NDS] and [RiH-FDS]. However, the normalized number size distributions are similar between startup and steady state for [Pell-NDS], [LMW-FDS], [LPG-OBS], [Alc-EBS] and [Kero-WS]. Note that particle sizes change rapidly over time during the burning process, and the 150 s scan time may be unable to capture rapid changes accurately. The use of instrumentation with faster high-resolution scans is important for future studies.

3.3 Morphology from HR-TEM

Figure 3 shows images of soot collected from the [LMW-NDS2] at the indicated varied magnification. Illustrative TEM images from [LMW-3SF], [Char-NDS], [LMW-NDS1], [HMW-NDS1], [Pell-NDS] and [Kero-WS] are given in Figure S11. For the others, no sample was observed on the TEM grid. The combination of low elemental carbon emission and potential for dissolution of organic content with related unknown extractability from the filter inhibited TEM observations from these fuel/stove combinations. Aggregates comprise semi-spherical primary particles with internal lamellae structure evident. The lamellae are observed as the dark lines in the image, arising from the phase contrast between the directly transmitted and diffracted electron beam in the microscope. The web structure in the background of the low magnification images is the lacy support of the TEM grid. Soot aggregates from the wood combustion were compact, tightly clustered with fractal dimension larger than 2.5. Primary particles within the aggregates spanned a broad size range, from 5 nm to nearly 50 nm. Soot derived from LMW combustion exhibited a measureable increase in nanostructure within the primary particles, which is consistent with the measured PNSD and may reflect high flame temperature associated with the LMW combustion. While the low magnification images show the aggregate morphology, the high magnification images show the fused connectivity between primary particles – evident at the “neck” between primary particles where the lamellae are observed to merge or bridge between the individual spherules. While some variations were observed across fuel-stove

combinations, similar soot structures were also observed. The merged nature of primary particles is clear from these images, and these images are consistent with soot formed from combustion processes, and not at low temperature or having formed as a secondary aerosol.

By the sample preparation method, dispersal of aggregates is not guaranteed, but overlapping aggregates can generally be discerned. The purpose here is not to correlate TEM aggregate size with the aerodynamic (mobility) size, for which real-time size-separated collection is necessary. The utility of the TEM images here is to provide physical reference to the mobility size as reported from the SMPS instrumentation. As illustration, similar size distributions have been reported from jet aircraft engines (running nominally on kerosene).⁴¹ In reference to the TEM images, a 2-dimensional projection in Figure 3 shows the individual spherules connected to form an aggregate, and across the fuel-stove combinations, the soot emissions are in this form. Typical of combustion-formed soot, varied primary particle sizes and numbers within an aggregate are observed. Given the primary particle size and that aggregates contain several to dozens of such particles illustrates that the aerodynamic or mobility size, commonly interpreted as a “diameter,” is a very different physical size metric. Any given mobility size will not correspond to a unique aggregate size by TEM, nor mass, because any number of primary particles and assemblies can match a particular aerodynamic size. Other studies explicitly investigate this mass-mobility relation.⁴² Of particular relevance for cookstoves and their use is that the respirable nature of emitted soot may be closely tied to aerodynamic size, which is one reason for the many literature studies reporting this metric; however, the climate impact related to optical properties and surface area for water condensation/ice nucleation is dependent on aggregate fractal morphology and total surface area, i.e. actual physical size and shape.

3.4 Comparison with previous literature results

Studies of UFP emissions from residential cookstoves are relatively scant compared with other combustion emissions sources, such as gasoline- and diesel-fueled vehicles. Some previous studies have reported UFP emissions from wood combustion in heating stoves or boilers, and more recently there are some reports about UFP emissions from wood burning in cookstoves.^{24–25, 27–29} UFP number emission factors for wood or pellet combustion in heating stoves or boilers are usually $\sim 10^{14}$ – 10^{15} particles/kg, or $\sim 10^{12}$ – 10^{13} particles/MJ on the basis of fuel energy,^{17–21, 43} which are comparable or slightly lower than results shown here for wood or pellets in cookstoves.

The three general types of PNSDs observed presently are also reported in studies of wood combustion emissions, regardless of the varied measurements and fuel-stove types used across the comparisons. A unimodal size distribution with a peak around 60–100 nm, or the highest abundance in the finest particle size range is often reported,^{24,25,29} but a bimodal distribution with peaks at ~ 120 nm and ~ 50 nm was also reported for wood combustion in a traditional Indian *Chulha* stove.²⁷ In another laboratory study, the peak modes of particles from wood combustion in a natural-draft stove and gasifier stove were 35 and 24 nm,²⁴ respectively, similar to our results for natural- and forced-draft wood-fueled stoves.

The PNSD GMD values for wood and pellet combustion emissions in heating stoves or boilers are typically 50–120 nm,^{17–21, 43} which are greater than the GMD values for wood burning cookstove emissions in the present study. In a field study in China, the PNSD GMD values measured using an electrical low-pressure impactor (ELPI, Dekati Ltd., Finland) were much higher showing 150 nm for firewood and 210 nm for wood branches.³⁷ In addition to the different instruments and dilution systems used, stove configurations are also an important factor. For example, a typical Chinese cookstove has a large combustion chamber and may be configured with an exhaust path through a heating bed (*Kang*) that often provides a long residence time and cooling promoting particle growth through coagulation and condensation.

In regard to temporal change of total UFP emissions and PNSDs, a previous study on wood combustion in masonry heaters and sauna stoves reported particle number emissions decreased from ignition to burn out.⁴³ Burning split beech wood logs (15–18% water content on a dry basis) in a residential cookstove,²² the PNC and GMD were highest at start-up (phases were defined according to the CO concentration, and the startup phase was from the ignition to a state with low and rather constant CO level). The PNC shifted to lower values in the intermediate and burn-out phases, changing with the combustion air supply. A similar temporal change in particle size was also observed in the present study for wood combustion in three-stone fire and natural draft cookstoves, but for the wood combustion in the forced-draft stove and for some non-solid fuels, there is only a minor difference in the startup and steady state PNSDs. Note again that startup transients may be missed due to the 150 s scan time limitation of the present study. Higher time-resolution scans are recommended for future studies.

3.5 Cookstove evaluation metrics

PM_{2.5} and CO mass emissions and thermal efficiency are used as metrics for most cookstove evaluations.⁴⁴ The [LMW-3SF] was among the fuel/stove combinations with the highest UFP number and PM_{2.5} mass emissions (Figure 4). The figure also indicates that combustion of gas and liquid fuels produces relatively lower UFP numbers and PM_{2.5} mass. [Pell-NDS] and [Char-NDS] emit less UFPs and PM_{2.5} compared with wood combustion in the natural-draft stoves. [HMW-NDS1] had higher UFP and PM_{2.5} emissions than [LMW-NDS1]. [LMW-FDS] had significantly lower PM_{2.5} mass emissions compared with the natural-draft stoves, but UFP number emissions were not significantly different between these two types of stoves. Generally, PM_{2.5} mass emission and UFP number emission are positively correlated ($r=0.723$ and $p<0.001$ for EFs on the basis of fuel energy, and combined from three phases, Spearman test), but PM_{2.5} mass emissions can only explain 33% of the variation in UFP number emissions ($EF_{UFP}=1.1\times10^{12}\cdot EF_{PM_{2.5}}+1.4\times10^{14}$, $R^2=0.328$). It is noted that [LMW-FDS] had significantly lower PM_{2.5} mass emissions (70–90% lower) than emissions from natural-draft cookstoves and the 3-stone fire, but a significant UFP emission reduction was only found when comparing to the 3-stone fire. These phenomena are interesting, but further investigation is needed. Presently, only one forced-draft stove fueled with low-moisture wood was included. Forced-draft is one of the technologies considered to improve combustion and stove performance, and consequently to reduce emissions and protect health.⁴⁵

In Figure 5, $PM_{2.5}$ mass, UFP number emissions, and F_{30} value are compared among different fuel-stove groups. [LPG-OBS] and [Alc-EBS] produced lower $PM_{2.5}$ mass, UFP number emissions, and F_{30} values. [Kero-WS] had low emissions of $PM_{2.5}$ mass and UFP number, but the F_{30} value is relatively high; moreover, [Kero-WS] had relatively high emissions of UFP in the low-power phase compared with those in the high-power phase, as discussed above. [Pell-NDS] had lower emissions of $PM_{2.5}$ mass and UFP number compared with uncompressed wood, but F_{30} values for [Pell-NDS] (58–77%) were higher than for wood burned in the 3-stone fire and in the NDSs. A relatively high F_{30} value is also found for [LMW-FDS] (~80%) and for [RiH-FDS] (60–70%). In another laboratory study, UFP emissions from wood combustion in a gasifier cookstove was reported to be about 50% lower compared with wood combustion in a natural-draft rocket stove, and 70% lower compared with a 3-stone wood fire, but the number of particles <30 nm (note the range of the SMPS was 14.6–661 nm diameter, same as the present study) were 3.5 and 3.1 times that of the 3-stone fire on a fuel energy basis.²⁴ Another recent study also found wood combustion in the stove with additional air injection produced more ultrafine particles < 20 nm than wood combustion in the three-stone fire on the basis of emissions per dry fuel mass.²⁶ Hawley and Volckens²⁹ observed fewer but smaller-sized particles from the gasifier stove compared with traditional three-stone fires, with the count median diameter of 19 and 92 nm, respectively, even though emissions from the gasifier produced much lower pro-inflammatory impacts. Therefore, clean fuels like LPG and alcohol may be promoted in interventions to alleviate household air pollution and protect human health. The use of kerosene should be prevented due to high fractions of ultrafine particles despite the total number emissions being less than those for solid wood combustion. In fact, the *WHO Guidelines for Indoor Air Quality for Household Fuel Combustion*⁴⁶ discourage household use of kerosene as existing evidence shows that it leads to PM, CO, SO₂, polycyclic aromatic hydrocarbon (PAHs) levels that exceed WHO guidelines; and risks of burns, fires and poisoning are also cause for concern.

Residential cookstoves are an important source of ultrafine particle emissions that have significant impacts on human health and climate; however, research on UFP emission factors and PNSD is in its infancy. The present study investigated eleven different fuel-stove combinations including gas, liquid, and solid fuels (rice hulls, wood and pellets), and different stove technologies. The reported emission factors could be applied in future emission inventories to reduce uncertainties. Considering various fuels and stoves used in practice, more studies are called for, especially for low-emission non-solid fuels and solid fuel combustion in forced-draft stoves. $PM_{2.5}$ mass emissions from forced-draft stoves are typically lower compared with emissions from natural-draft wood fueled stoves, but UFP number emissions and F_{30} values may not be lower to the same degree. Emissions should be compared on a delivered energy basis when evaluating intervention options. This study suggests that evaluation of pollutant indicators in addition to CO and $PM_{2.5}$ mass emissions may be valuable, and PNSD is also important to take into consideration.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1-

Number size distributions of UFPs in the cold-start, high-power phase for tested fuel-stove groups. (A) unimodal distributions with peaks ~30–40 nm; (B) unimodal distributions with peaks < 20 nm; and (C) bimodal distributions. Data show means (line) and standard deviations (shaded area).

Figure 2- Normalized particle number size distributions for startup and steady state for [LMW-3SF], [LMW-NDS1], [LMW-FDS] and [Pell-NDS]. Results for all tested fuel-stove groups are presented in Figure S9. Startup is the first 150 s (the time resolution set in the current SMPS system), while steady state is 10 or 12.5 min after ignition (depending on fuel/stove types). Results shown are means (line) and standard deviations (shaded area) from repeat trials.

Figure 3- TEM images illustrating the soot aggregate morphology, primary particle connectivity and internal nanostructure. This particular sample was collected from low-moisture wood fuel burned in a natural drafts stove (LMW-NDS2).

Figure 4.

Comparison of UFP number emissions with PM_{2.5} mass emission during the high- and low-power test phases. Data shown are means and standard deviations.

Figure 5-
Emissions of PM_{2.5} mass, UFP numbers and fractions of particles smaller than 30 nm for tested fuel/stove types during the cold-start high-power phase. Fuel-stove combinations in the figure are arranged in an increasing trend in PM_{2.5} mass emission.

Table 1

Information on fuels and stoves in this study

No.	Fuel	Fuel moisture (wet basis)	Cookstove (manufacturer and model)	Forced-/Natural- Draft	Designated code*
1	LPG	n/a	Peru Solgas/Repsol LPG	ND	[LPG-OBS]
2	Alcohol	n/a	Dometic CleanCook, Model A1	ND	[Alc-EBS]
3	Kerosene	n/a	Butterfly wick, Model 2668	ND	[Kero-WS]
4	Pellet, wood	Low (~4.2%)	StoveTec Prototype	ND	[Pel-NDS]
5	Charcoal	Low (~4.0%)	EcoZoom Jet	ND	[Char-NDS]
6	Rice hulls	Low (~10%)	Belonio TLUD gasifier	FD	[RH-FDS]
7	Wood	Low (~10%)	3-stone fire	ND	[LMW-3SF]
8	Wood	Low (~6.5%)	JikoPoa, designed by Burn Design Labs	ND	[LMW-NDS1]
9	Wood	High (~18%)	JikoPoa, same as #8	ND	[HMW-NDS1]
10	Wood	Low (~8.7%)	Philips Electronics India Ltd, Model HD4008	ND	[LMW-NDS2]
11	Wood	Low (~8.7%)	Philips Electronics India Ltd, Model HD4012	FD	[LMW-FDS]

* OBS-Open Burner Stove; EBS-Evaporative Burner Stove; WS-Wick Stove; NDS-Natural Draft Stove; FDS-Forced Draft Stove; 3SF-Three Stone Fire; LMW-Low Moisture Wood; HMW-High Moisture Wood.

Table 2.

Thermal efficiency (TE), and emissions of UFPs from different fuel-stove types during the high-power cold-start, high-power hot-start and low-power phases. Emissions factors are presented on the basis of fuel mass (particles/kg), fuel energy (particles/MJ), useful energy delivered (particles/MJ_d) and rate (particles/hour). Results listed as means $\pm$ standard deviations from triplicate tests.

		[LPG-OBS]	[Alc-EBS]	[Kero-WS]	[Pel-NDS]	[Char-NDS]	[RiH-FDS]
High-power Cold-start phase	TE, %	56 $\pm$ 2	55 $\pm$ 2	41 $\pm$ 1	53 $\pm$ 6	31 $\pm$ 1	43 $\pm$ 4
	particles/kg	(4.5 $\pm$ 6.3) $\times 10^{12}$	(2.3 $\pm$ 1.8) $\times 10^{12}$	(1.5 $\pm$ 0.5) $\times 10^{15}$	(2.9 $\pm$ 0.4) $\times 10^{15}$	(5.5 $\pm$ 0.3) $\times 10^{15}$	(6.2 $\pm$ 1.6) $\times 10^{15}$
	particles/MJ	(1.0 $\pm$ 1.4) $\times 10^{11}$	(1.1 $\pm$ 0.9) $\times 10^{11}$	(3.5 $\pm$ 1.0) $\times 10^{13}$	(1.6 $\pm$ 0.2) $\times 10^{14}$	(1.8 $\pm$ 0.1) $\times 10^{14}$	(4.3 $\pm$ 1.1) $\times 10^{14}$
	particles/MJ _d	(1.7 $\pm$ 2.4) $\times 10^{11}$	(2.0 $\pm$ 1.6) $\times 10^{11}$	(8.6 $\pm$ 2.6) $\times 10^{13}$	(3.0 $\pm$ 0.5) $\times 10^{14}$	(5.7 $\pm$ 0.2) $\times 10^{14}$	(1.0 $\pm$ 0.3) $\times 10^{15}$
	particles/hour	(5.4 $\pm$ 7.6) $\times 10^{11}$	(4.6 $\pm$ 3.4) $\times 10^{11}$	(4.0 $\pm$ 1.1) $\times 10^{14}$	(7.7 $\pm$ 1.4) $\times 10^{14}$	(1.7 $\pm$ 0.1) $\times 10^{15}$	(2.6 $\pm$ 0.8) $\times 10^{15}$
High-power Hot-start phase	TE, %	58 $\pm$ 2	56 $\pm$ 2	43 $\pm$ 1	55 $\pm$ 2	38 $\pm$ 3	49 $\pm$ 3
	particles/kg	(1.7 $\pm$ 1.4) $\times 10^{12}$	(4.6 $\pm$ 3.3) $\times 10^{12}$	(1.6 $\pm$ 0.2) $\times 10^{15}$	(1.4 $\pm$ 0.6) $\times 10^{15}$	(5.8 $\pm$ 0.8) $\times 10^{15}$	(6.7 $\pm$ 0.6) $\times 10^{15}$
	particles/MJ	(3.8 $\pm$ 3.0) $\times 10^{10}$	(2.3 $\pm$ 1.6) $\times 10^{11}$	(3.6 $\pm$ 0.4) $\times 10^{13}$	(7.3 $\pm$ 3.1) $\times 10^{13}$	(1.9 $\pm$ 0.2) $\times 10^{14}$	(4.7 $\pm$ 0.4) $\times 10^{14}$
	particles/MJ _d	(6.4 $\pm$ 5.1) $\times 10^{10}$	(4.1 $\pm$ 3.1) $\times 10^{11}$	(8.5 $\pm$ 0.9) $\times 10^{13}$	(1.3 $\pm$ 0.5) $\times 10^{14}$	(4.9 $\pm$ 0.5) $\times 10^{14}$	(9.5 $\pm$ 0.9) $\times 10^{14}$
	particles/hour	(1.9 $\pm$ 1.5) $\times 10^{11}$	(1.0 $\pm$ 0.7) $\times 10^{12}$	(4.2 $\pm$ 0.5) $\times 10^{14}$	(4.1 $\pm$ 1.7) $\times 10^{14}$	(2.6 $\pm$ 0.1) $\times 10^{15}$	(2.4 $\pm$ 0.3) $\times 10^{15}$
Low-power phase	particles/kg	(2.9 $\pm$ 3.5) $\times 10^{12}$	(9.6 $\pm$ 8.1) $\times 10^{11}$	(3.9 $\pm$ 1.0) $\times 10^{15}$	(3.8 $\pm$ 1.4) $\times 10^{15}$	(5.6 $\pm$ 3.2) $\times 10^{14}$	(8.8 $\pm$ 2.5) $\times 10^{15}$
	particles/MJ	(6.5 $\pm$ 7.9) $\times 10^{10}$	(4.7 $\pm$ 3.9) $\times 10^{10}$	(9.0 $\pm$ 2.4) $\times 10^{13}$	(2.0 $\pm$ 0.8) $\times 10^{14}$	(1.8 $\pm$ 1.0) $\times 10^{13}$	(3.8 $\pm$ 1.8) $\times 10^{14}$
	particles/hour	(2.3 $\pm$ 2.7) $\times 10^{11}$	(1.9 $\pm$ 1.6) $\times 10^{11}$	(7.1 $\pm$ 1.3) $\times 10^{14}$	(9.2 $\pm$ 3.2) $\times 10^{14}$	(5.0 $\pm$ 2.8) $\times 10^{13}$	(1.6 $\pm$ 0.5) $\times 10^{15}$
		[LMW-3SF]	[LMW-NDS1]	[HMW-NDS1]	[LMW-NDS2]	[LMW-FDS]	
High-power Cold-start phase	TE, %	15 $\pm$ 1	24 $\pm$ 1	23 $\pm$ 1	34 $\pm$ 3	36 $\pm$ 1	
	particles/kg	(5.8 $\pm$ 1.6) $\times 10^{15}$	(4.5 $\pm$ 0.5) $\times 10^{15}$	(6.7 $\pm$ 0.9) $\times 10^{15}$	(5.4 $\pm$ 0.6) $\times 10^{15}$	(6.0 $\pm$ 0.6) $\times 10^{15}$	
	particles/MJ	(3.3 $\pm$ 0.9) $\times 10^{14}$	(2.5 $\pm$ 0.3) $\times 10^{14}$	(3.7 $\pm$ 0.5) $\times 10^{14}$	(3.1 $\pm$ 0.3) $\times 10^{14}$	(3.4 $\pm$ 0.3) $\times 10^{14}$	
	particles/MJ _d	(2.3 $\pm$ 0.7) $\times 10^{15}$	(1.0 $\pm$ 0.1) $\times 10^{15}$	(1.6 $\pm$ 0.3) $\times 10^{15}$	(9.0 $\pm$ 0.7) $\times 10^{14}$	(9.4 $\pm$ 0.9) $\times 10^{14}$	
	particles/hour	(9.4 $\pm$ 1.6) $\times 10^{15}$	(3.6 $\pm$ 0.5) $\times 10^{15}$	(4.5 $\pm$ 0.1) $\times 10^{15}$	(4.1 $\pm$ 0.6) $\times 10^{15}$	(5.6 $\pm$ 0.3) $\times 10^{15}$	
High-power Hot-start phase	TE, %	15 $\pm$ 1	25 $\pm$ 2	25 $\pm$ 1	35 $\pm$ 3	40 $\pm$ 1	
	particles/kg	(4.2 $\pm$ 1.5) $\times 10^{15}$	(3.8 $\pm$ 0.9) $\times 10^{15}$	(5.7 $\pm$ 1.1) $\times 10^{15}$	(5.4 $\pm$ 1.1) $\times 10^{15}$	(4.8 $\pm$ 1.2) $\times 10^{15}$	
	particles/MJ	(2.4 $\pm$ 0.9) $\times 10^{14}$	(2.1 $\pm$ 0.5) $\times 10^{14}$	(3.1 $\pm$ 0.6) $\times 10^{14}$	(3.0 $\pm$ 0.6) $\times 10^{14}$	(2.7 $\pm$ 0.7) $\times 10^{14}$	
	particles/MJ _d	(1.6 $\pm$ 0.6) $\times 10^{15}$	(8.4 $\pm$ 0.2) $\times 10^{14}$	(1.3 $\pm$ 0.3) $\times 10^{15}$	(8.9 $\pm$ 2.4) $\times 10^{14}$	(6.8 $\pm$ 1.9) $\times 10^{14}$	

	particles/hour	$(8.0\pm0.2)\times10^{15}$	$(3.61\pm0.9)\times10^{15}$	$(4.6\pm0.6)\times10^{15}$	$(4.0\pm0.8)\times10^{15}$	$(5.0\pm1.0)\times10^{15}$
Low-power phase	particles/kg	$(8.8\pm2.9)\times10^{15}$	$(5.7\pm0.8)\times10^{15}$	$(7.0\pm0.4)\times10^{15}$	$(5.2\pm1.0)\times10^{15}$	$(6.4\pm0.6)\times10^{15}$
	particles/MJ	$(5.0\pm1.6)\times10^{14}$	$(3.1\pm0.4)\times10^{14}$	$(3.8\pm0.2)\times10^{14}$	$(2.9\pm0.6)\times10^{14}$	$(3.6\pm0.4)\times10^{14}$
	particles/hour	$(8.4\pm2.2)\times10^{15}$	$(3.2\pm0.6)\times10^{15}$	$(4.2\pm0.4)\times10^{15}$	$(2.0\pm0.4)\times10^{15}$	$(2.2\pm0.2)\times10^{15}$