

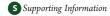


pubs.acs.org/EF

Emission Characterization and Efficiency Measurements of High-Efficiency Wood Boilers

Sriraam R. Chandrasekaran, James R. Laing, Thomas M. Holsen, Suresh Raja, and Philip K. Hopke*

Center for Air Resources Engineering and Science, Clarkson University, Post Office Box 5708, Potsdam, New York 13699, United States



ABSTRACT: Detailed gaseous and particle emissions along with thermal efficiency measurements were made on three mid-sized high-efficiency wood boilers with thermal output capacities of $150 \,\mathrm{kW}$ ($514\,000 \,\mathrm{Btu} \,\mathrm{h}^{-1}$) (n=2) and $500 \,\mathrm{kW}$ ($1.7 \,\mathrm{mmBtu} \,\mathrm{h}^{-1}$) (n=1). Wood chips and commercial wood pellets were used as fuel. Continuous emissions of CO, NO₃₁ SO₂₁ fine particle mass (PM_{2.5}), and ultrafine particle number distributions were determined using a dilution tunnel sampling system. PM_{2.5} and semivolatile organic compound characterization was performed. Low concentrations of CO, organic carbon (OC), and elemental carbon (EC) during steady-state boiler operation indicated good combustion conditions. Fine particle mass from wood pellets was predominantly K and SO₄²⁻, with <8% OC and <2% EC. Inorganic emissions (PM_{2.5}, NO_x, and SO₂) were found to depend upon fuel quality, which indicates the need for wood pellet fuel standards in the U.S. Cd, Pb, Ti, Rb, and Zn were found to be enriched in PM_{2.5}, which is of concern for human health. Levoglucosan was a predominant organic compound found for all fuels, ranging from 38 to 82 µg/MJ. Total particle and semivolatile polycyclic aromatic hydrocarbon (PAH) emissions were relatively low (19.4–92.8 μ g/MJ). The thermal efficiencies of the wood pellet boilers determined using the provisional American Society of Heating, Refrigeration and Air Conditioning Engineers (ASHRAE) standard 155p ranged between 70 and 86% for the 150 kW boiler tested and between 75 and 91% for the 500 kW boiler. The use of ASHRAE 155p allowed for the determination of the linear relationship between the energy input and the energy output over the range of energy outputs rather than only determining the efficiency at minimum and full load as is the current standard practice. Boiler capacity had no significant effect on efficiency; however, the operating conditions, such as fuel feed rate, outlet water temperature, and building demand, did affect the results.

■ INTRODUCTION

There has been increasing interest in developing sustainable energy sources, especially biomass-based fuel, such as wood pellets and wood chips, for space-heating purposes because of concerns about fossil fuel depletion and global climate change. Biomass can be renewable and a CO₂-neutral energy source. It can be produced domestically and stimulate local economies. It has been shown to be a promising source of alternate energy for commercial or industrial hot air, hot water, steam, and electricity, especially in the northeastern U.S.² In the commercial building sector, space heating ranks second in terms of energy use.^{3,4}

If an increase in the use of biomass energy in the U.S. is to be achieved, health and environmental considerations must be considered. Conventional wood stoves, which make up the majority of the fleet in the U.S., have relatively low efficiency and significant emissions of CO, soot, particulate matter, and other harmful pollutants, which are a concern from the perspectives of global climate change and human health.⁵ Many European countries have emission, efficiency, and fuel standards for biomass combustion devices. These standards have driven the development of many advanced combustion systems that provide substantially higher thermal efficiency and lower emissions than conventional systems. Currently, there is little information available on the emissions and efficiency of mid-sized biomass boilers (100-500 kW). A majority of the combustion devices investigated previously have either been residential burners and boilers $(2-15~\text{kW})^{7-10}$ or large district heating boilers (>1 MW).11-14 Studies that have looked at mid-sized boilers have

generally only measured fine particle mass (PM_{2.5}). It is important to fully characterize the emissions from these boilers if they are going to replace existing oil and natural gas boilers.

The objective of the present study was to characterize the emissions and efficiency of three high-efficiency boilers with thermal capacities of 150 kW (514 000 Btu $\rm h^{-1}$) (n=2) and 500 kW (1.7 mmBtu $\rm h^{-1}$) (n=1). In this study, the American Society of Heating, Refrigeration and Air Conditioning Engineers (ASHRAE) standard 155p new provisional protocol was used to determine the full load, partial load, and seasonal efficiency. The standard can be applied to determine the space heating performance and is applicable to all boilers with energy input values ranging from 300 000 to 12 500 000 Btu $\rm h^{-1}$. Previous performance standards do not account for heat losses in the heat exchanger or only account for thermal efficiency at full-load conditions. ASHRAE 155p measures the efficiency over the range of energy output values rather than only making measurements at minimum and fuel boiler loads.

■ MATERIALS AND METHODS

Boiler Descriptions. Measurements were carried out on three commercial high-efficiency boilers: a 150 kW boiler installed at Clarkson University's Walker Center in Potsdam, NY (WAC), an identical 150 kW

Received: August 19, 2011
Revised: October 12, 2011
Published: October 17, 2011



boiler at ACT Bioenergy's facility in Schenectady, NY that was subsequently installed at the Cayuga Nature Center in Ithaca, NY (CNC), and a 500 kW boiler integrated with a solar hot-water system installed at the Wild Center Museum in Tupper Lake, NY (WIC). The smaller boilers are 150 kW Hamont CATfire wood boilers imported by ACT Bioenergy (Advance Climate Technologies, LLC), and the Wild Center boiler is a 500 kW Hamont CATfire manufactured by ACT Bioenergy.

All boilers were of the same general design (pictures of the WAC and WIC boilers can be found in Figures S1 and S2 of the Supporting Information). The boilers employ fully automated bottom fed fuel feeding systems and a triple air-staging combustion process. Air staging is accomplished by heating the fuel bed in an oxygen-deficient environment and tangentially injecting secondary and tertiary air at a high air/fuel ratio to burn pyrolysis gases. Thorough mixing of combustion air with pyrolysis gases allows the boiler to operate at low excess air levels, increasing the temperature in the combustion zone and, thereby, increasing the combustion efficiency. Low temperatures and the presence of oxygen in the fuel bed have been observed to reduce NO_x and inorganic particle emissions. ¹⁶ The boilers were equipped with a cyclone to remove coarse particles from the flue gas. Prior to the measurements, the boilers were run for at least 1 h to bring them to the operating temperature.

The WAC boiler was not American Society of Mechanical Engineer (ASME)-certified and, thus, had to be operated unpressurized. It had an external heat exchanger to transfer heat from the boiler water loop to the building water loop (see Figure S3 of the Supporting Information). The CNC and WIC boilers (see Figure S4 of the Supporting Information) were ASME-certified and did not have an external heat exchanger.

Fuels. Calorific value, moisture, ash, nitrogen, and sulfur contents, and elemental compositions of the fuels were determined (Table 1 and Table S1 of the Supporting Information). Most of the elements were determined by inductively coupled plasma—mass spectrometry (ICP—MS). Nitrogen analysis was performed according to American Society for Testing and Materials (ASTM) D5291. Sulfur and chlorine content were determined by ion chromatography (IC) analysis of the

Table 1. Fuel Characteristics

	pellets A	wood chips	pellets B
gross calorific value (Btu/lbs)	8180	6371	8060
moisture (%)	4.6	26	5.1
ash (%, dw)	0.71	1.79	0.47
N (mg/kg, dw)	1300	3700	1424
S (mg/kg, dw)	74.1	175	63.6
Cl (mg/kg, dw)	38.8	nm	nm

oxygen bomb washings according to ASTM E775 and United States Environmental Protection Agency (U.S. EPA) SWP-846 method 5050, respectively. Commercially available wood pellets were used at the WAC (pellets A) and the WIC (pellets B). The CNC used wood chips consisting of waste wood and forest residue. The wood pellets had low moisture (4.61 and 5.10%) and ash [0.71 and 0.47% dry weight (dw)] contents compared to the wood chips (26.4% moisture and 1.79% dw ash). Nitrogen, sulfur, and trace element concentrations were also significantly higher for the wood chips.

Emission Measurements. A dilution sampling system with a 2.5 µm in-stack cyclone conforming to CTM-039 of the U.S. EPA was used for emission measurements (Figure 1). Dilution sampling simulates ambient conditions by rapidly mixing hot flue gas with high-efficiency particulate air (HEPA)-filtered ambient air, enhancing secondary aerosol formation.¹⁷ Flow rates of sample and dilution air were calculated using the differential pressure of factory-calibrated venturi flow elements and the air temperature. Dilution ratios between 20 and 40 were used. CO, NOx, and SO2 were measured continuously using ambient gas monitors (models 42i, 43i, and 48i, Thermo Scientific). Continuous measurements of the PM_{2.5} mass concentration were made using a Filter Dynamics Measurement System (FDMS), consisting of a Tapered Element Oscillating Microbalance (TEOM) particle monitor with an FDMS kit (models 8500b and 1400ab, Thermo Scientific). Ultrafine particle number concentration and particle size distributions in the range from 5.6 to 560 nm were measured using a fast mobility particle sizer spectrometer (FMPS, model 3091, TSI, Inc.). All emission values are reported at dry standard state conditions (283.34 K and 101.325 kPa). Continuous measurements were made for 17.2, 7.2, and 6.0 h for the WAC, CNC, and WIC, respectively. Emission factors were converted to nominal emissions (1 MJ⁻¹) per fuel energy input because the flow rate in the stack and fuel energy input rate were known.

Quartz fiber filters were analyzed for organic carbon (OC) and elemental carbon (EC) by the National Institute for Occupational Safety and Health (NIOSH) 5040 method using a Sunset Laboratories analyzer. Prior to use, the filters were baked in a muffle furnace at 550 °C for 16 h to remove organics. OC was artifact-corrected using two parallel sample lines: the first with a quartz filter and the second with a Teflon filter followed by a quartz filter. The quartz filter downstream of the Teflon filter provided an estimate of the gas-phase OC. PA 1.5 cm² punch from the quartz fiber filters was sonicated in 10 mL of Milli-Q water for 60 min and analyzed for major anions and cations by IC. The Teflon filters were microwave-digested (MARS Express, CEM Corp.) in 10 mL of 50% HNO3 and analyzed for trace metals by ICP—MS using EPA Method IO-3.5.

Semivolatile and particle organic compounds were collected on quartz filters followed by polyurethane foam plugs (PUFs) and analyzed

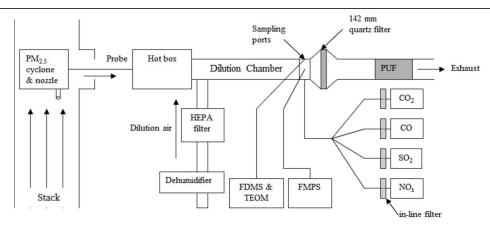


Figure 1. Schematic of dilution tunnel and measurement devices.

by gas chromatography—mass spectrometry (GC-MS). A total of 133 compounds were included in the analysis. Prior to use, the PUF glass linears were baked in a muffle furnace at 500 °C for 1 h and the quartz filters were baked at 550 °C for 16 h. PUFs were extracted in 100 mL of dichloromethane, dried with nitrogen, wrapped in aluminum foil, and sealed before use. After collection, the filters were extracted with a 100 mL solution of dichloromethane and methanol (4:1, v/v) and the PUFs were extracted in 100 mL of dichloromethane using an ASE 300 accelerated solvent extraction system (Dionex Corp., Sunnyvale, CA). After the extract was concentrated by evaporation with ultrapure nitrogen, the sample was split into two fractions. One aliquot was analyzed immediately, and another was reacted with 99% bis(trimethylsilyl)trifluoroacetamide (BSTFA) and 1% trimethylchlorosilane (TMCS) to convert the polar compounds into their trimethylsilyl derivatives before being analyzed.²¹ GC-MS analysis was performed using a DSQ II single-quadrupole GC-MS system (Thermo Scientific, Inc., Franklin, MA) equipped with a 30 m length \times 0.25 mm inner diameter \times 0.25 μ m film thickness SGE forte GC capillary column (Restek Co., Ballefonte, PA).

Quartz filters and PUFs collected at the WAC were also analyzed for polychlorinated dibenzodioxins and polychlorinated dibenzofurans (PCDD/Fs). Samples were extracted with toluene, and analysis was performed at State University of New York at Fredonia (SUNY Fredonia). A total of 7 polychlorinated dibenzodioxins were analyzed for, including 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, and OCDD, along with 10 polychlorinated dibenzofurans, including 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-eCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,4,6,7,8-HxCDF, 1,2,3,7,8,9-HyCDF, 1,2,3,7,8,9-HyCDF, 1,2,3,7,8,9-HyCDF, 1,2,3,4,7,8,9-HpCDF, 1,2,3,4,7,8,9-HpCDF, 1,2,3,4,7,8,9-HpCDF, 1,2,3,4,7,8,9-HpCDF, and OCDF.

Thermal Efficiency Measurement Methods. Boiler efficiency was determined using the direct method of dividing the useful heat output of the boiler by the energy input of the fuel (eq 1).

$$\eta = \frac{\text{heat input}}{\text{heat output}} \tag{1}$$

Heat input was calculated from the gross calorific value of the fuel. Heat output from the boiler was determined by the temperature differences and flow rates in the output and return water pipe. Temperatures at the WAC were measured with thermocouples connected to a portable handheld data logger (Omega DAQPRO-5300). A handheld ultrasonic flow meter with type M1 transducers (Shenitch STUF-200H) was used to measure the water flow rate.

The WIC boiler had an automated reporting system for the input and output energies. On the basis of real-time measurements of the fuel feed rate via the boiler control system and the higher heating value of the pellets, the input energy was calculated to determine the boiler efficiency based on boiler loads. The fuel feed rate for CNC and WAC was measured by hand feeding a known quantity of pellets into the feeding bin of the boiler and recording the time of consumption.

■ RESULTS AND DISCUSSION

Gaseous and Particulate Emissions. During continuous measurement periods, the WAC and CNC boilers ran at steady state and operated at thermal inputs of 114 and 90 kW, respectively. The WIC boiler did not run at steady state throughout the continuous emissions testing because of an insufficient heat demand in the building. Thermal inputs for the WIC boiler ranged from 378 to 621 kW throughout testing. Testing at the WIC boiler was performed soon after boiler installation and fuel auger rates were manually chosen. During one portion of the testing, the fuel feed rate, calculated from the fraction of time that the fuel auger ran and the weight of pellets delivered per unit time

Table 2. Emission Factors (mg/MJ) with 95% Confidence Intervals

boiler	Walker Center	Cayuga	Wild Center
fuel	pellets A	wood chips	pellets B
СО	107 ± 2.51	125 ± 4.29	734 ± 125
NO_x	17.3 ± 0.17	127 ± 1.67	35.9 ± 1.34
SO_2	$\boldsymbol{0.21 \pm 0.02}$	$\boldsymbol{1.42 \pm 0.02}$	$\textbf{0.44} \pm \textbf{0.02}$
$PM_{2.5}$	26.0 ± 0.47	41.1 ± 1.40	26.3 ± 3.53
$N_{\rm FMPS}$ ($\times 10^{11}$, number/MJ)	134 ± 1.15	360 ± 6.03	230 ± 14.3
$\mathrm{GMD}_{\mathrm{FMPS}}$	91.4 ± 0.31	80.0 ± 0.26	70.3 ± 0.75
OC	$\boldsymbol{1.90 \pm 0.20}$	$\textbf{0.70} \pm \textbf{0.15}$	$\boldsymbol{0.58 \pm 0.50}$
EC	0.11 ± 0.01	$\textbf{0.02} \pm \textbf{0.04}$	$\textbf{0.24} \pm \textbf{0.19}$

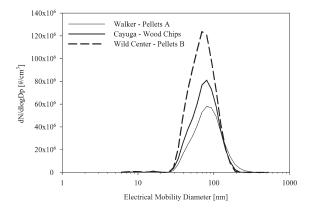


Figure 2. Steady-state ultrafine particle number size distributions for WAC, CNC, and WIC boilers.

when the auger ran, was determined to be higher than the rated capacity of the boiler. All filter and PUF measurements were taken during steady-state periods.

Carbon monoxide emissions, which indicate the completeness of combustion, were significantly higher (*p* value < 0.001) for the WIC boiler (734 mg/MJ) than for the WAC and CNC boilers (107 and 125 mg/MJ) (Table 2). The CO concentration at the WIC boiler was highly variable, characterized by intermittent large peaks of high concentration. Testing was performed soon after installation, and the boiler was not running at steady state, which may account for these results. Previous work has shown that modern pellet burners and boilers (5–20 kW) operating with a nominal feed rate have CO emissions ranging from 21 to 194 mg/MJ, and when operating intermittently, the emission factors range from 155 to 1100 mg/MJ. Sippula et al. ¹³ observed CO emission factors of 6.60–117 mg/MJ for four district heating boilers (5–15 MW) operating at steady-state.

Nitrogen oxide (NO_x) and sulfur dioxide (SO_2) emissions were significantly higher (p value < 0.001) from wood chips compared to wood pellets. NO_x emissions from biomass burning are all derived from nitrogen in the fuel because temperatures in the wood boilers are not high enough to form significant amounts of thermal or prompt NO_x . The higher NO_x and SO_2 emissions from wood chips are due to higher nitrogen and sulfur in the wood chips compared to wood pellets. This relationship has been observed in other studies. A relatively low percentage of fuel-bound nitrogen was emitted as NO_x (14, 25, and 21% for pellets A, wood chips, and pellets B, respectively), possibly because of the air-staging technology employed by the boilers.

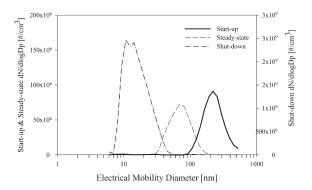


Figure 3. Ultrafine particle number size distributions for startup, steady-state, and shutdown conditions.

Table 3. Chemical Composition of PM_{2.5} from Wood Pellets

Tuble 3.	enemical composition of 1 W	2.5 Hom Wood I the			
	Walker Center (WAC)	Wild Center (WIC)			
species	pellets A	pellets B			
	Organic and Elemental Carbon (wt				
OC	7.67	4.89			
EC	0.00	1.95			
	Ionic Species (wt % of PM _{2.5})				
Na^+	0.18	0.18			
K^{+}	40.6	15.3			
Mg^+	0.06	na ^a			
Ca ²⁺	0.05	na			
SO_4^{2-}	20.0	24.4			
Cl^-	0.44	1.68			
Elemental Species (wt % of PM _{2.5})					
Al	${<}\mathrm{DL}^{b}$	0.056			
As	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>			
Ba	4.13×10^{-3}	0.059			
Ca	<dl< td=""><td>0.53</td></dl<>	0.53			
Cd	1.07×10^{-3}	1.64×10^{-3}			
Co	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>			
Cr	8.69×10^{-4}	0.013			
Cu	0.011	0.053			
Fe	<dl< td=""><td>0.12</td></dl<>	0.12			
Mg	0.013	0.089			
Mn	0.040	0.39			
Ni	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>			
Pb	0.014	0.027			
Rb	0.089	0.064			
Sb	<dl< td=""><td><dl< td=""></dl<></td></dl<>	<dl< td=""></dl<>			
Sn	<dl< td=""><td>2.70×10^{-3}</td></dl<>	2.70×10^{-3}			
Sr	5.36×10^{-4}	0.012			
Tl	1.27×10^{-4}	2.82×10^{-4}			
V	6.93×10^{-5}	6.97×10^{-5}			
Zn	0.15	0.29			
a na = not avaiable. b <dl =="" below="" detection="" limit.<="" td="" the=""></dl>					

Emissions of PM_{2.5} were highest (p value < 0.001) for wood chips (41.1 mg/MJ) compared to commercial pellets (26.0 and 26.3 mg/MJ for the WAC and WIC, respectively). Because the carbonaceous fraction of PM_{2.5} was very low (<8%), the higher

Table 4. Recovery Fraction in PM_{2.5} of Trace Metals

	recovery in PM _{2.5} (wt %)		
	Walker Center (WAC)	Wild Center (WIC)	
element	pellets A	pellets B	
Al	<1.23	2.10	
As	<33.8	<37.7	
Ba	0.54	4.61	
Ca	<0.44	0.40	
Cd	411	848	
Co	<16.1	<12.6	
Cr	3.95	19.4	
Cu	23.1	17.2	
Fe	<7.01	2.90	
Mg	0.06	0.25	
Mn	0.74	6.30	
Ni	<26.5	<9.83	
Pb	96.3	87.1	
Rb	69.1	25.9	
Sb	<33.8	<23.2	
Sr	0.10	1.62	
Tl	<137	278	
V	4.18	0.31	
Zn	45.6	27.8	

particle emissions from wood chips are due to higher fuel ash content in the wood chips. Residential pellet stoves and boilers $(5-20~\mathrm{kW})$ previously tested at steady state have comparable emission factors $(13-58~\mathrm{mg/MJ}).^{7-10}$ A survey compiled from 23 institutions in 8 European countries found automated pellet stoves and boilers to have an averaged PM emission factor of 32 $\mathrm{mg/MJ}.^6$

Ultrafine Particle Number Size Distributions. Steady-state particle number size distributions for the three boilers were lognormal (Figure 2) with geometric mean diameters (GMDs) between 70 and 92 nm (Table 2). During startup periods, the distribution remained log-normal but the GMD was much larger than during steady-state (234 nm) (Figure 3). This larger size distribution was likely the result of incomplete combustion producing larger unburnt fuel particles because they ablate from the surface of the pellet or chip. The number size distribution during shutdown conditions was much smaller than during steady-state (GMD of 17.1 nm), with most of the particles in the accumulation mode. These particles are formed by evolution of vapor that nucleate into small particles that are not combusted because the active flaming process has ceased.

Particle Chemical Characterization. The majority of resolved fine PM mass consisted of potassium (K⁺) and sulfate ($\mathrm{SO_4}^{2-}$) (87 and 79% for the WAC and WIC, respectively) (Table 3). There were also minor amounts of $\mathrm{Na^+}$, Mg, Ca, and Zn. The low amount of chloride relative to sulfate may be due to gas-phase sulfation reactions, in which $\mathrm{SO_2}$ reacts with alkali chlorides to form particle alkali sulfates and gaseous HCl. ²³ The chemical analysis resolved 69 and 50% of the total PM_{2.5} mass for the WAC and WIC, respectively. The unresolved portion, which is the difference between the chemical analysis and PM_{2.5} mass, is likely oxygen, hydrogen, and other elements that are part of organic compounds, inorganic hydroxides and oxides, or carbonates. The unresolved fraction also reflects analytical uncertainties.

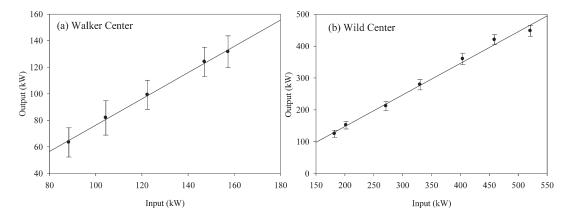


Figure 4. Thermal energy input rate and output rate of the (a) Walker Center and (b) Wild Center boilers (input rate calculated using the gross calorific value).

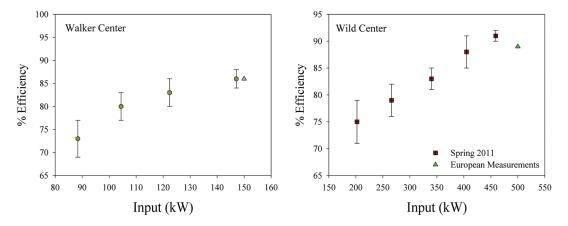


Figure 5. Thermal efficiency of the (a) Walker Center and (b) Wild Center boilers for various energy inputs.

The trace metal recovery fractions were estimated by comparing the elemental concentrations in the fuel to the concentrations in PM_{2.5}. Cd, Cu, Pb, Tl, Rb, and Zn were enriched in fine PM, with recoveries greater than 20% (Table 4). From a public-health perspective, this is important because, despite low levels in the fuel, exposure can be significant. In addition, heavy metals tend to adsorb onto existing particles to be surface-enriched, which increases their bioavailability. 24

Semivolatile and Fine Particle Organic Emissions. Quartz fiber filter and PUF measurements were summed to provide semivolatile and particle organic compound concentrations (see Table S2 of the Supporting Information). Gas-particle partitioning was not determined because of gas-phase adsorption of organic species onto quartz filters. ¹⁷ Most previous studies of organic compounds have focused on fireplace and open burning^{25–28} that produce emissions with much higher concentrations than found in this study. This distinction has implications for source apportionment studies. Although levoglucosan, a molecular marker of wood combustion, was a predominant compound found for all three fuels, the concentrations were relatively low (37.9, 43.0, and 82.3 μ g/MJ for pellets A, wood chips, and pellets B, respectively). Total polycyclic aromatic hydrocarbon (PAH) emissions were relatively low (19.4–92.8 μ g/MJ), which is comparable to previous studies of modern pellet stoves and boilers at steady state $(7-320 \,\mu g/MJ)$.^{7,8}

Four sets of filter/PUFs collected from wood pellets at WAC were analyzed for PCDD/Fs. Included in the analysis were 7 polychlorinated dibenzodioxins and 10 polychlorinated dibenzofurans. All 17 species were below the detection limit, yielding a total PCDD/F emission concentration of <22.4 pg/m³ and an emission factor of <14.5 pg/MJ. PCDD/F formation is dependent upon the chlorine content of the fuel and is primarily of concern when burning treated or contaminated waste wood or annual biofuels. ²⁹ Because of the low Cl content of pellets A (39 mg/kg of dry fuel; Table 1), low PCDD/F concentrations were expected.

Thermal Efficiency Measurements. Both the WAC (70–86%) and WIC (68–91%) boilers had similar thermal efficiencies ranges at the various energy input rates tested. Boiler input and output energy were linearly related (Figure 4), and efficiency increased with the input rate (Figure 5). The maximum efficiency measured was about 86% for the WAC boiler and 91% for the WIC boiler (see Figure S5 of the Supporting Information). The measured efficiencies were in good agreement with European measurements of the same model boilers (Figure 5).

Thermal Efficiency Optimization. The building demand is affected by a number of parameters. In addition to the outdoor temperature, the wind velocity, the solar gain, the presence of machines or equipment that radiate heat, and the number of persons present in the building have to be considered. However, the most dominant factor of those above-mentioned affecting the

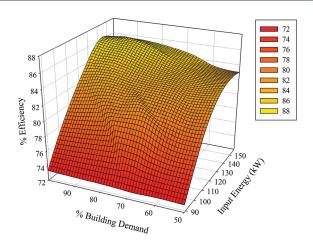


Figure 6. Optimization of operating parameters in the Walker Center boiler.

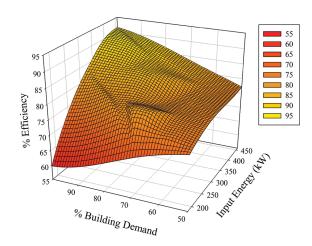


Figure 7. Optimization of operating parameters in the Wild Center boiler.

energy consumption is the outdoor temperature. 30 In this analysis, the building demand was calculated using the average outdoor temperature and the required building temperature (18 $^{\circ}$ C) as follows:

$$\% \text{ bd} = \frac{T_{\text{avg}} - T_{\text{set}}}{T_{\text{set}}} \times 100 \tag{2}$$

where bd is the building demand (%), $T_{\rm avg}$ is the average ambient temperature (°F), and $T_{\rm set}$ is the set temperature in the building (°F) (usually 18 °C).

Several experiments were conducted on the WAC and WIC boilers to examine the boiler performance as a function of operating parameters (Figures 6 and 7). The plots represent the variation in efficiency with respect to building demand and the boiler energy input. As the input rate and the building demand increased, the efficiency increased. However, for lower boiler inputs and higher building demands, the efficiency rose slightly and then did not increase further. This behavior probably meant that the boiler was unable to meet the building demand. It was not possible to measure efficiency at lower boiler input rates when the demand in the building was high (Figures 6 and 7).

The experimental conditions were such that the full domain of the parameters could not be examined. Thus, Figures 6 and 7 only present the range of parameter values that were examined. For instance, it was not possible because of the needs for use of the building to measure efficiency with lower boiler input rates when the demand in the building was high because this would have resulted in an insufficient heat supply. High input rates and low building demand would have caused the boiler to have incomplete combustion, and that would result in incompletely combusted fuel being expelled into the ash bin. Both of these conditions could be expected to result in lower efficiency.

The WIC boiler had an automated online reporting system from solarwave.net. This system is a green-energy-monitoring program that works with existing programs and hardware to deliver web-based monitoring and reporting. The optimization plot for the WIC boiler was based on the real time data obtained from solarwave.net. Operating the boiler at the highest fuel feed rate and outlet water temperature proved to be most efficient at the higher building demands. Operating the wood boiler at low building demands during the shoulder heating seasons resulted in lower efficiency because the boiler was operating at low loads. Installing thermal storage could provide stored excess energy that can then be used to support periods of low building heat demand. Tapping stored hot water would significantly reduce the boiler cycling, allowing it to operate mostly at high-load, high-efficiency conditions, regardless of the heat demand in the building. This approach would also result in lower emissions.

CONCLUSION

Emission characterization and thermal efficiency measurements were performed on commercial-sized wood boilers. Thermal efficiency values estimated using the ASHRAE 155p methodology ranged between 70 and 86% for the WAC boiler and 75 and 91% for the WIC boiler. The use of ASHRAE 155p allowed for the determination of the linear relationship between the energy input and the energy output over the range of energy output rather than only determining the efficiency at minimum and full load, which is the current standard practice. The thermal efficiency was found to be the function of both energy input and building demand. Low CO emissions were observed for the WAC and CNC boilers during steady-state operation, and emitted fine particle mass was predominantly K⁺ and SO₄²⁻, with <8% combustible carbon (OC and EC). The small amount of carbonaceous emissions indicated high combustion efficiencies. In comparison to other wood combustion technologies, the emissions are significantly lower but they are not as low as for oil or natural gas boilers; therefore, controls may be required if wood boilers are to replace these fossil-fuel-based heat sources, especially in buildings that house sensitive populations, such as schools or hospitals. Fuel quality was observed to influence the emissions of PM_{2.5}, NO_x, and SO₂. Some heavy metals (Cd, Pb, Ti, Rb, and Zn) were found in the emitted PM_{2.5}, suggesting a need for testing of wood pellets to meet appropriate fuel-quality standards. If lower emissions are required, emission control technologies can be implemented because they have been found to work well for wood combustion.31

ASSOCIATED CONTENT

Supporting Information. Fuel characteristics of the two wood pellets and wood chips used as fuel (Table S1), semivolatile and particle organic compound emission factors (μ g/MJ) using wood pellets and wood chips as fuel (Table S2), Walker Center

150 kW boiler (Figure S1), Wild Center 500 kW boiler (Figure S2), schematics of the Walker Center boiler (Figure S3), and schematics of the Wild Center boiler (obtained from http://beta.solarwave.net/WildCenterDownload.php) (Figure S4). This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*Telephone: 315-268-3861. Fax: 315-268-4410. E-mail: hopkepk@clarkson.edu.

ACKNOWLEDGMENT

The authors acknowledge the financial support provided by the New York State Energy Research and Development Authority (NYSERDA 10672). The authors thank David Dungate for the technical support and Chris Rdzanek and Bob Andrews of the Wild Center and John Frary and other WAC staff for their support. The authors also thank Mark Omara for his assistance.

■ REFERENCES

- (1) Luque, R.; Davila, L. H.; Campelo, J. M.; Clark, J. H.; Hidalgo, J. M.; Luna, D.; Marinas, J. M.; Romero, A. A. *Energy Environ. Sci.* **2008**, *1*, 542–564.
- (2) Richter, D. D.; Jenkins, D. H.; Karakash, J. T.; Knight, J.; McCreery, L. R.; Nemestothy, K. P. *Science* **2009**, 323, 1432–1433.
 - (3) Hewet, M. J. ASHRAE J. 2005, 10-21.
- (4) Energy Information Administration (EIA). Commercial Building Energy Consumption Survey; EIA: Washington, D.C., 2002.
- (S) Naeher, L. P.; Brauer, M.; Lipsett, M.; Zelikoff, J. T.; Simpson, C. D.; Koenig, J. Q.; Smith, K. R. *Inflamation Toxicol.* **2007**, *19*, 67–106.
- (6) New York State Energy Research and Development Authority (NYSERDA). Biomass Combustion in Europe: Overview on Technologies and Regulations; NYSERDA: Albany, NY, 2008.
- (7) Boman, C.; Nordin, A.; Boström, D.; Öhman, M. Energy Fuels **2004**, *18*, 338–348.
- (8) Johansson, L. S.; Leckner, B.; Gustavsson, L.; Cooper, D.; Tullin, C.; Potter, A. Atmos. Environ. 2004, 38, 4183–4195.
- (9) Sippula, O.; Hytonen, K.; Tissari, J.; Raunemaa, T.; Jokiniemi, J. Energy Fuels 2007, 21, 1151–1160.
- (10) Tissari, T.; Sippula, O.; Kouki, J.; Vuorio, K.; Jokiniem, J. Energy Fuels 2008, 22, 2033–2042.
- (11) Brunner, T.; Obernberger, I.; Jöller, M.; Arich, A.; Pölt, P. Behavior of ash forming compounds in biomass furnaces—Measurement and analyses of aerosols formed during fixed-bed biomass combustion. *Proceedings of the Aerosols from Biomass Combustion, International Seminar IEA Bioenergy Task* 32; Zurich, Switzerland, 2001; pp 75—80.
- (12) Lillieblad, L.; Szpila, A.; Strand, M.; Pagels, J.; Rupar-Gadd, K.; Gudmundsson, A.; Swietlicki, E.; Bohgard, M.; Sanati, M. *Energy Fuels* **2004**, *18*, 410–417.
- (13) Sippula, O.; Hokkinen, J.; Puustinen, H.; Yli-Pirila, P.; Jokiniemi, J. Energy Fuels **2009**, 23, 2974–2982.
- (14) Wierzbicka, A.; Lillieblad, L.; Pagels, J.; Strand, M.; Gudmundsson, A.; Gharibi, A.; Swietlicki, E.; Sanati, M.; Bohgard, M. Atmos. Environ. 2005, 39, 139–150.
- (15) Ware, M. Establishing and energy efficiency recommendations for commercial boilers. *Proceedings of the 2000 American Council for an Energy-Efficient Economy (ACEEE) Summer Study on Energy Efficiency in Buildings*; Pacific Grove, CA, 2000; Panel 3. Commercial Buildings: Technologies, Design, and Performance Analysis.
 - (16) Nussbaumer, T. Energy Fuels 2003, 17, 1510-1521.
- (17) Holdemann, L. M.; Cass, G. R.; Markowski, G. R. Aerosol Sci. Technol. 1989, 10, 193–204.

- (18) National Institute for Occupational Safety and Health (NIOSH). NIOSH elemental carbon (diesel particulate) method 5040. *NIOSH Manual of Analytical Methods (NMAM)*, 4th ed.; NIOSH: Atlanta, GA, 1999.
- (19) Turpin, B. J.; Saxena, P.; Andrews, E. Atmos. Environ. 2000, 34, 2983–3013.
- (20) Raman, S. R.; Hopke, P. K.; Holsen, T. M. Environ. Monit. Assess. **2008**, 144, 351–366.
 - (21) Simoneit, B. R. T. Appl. Geochem. 2002, 17, 129–162.
- (22) Holsen, T. M.; Hopke, P. K.; Pagano, J. J.; Milligan, M. S. Chemical Analysis of the Great Lakes Fish Monitoring Program (GLFMP); United States Environmental Protection Agency (U.S. EPA): Washington, D.C., 2008; EPA Contract GL 96594201.
- (23) Christensen, K. A.; Livbjerg, H. Aerosol Sci. Technol. 1996, 25, 185–199.
- (24) Lightly, J. S.; Veranth, J. M.; Sarofim, A. F. J. Air Waste Manage. Assoc. 2000, 50, 1565–1618.
- (25) Fine, P. M.; Cass, G. R.; Simoneit, B. R. T. Environ. Sci. Technol. **2001**, 35, 2665–2675.
- (26) Mazzoleni, L. R.; Zielinska, B.; Moosmüller, H. Environ. Sci. Technol. 2007, 41, 2115–2122.
- (27) Schauer, J. J.; Kleeman, M. J.; Cass, G. R.; Simoneit, B. R. T. *Environ. Sci. Technol.* **2001**, *35*, 1716–1728.
- (28) Simoneit, B. R. T.; Schauer, J. J.; Nolte, C. G.; Oros, D. R.; Elias, V. O.; Fraser, M. P.; Rogge, W. F.; Cass, G. R. *Atmos. Environ.* **1999**, 33, 173–182.
- (29) Lavric, E. D.; Konnov, A. A.; Ruyck, J. D. Biomass Bioenergy **2004**, 26, 115–145.
- (30) Braida, N. P. D. Workshop on Energy Efficiency Measurements and Validation Methodologies for Pilots; Habitat et Territoires Conseil (eSESH Project): Paris, France, 2010; http://ict-sustainablehomes.org/wp-content/plugins/alcyonis-event-agenda//files/eSESH_N_Pilat.pdf (accessed on Sept 23, 2011).
- (31) Omara, M.; Hopke, P. K.; Raja, S.; Holsen, T. M. Energy Fuels **2010**, 24, 6301–6306.