Research paper

Comparative emissions characterization of a small-scale wood chip-fired boiler and an oil-fired boiler in a school setting

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ABSTRACT

To investigate the differences in emissions performance between wood chip and oil-fired boilers under real-world operating conditions, a series of stack tests were performed to measure air pollutant emissions at a wood chip stoker boiler with multi-cyclone emission controls. This type of wood combustion boiler technology (inclined grate stoker boiler with a heat input rating between 880 and 4700 kW) represents the largest number of installations in the northeastern United States. The monitored air pollutant stack emissions from the wood chip boiler were compared to those from an oil-fired boiler used as backup at the same location. The stack test results found that total particulate matter emissions from the wood-fired boiler with a multi-cyclone emission control device were almost 30 times higher on a mass per energy input basis than the oil-fired boiler burning No. 2 distillate oil with 2.2 g kg^{-1} sulfur content under similar real-world operating conditions.

1. Introduction

Under the U.S. Clean Air Act, small combustions boilers are regulated for air pollution control purposes as "area sources" if they emit or have the potential to emit less than 9070 kg per year of a single "hazardous air pollutant," or less than 22,700 kg per year of combined hazardous air pollutants (see slide 4 in Ref. [1]). The specific "hazardous air pollutants" are listed under the U.S. Clean Air Act, and examples include benzene and a number of metal compounds. Small boilers also emit other regulated pollutants not listed as hazardous air pollutants, such as nitrogen oxides (NOx), carbon monoxide (CO), sulfur oxides (SOx), volatile organic compounds (VOC), and particulate matter (PM).

In the United States, there are approximately 1.3 million boilers installed at area sources such as industrial, commercial, and institutional settings. Within this universe of boilers, an estimated 53% are installed in institutional settings (e.g., hospitals, schools, churches, government buildings), 47% are installed in commercial settings (e.g., shopping malls, apartment buildings, hotels), and less than 1% are located in industrial settings (e.g., manufacturing, refining, mining) (see slide 14 in Ref. [1]).

The U.S. Environmental Protection Agency (U.S. EPA) sets specific air pollution control requirements, such as operational practices or numerical emission standards, for different types and sizes of area

source boilers [2]. For area source boilers subject to numerical emission standards, air pollutant emissions are evaluated through stack emissions testing conducted according to established U.S. EPA test methods. PM emissions from oil-fired and wood-fired area source boilers are tested according to the same test methods (see Table 4 in Ref. [2]). For regulatory compliance purposes, the test methods are applied under high-load steady-state conditions, and the emissions testing is typically required to be conducted while the air pollution source is operating at its maximum operating load (see Table 6, Item 4 in Ref. [2]). We note that the U.S. PM emission limits are set on an emissions per energy input basis, although if expressed in terms of emissions per unit of energy output, operation under part load may have higher emission rates due to poorer combustion conditions.

For oil-fired boilers, the maximum operating load typically produces the greatest gross amount of emissions. Gross emissions of wood-fired boilers, however, typically are higher under low-load operating conditions rather than at maximum load. Therefore, the high-load steady-state test conditions used for determining compliance with the numerical emission limits may not well represent "typical use" operating conditions for area source wood-fired boilers. This has also been seen with advanced residential wood stove technologies where measured air emissions under real-world operating conditions were higher than observed under standard test conditions [3,4].

Higher total emissions under typical operating conditions that are

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Table 1
Load conditions and fuel characteristics during individual test runs.

Test Run	Load Conditions	Fuel Characteristics
Bole Chip Run#1	• low load (35–43% capacity) for entire run	12.4 MJ kg^{-1}, 0.85% ash, 41% moisture
Bole Chip Run#2	• high load (70% capacity) for 101 min	12.1 MJ kg^{-1}, 0.85% ash, 42% moisture
	• medium load (51%–69% capacity) for 150 min	
Bole Chip Run#3	• low load (35–48% capacity) for entire run	12.6 MJ kg^{-1}, 0.78% ash, 43% moisture
Mill Chip Run#1	• high load (70% capacity) for 101 min	11.6 MJ kg^{-1}, 0.45% ash, 45% moisture
	• medium load (54%–69% capacity) for ~162 min	
Mill Chip Run#2	• high load (70% capacity) for ~216 min	12.1 MJ kg^{-1}, 0.53% ash, 43% moisture
	• medium load (62%–69% capacity) for 90 min	
Mill Chip Run#3	• medium load (53%–69% capacity) for ~63 min	11.8 MJ kg^{-1}, 0.41% ash, 44% moisture
	• low load for ~160 min	

Table 2
Listing of test methods applied in wood chip and oil boiler emissions testing.

Test Method	Description
US EPA Method 1	Sample and Velocity Traverses for Stationary Sources
US EPA Method 2	Determination of Stack Gas Velocity and Volumetric Flow Rate (Type S Pitot Tube)
US EPA Method 3A	Determination of Oxygen and Carbon Dioxide Concentrations in Emissions from Stationary Sources (Instrumental Analyzer Procedure)
US EPA Method 4	Determination of Moisture Content in Stack Gases
US EPA Method 5	Determination of Particulate Matter Emissions from Stationary Sources
US EPA Method 6C	Determination of Sulfur Dioxide Emissions from Stationary Sources (Instrumental Analyzer Procedure)
US EPA Method 7E	Determination of Nitrogen Oxide Emissions from Stationary Sources (Instrumental Analyzer Procedure)
US EPA Method 10	Determination of Carbon Monoxide Emissions from Stationary Sources (Instrumental Analyzer Procedure)
US EPA Method 18	Determination of Gaseous Organic Compound Emissions by Gas Chromatography
US EPA Method 0010	Modified Method 5 Sampling Train for Measurement of Semivolatile Compounds
US EPA Method 25A	Determination of Total Gaseous Organic Concentration Using a Flame Ionization Analyzer
US EPA Method 26	Determination of Hydrogen Halide and Halogen Emissions from Stationary Sources
US EPA Other Test Method 28	Dry Impinger Method for Determining Condensable Particulate Emissions from Stationary Sources
CARB Method 430	Determination of Formaldehyde and Acetaldehyde in Emissions from Stationary Sources

Table 3
CEMS specifications.

Calibration Error	Less than ± 2% of span for the zero, mid-and hi-range calibration gases
System Bias	Less than ± 5% of span for the zero, mid-or hi-range calibration gases
Zero Drift	Less than ± 3% of span over the period of each test run
Calibration Drift	Less than ± 3% of span over the period of each test run

not captured by high-load emissions compliance test methods have public health implications as wood smoke is known to contain a number of health-damaging pollutants, including several carcinogens [5,6], with sensitive subpopulations, such as the elderly, being at higher risk [7]. Because installations of wood-fired boilers are occurring at locations with local populations that can be more sensitive to emitted air pollutants, such as schools and hospitals, a better understanding of the real-world performance of these technologies is needed to provide greater information on their local health impact implications. This is particularly salient in light of advanced wood combustion systems increasingly being promoted as low-carbon renewable fuel technologies as well as an emerging market for the use of locally-produced biomass [8], with significant opportunities in highly populated areas [9]. The positive aspects of these efforts can be undermined if operational practices associated with wood fuel combustion result in increased human exposures to higher concentrations of health-damaging air pollutants [10,11].

To further investigate the differences in emissions performance between wood chip and oil-fired boilers under real-world operating conditions, a series of stack tests was conducted in this study to measure air pollutant emissions at a wood chip stoker boiler with multi-cyclone emission controls installed at a school location in the northeastern United States. This type of wood boiler (inclined grate stoker boiler with a heat input rating between 880 and 4700 kW) represents the largest number of installations in this region. Two types of wood chips were tested, mill end and bole, with moisture mass fraction ranging

from 41% to 45%. The monitored air pollutant stack emissions from the wood chip boiler were compared to those from a distillate oil-fired boiler burning No. 2 distillate oil with 2.2 g kg^{-1} sulfur content used as backup at the same school setting. We also compare the wood chip stoker boiler PM emissions to reported results from other wood chip boiler technologies installed in the northeastern U.S. and in Europe.

2. Material and methods

We measured air pollutant emissions from a 2050 kW wood chip stoker boiler with a multi-cyclone for emission control installed for primary heating purposes at a northeastern U.S. school setting. The wood boiler is a Messersmith combustor coupled to a Hurst Model SI-265 boiler that typically consumes approximately 725 t of wood per year at the school. The combustor is an inclined grate stoker with two augers to feed wood chip fuel to the top of the grate. There are five zones of primary combustion air (under-fire air), and a refractory shelf that separates the primary combustion zone from the boiler. As unburned wood gases leave the primary combustion zone, secondary combustion air (over-fire) is introduced to the gases so the combustion is completed in the combustion chamber of the Hurst boiler. The Hurst boiler is a fire tube unit with a three-pass design.

The combustion control system employs a step modulated fuel feed rate, having three stoking rates, plus a pilot mode in which stoking occurs intermittently at the lowest rate. The low fire, medium fire, and high fire stoking rates are manually adjustable by potentiometers.

Table 4
Individual test run results and three-run averages of total, filterable, and condensable PM emissions rates for stoker boiler combusting 40% moisture mass fraction wood chips and boiler combusting heating oil. Rates in mg MJ^{-1} are on energy input basis.

	Run 1	Run 2	Run 3	Average
Bole Chip Test Load	low	high to medium	low	–
Total PM (g h^{-1})	419.6	1032.4	546.6	666.2
Filterable	393.7	1003.8	517.1	638.2
Condensable	25.9	28.6	29.5	28.0
Total PM (mg MJ^{-1})	99.7	159.4	114.3	124.4
Filterable	93.7	155.1	108.3	119.0
Condensable	6.0	4.3	6.0	5.4
Mill End Chip Test Load	high to medium	high to medium	medium to low	–
Total PM (g h^{-1})	551.1	695.8	250.4	499.1
Filterable	467.7	676.8	225.0	456.5
Condensable	83.5	19.1	24.9	42.5
Total PM (mg MJ^{-1})	97.1	155.9	84.2	112.4
Filterable	82.5	151.6	75.6	103.2
Condensable	14.6	4.3	8.6	9.2
Heating Oil Test Load	medium	medium	high	–
Total PM (g h^{-1})	17.2	9.5	11.3	12.7
Filterable	13.6	5.9	6.4	8.6
Condensable	3.6	4.1	5.0	4.2
Oil Total PM (mg MJ^{-1})	6.0	3.4	3.0	4.2
Filterable	4.7	2.1	1.7	2.9
Condensable	1.3	1.3	1.3	1.3

Table 6
Measured emission rates of NOx, CO, SO$_2$, and VOC from wood chip and oil boilers (mg MJ^{-1} energy input).

	Bole Chip	Mill End Chip	No. 2 Distillate Oil, 0.22% Sulfur
NOx	124.2	126.0	56.7
CO	193.0	193.9	0.4
SO$_2$	15.0	5.6	92.9
VOC	21.1	6.4	0.4

oxygen, hydrogen, nitrogen, sulfur, trace elements including metals, ash and moisture content, and heating value. The chip samples were finely ground and pressed into pellets for analysis. Standard ASTM test methods were applied, which were ASTM E870-82 for preparation [12], ASTM E711-87 for calorific content [13], ASTM D1102–84 for ash content [14], and ASTM E871-82 for moisture analysis [15] Basic elemental analysis was conducted using ASTM methods D-3176 modified (ICP-MS) [16]. Trace elements in the wood chip samples were determined from a sample of ash. A wood sample was placed in a muffle furnace to ash the sample. The ash samples were then acid digested in nitric acid using a CEM MARS 5 microwave digestion system. The resulting solution was filtered and analyzed using a Thermo X-Series ICP-MS with collision cell technology (CCT). Only relevant elements that had detectable levels in most samples are reported. Variability across samples was large, with standard deviations approaching or exceeding the averages. The wood chip characteristics used in this study are provided in Table 1, with additional details of test results in the Supplementary Material (Tables S1–S5). We note that moisture mass fraction ranged from 41 to 43% for the bole chips and 43–45% for the mill chips, which was at or below the upper end of the woodchip boiler manufacturer's recommended maximum content of 45%.

A sample of No. 2 distillate oil was obtained during the testing for analysis of trace metal and sulfur content by a certified and accredited testing laboratory (Intertek). Oil samples were shipped to the Intertek laboratory in Deer Park, TX for analysis. Sample chain of custody was maintained such that metals analyses and sulfur results could be tracked to each sample. Trace element analysis other than sulphur was done by ICP-MS using the laboratory's standard test method (ITM_1051), with minimum detection levels between 1 and 10 μg kg^{-1} depending on the element. Oil sulfur content was tested using energy-dispersive X-ray fluorescence spectrometry according to ASTM Method D4294 [17]. This method is typically applied to a concentration range of 150 to 50,000 mg kg^{-1} sulfur in fuel oils. Sulfur content averaged

Programmable logic controllers automatically select the proper fuel feed rate by differencing the boiler supply water temperature and the supply water temperature operating point.

The wood boiler was fired with two types of wood chips – mill end and bole. Mill end chips have bark removed before chipping while bole tree chips do not. The bole chips underwent processing to remove "fines" (particles less than ∼ 3 mm in size) prior to delivery to the testing site.

Wood chip samples were collected at the school site and sent to an analytical laboratory at Clarkson University (Potsdam, New York) operated by Prof. Phil Hopke. Wood sample analysis included carbon,

Table 5
Comparison of U.S. particulate matter regulatory standards (gray-shaded rows) with measured emission rates at wood combustion and oil-fired boilers.

Boiler type	Fuel	Boiler size (kW)	PM emissions rate[a] (g MJ^{-1})	Control technology
U.S. EPA new small biomass boiler PM standard#1	Biomass	2930–8792	30.1	–
U.S. EPA new small biomass boiler PM standard#2	Biomass	≥8792	12.9	–
School stoker, this study	Bole chip	2050	120.4	Multi-cyclone
School stoker, this study	Mill end chip	2050	112.6	Multi-cyclone
Gammie [20], school#1 gasification combustion	Mill end hardwood chip	2667	67.9	Multi-cyclone
Gammie [21], school#2 gasification combustion	Mill end hardwood chip	1348	28.4	Multi-cyclone
Chandrasekaran et al. [22] Walker Center gasification staged combustion	Pellet	146	25.8	Mono-cyclone
Chandrasekaran et al. [22] Cayuga gasification staged combustion	Wood chip residue	146	41.3	Mono-cyclone
Chandrasekaran et al. [22] Wild Center gasification staged combustion	Pellet	498	26.2	Mono-cyclone
Sippula et al. [23] "boiler A" rotating grate	Sawdust + bark	5011	34.8	Multi-cyclone, wet scrubber
Sippula et al. [23] "boiler B" rotating grate	Wood chips (forest residue)	15,005	15.9	Multi-cyclone, ESP
Sippula et al. [23] "boiler C" rotating grate	Sawdust + bark	9994	34.8	Multi-cyclone, ESP
Sippula et al. [23] "boiler D" gasification combustion	Wood chips (forest residue)	7004	52.0	Mono-cyclone
U.S. EPA new oil boiler	Oil	2930	12.9	–
School oil boiler, this study	No. 2 distillate oil 2.2 g kg^{-1} sulfur	1170	3.44	none
McDonald [24] conventional cast iron boiler	No. 2 distillate oil 11 mg kg^{-1} sulfur	58.6	0.026	none
McDonald [24] conventional warm air furnace	No. 2 distillate oil 11 mg kg^{-1} sulfur	29.3	0.056	none

[a] Emission rates are based on energy input. For boilers with installed control technologies, emission rates are post-control.

2.2 g kg^{-1}, which is consistent with a 2.0 g kg^{-1} average sulfur content in a larger 2010 regional analysis of No. 2 distillate oil sold in the northeastern U.S [18]. Trace metal content was also consistent with the 2010 distillate oil regional analysis. Full results are presented in the Supplementary Material (Table S6).

Three test runs for each fuel type were carried out, for a total of nine runs (three mill chip, three bole chip, and three distillate oil). A listing of U.S. EPA and other test methods used for the combustion species measured during the test runs is provided in Table 2.

Air emission sampling for the wood chip boiler was performed using the existing 61 cm internal diameter vertical stack (Supplemental Material Fig. S1). Two 15 cm diameter sampling ports were located 90° apart approximately 2 m above the school roof (~6 stack diameters downstream of the nearest flow disturbance), and a second tier of two 15 cm diameter ports were installed approximately 2.5 m above the lower pair. During each run of approximately 4 h in total duration, combustion species were measured in the stack with six sampling trains that were switched in and out at the four ports according to the species being measured, typically in periods of 2 h or less. A single sampling port was installed approximately 1 m below the existing ports for use with a continuous emission monitoring system (CEMS) probe. The CEMS probe blocked less than 3% of the stack area and therefore did not represent a flow disturbance. Additional details on sampling train arrangements and schematics are given in Rector et al. [19].

Emissions from the wood chip boiler performance testing were compared to a conventional distillate oil-fired boiler with no emission control equipment (which is typical for conventional oil-fired boilers burning distillate in the U.S.) that serves as a backup to the wood chip boiler at the same location. The 1170 kW boiler was manufactured in 1993 by Industrial Combustion, Inc. (Highland Automatic Boiler Model 508-5), and installed at the school location in 2002 to serve as the primary heat source at the school, but was replaced by the woodchip boiler in 2007.

Sampling for the oil boiler was performed using a temporary horizontal stack that was set up to satisfy the requirements of EPA sampling Test Method 1. This stack was 38 cm square. Two sets of 10 cm diameter sampling ports were installed that were accessible from the school roof. Each set of sampling ports was located ~8 equivalent duct diameters downstream of the nearest flow disturbance.

During testing, the wood chip boiler operated under normal conditions and under a variety of typical loads. Fuel load varied from high to low throughout the testing periods. Table 1 provides the load conditions and fuel characteristics during each wood chip boiler test run. Load on the oil boiler during testing ranged from medium to high.

The CEMS was used to quantify nitrogen oxides (NOx), sulphur oxides (SOx), carbon monoxide (CO), volatile organic compounds (VOC), O_2, and CO_2 emissions. The system consisted of a heated stainless-steel probe of sufficient length to allow a three-point traverse during sampling and a heated Teflon® sample line (0.95 cm OD, 0.016 cm wall) to transport the sample gas from the probe to the sample gas conditioning system. The CEMS met or exceeded all specifications listed in Table 3.

The sample gas conditioning system consisted of an in-stack sintered filter located at the probe inlet to remove particulates from the sample gas stream, and a thermoelectric condenser/electronic chiller sample gas conditioning system for moisture removal. A diaphragm-type vacuum pump was used to draw the sample gas from the probe through the conditioning system to the analysers. The pump head was stainless steel, the valve disks were Viton® rubber, and the diaphragm was Teflon® coated.

A three-way fitting, located at the base of the probe, was utilized to allow the operator to select sampling from either the sample stream or the calibration gas stream. The sample or conditioned gas was distributed to the analysers using a series of flow meters, valves, and backpressure regulators. These allowed the operator to maintain constant sample flow and pressure during sampling and calibration.

NOx was measured in accordance with US EPA Method 7E using a Thermo Environmental Instruments Inc. (TEI) Model 42H NOx chemiluminescent monitor in a range from 0 to 500 cm^3 m^{-3} dry basis NOx. SOx was measured with a continuous emissions analyzer according to US EPA Method 6C, and set at a range of 0–250 cm^3 m^{-3} dry basis SO_2. CO was measured according to US EPA Method 10 using a TEI Model 48 gas filter correlation infrared analyzer set at a range of 0–1000 cm^3 m^{-3} CO. A VOC gas sample was continuously extracted from the exhaust stack through a heated Teflon sample line routed to a TECO 51 flame ionization analyser in accordance with US EPA Method 25A. VOC measurements were expressed in equivalents to the calibration gas, propane. Calibration was done with three concentrations of propane in a balance of high purity nitrogen: 85.8, 50.1 and 30.4 cm^3 m^{-3} as propane. O_2 and CO_2 were measured according to US EPA Method 3A using a California Analytical Model 100F O_2 analyzer with a range of 0–25% volume fraction for O_2, and a California Analytical Model ZRH non-dispersive infrared CO_2 analyzer with a range of 0–20% volume fraction for CO_2.

A PC-based data logger system with digital strip chart recorder (Monarch Instrument DataChart 4600) was used to record analyser responses to the sample and calibration gas streams. The chart recorder was operated continuously while the personal computer recorded the test run data in 15-s interval averages.

A NOx and O_2 stratification test was completed prior to testing. The eight traverse points determined by U.S. EPA Method 1 were sampled to ensure no deviation greater than 10% of the mean concentration at any point (or greater than 3 mg kg^{-1} NOx or 0.3% O_2 absolute difference). The CEMS sampling was conducted at a single point due to the lack of stratification.

Raw data averages collected for each pollutant or diluent gas concentration for each test run were determined by exporting the collected data from the data logger into an Excel™ spreadsheet file. The raw data were then corrected to determine the final concentrations by the calibration drift and bias test results.

For the stack PM measurements, filterable PM is defined as emitted directly by the boiler as a solid or liquid at stack conditions and captured on the filter of the stack test train. Condensable PM exists in the vapor phase at stack conditions, then condenses and/or reacts upon cooling and dilution in the ambient air to form solid or liquid PM immediately after discharge from the stack. For both filterable and condensable PM sampling, filters were collected during each of three 60 min test runs. Collected filterable and condensable PM samples from each run were sent to a third-party commercial laboratory (Maxxam Analytics, Inc.) for analysis.

Total filterable PM was quantified in the front-half of the sampling train according to US EPA Reference Test Method 5. Total condensable PM was measured in the back-half of the sampling train according to US EPA Other Test Method 28. The front-half for sampling filtered PM consisted of a glass button hook nozzle, a heated glass lined sample probe, and a tared glass fiber filter in a holder in an oven box. The sample probe and oven box were maintained at a temperature greater than 120C during sampling to prevent moisture condensation. After a post-test leak check, the front-half of the sampling train was disassembled, all open ends were sealed, and the sampling train components were moved to a cleanup area for recovery. The filter was carefully removed with forceps and placed in a labeled plastic petri dish. Any particulate matter or filter fragments that adhered to the filter holder gasket were also transferred to the petri dish. The petri dish was then sealed for laboratory analysis. In addition, the nozzle, probe liner, cyclone bypass, and filter holder front half were rinsed and brushed three times (or until there was no discoloration in the rinse) with acetone. These rinses were collected in a labeled glass or Nalgene® sample jar. The sample jar was sealed with the liquid level marked for laboratory analysis.

The back-half sampling train for condensable PM collection consisted of a condenser and a condensate dropout impinger without

bubbler tube following the out-of-stack filterable PM heated filter assembly. A modified Greenburg Smith impinger with no taper followed and served as a backup dropout impinger. The first two impingers were placed in an insulated "cold box" containing water at less than 30C which was recirculated through the condenser during sampling. The condenser was capable of cooling the stack gas to less than 30C. The condensable PM filter holder containing a Teflon® filter followed and was equipped with a thermocouple measuring the temperature of the sample gas. The temperature of the sample gas was maintained at or below 30C. A modified Greenburg Smith impinger containing 100 cm^3 of water followed, serving as the moisture trap to collect the moisture that passed through the condensable PM filter. A fourth impinger contained a known quantity of indicating silica gel. The temperature at the outlet of the fourth impinger was maintained below 20C during sampling by an ice bath containing the third and fourth impingers. A vacuum line connected the outlet of the fourth impinger to a control module consisting of a leak free sampling pump, a calibrated critical orifice, an inclined manometer, and a calibrated dry gas meter. As soon as possible following the post-test leak check, the front-half of the sampling train was removed from the back-half, and the back-half was configured for a post-test nitrogen purge. If no visible water or condensate was collected before the condensable PM filter (in the condensate dropout impinger or backup dropout impinger) the nitrogen purge cycle was eliminated. The CPM filter sample was carefully removed from the filter holder with forceps and placed in a labeled plastic petri dish. Any particulate matter or filter fragments that adhered to the filter holder gasket were transferred to the petri dish using the forceps. The petri dish was then sealed for transport to the analytical laboratory.

The aqueous liquid contents of the dropout and backup dropout impingers were quantitatively transferred into a clean glass or plastic sample bottle. The probe extension, condenser, each impinger and the connecting glassware, and the front-half of the condensable PM filter holder were rinsed twice with water. The rinse water was recovered into the same bottle as the impinger contents and the liquid level marked. This sample container held the water soluble condensable PM captured in the sampling train.

Following the water rinses the sampling train components were rinsed with acetone, then twice with methylene chloride. These solvent rinses were collected in a glass sample bottle with the liquid level marked. This sample container held the organic condensable PM sample fraction captured in the sampling train.

The condensable PM sample fractions were maintained at or below 30C during overnight delivery transport to the analytical laboratory.

3. Results and discussion

3.1. Particulate matter emission rates from the combustion of wood chips and heating oil

Table 4 provides a summary of the filterable and condensable results of the PM testing for each test run and the average across the three runs for each fuel type. On average, total PM emissions on an energy input basis from the bole and mill end chips were about 30 times higher than those of the oil test at the same location. Review of the filterable PM versus the condensable PM shows similar results between the bole and mill end chip fuels. Approximately 8% of the PM in the mill chip was emitted in the condensable fraction, while 5% of the PM in the bole chips was emitted in the condensable fraction. The condensable PM fraction from the oil boiler was significantly higher than that from the wood chips, with 37.5% of the PM found in the condensable fraction. We would expect the condensable PM fraction from oil combustion to be lower than from the wood chip boiler due to higher combustion temperatures in the oil boiler. We are uncertain for the higher relative condensable fraction with oil in this study, but note that the oil boiler is used as backup to the chip boiler, and may not have been maintained for optimal operation.

Table 5 compares the PM emission results from this study to results appearing in previously published studies. Included in the table are results from two close-coupled gasification boilers burning mill end chips with high efficiency multi-cyclone for PM emission control located at two different school locations in the northeastern U.S. [20,21]; three different wood gasification staged combustion boilers at different institutional settings also in the northeastern U.S. [22]; wood chip combustion results at district heating systems in Europe [23]; and two conventional oil devices burning 11 mg kg^{-1} sulfur No. 2 distillate oil [24]. Table 5 also includes for comparison more recent PM emission standards established by the U.S. EPA after the completion of these stack tests that would apply to new wood- and oil-fired boiler installations in the United States.

As seen in Table 5, wood-fired boilers equipped with ESPs tend to reduce PM emissions by approximately 90% while units using multi- or mono-cyclone technologies only reduce PM emissions by 10% [25], although the PM results of the close-coupled gasification mill end chip boiler #2 with a high-efficiency multi-cyclone compare well to ESPs.

Hopke and Holsen [26] concluded that without emission controls, staged combustion wood chip boilers burning 30% moisture mass fraction wood chip (much lower than typically available) could achieve PM emissions of 47.3 mg MJ^{-1}. The study concluded that staged combustion systems in the U.S. represent a significant improvement over the stoker design. The study's staged units, however, did not represent state-of-the-art in boiler design, and therefore additional opportunities may exist to improve combustion efficiency with the potential of lower emissions and higher thermal efficiencies.

Lowest PM emissions in this study were observed with the oil-fired boiler burning distillate (No. 2) fuel oil with 2.2 g kg^{-1} sulfur content.

3.2. Other emission rates from the combustion of wood chips and heating oil

The CEMS results for emission rates of NOx, CO, SO$_2$ and VOC are shown in Table 6 for the wood chip stoker and oil boiler measurements. The bole chip and the mill end chip results showed fairly similar NOx emission rates of 124.2 mg MJ^{-1} for the bole chips and 126.0 mg MJ^{-1} for the mill end chips. The NOx emission rate for the oil boiler was 56.7 mg MJ^{-1}, which was 55% lower than the wood chips.

For CO, the results showed fairly similar emission rates of 193.0 mg MJ^{-1} for the bole chip and 193.9 mg MJ^{-1} for the mill end chip. The 2.2 g kg^{-1} S distillate heating oil CO emission rate was significantly lower at 0.4 mg MJ^{-1}. The CEMS data collected over the course of several days indicated that CO emissions from the wood chip boiler are highly variable and dependent on load, state of operation, fuel moisture content, and air to fuel ratios. Emission rates varied from levels less than 10 mg kg^{-1} to more than 1.5 g kg^{-1}. We reviewed CO rates in comparison to load or changes in wood chip boiler operation and did not find any correlation between these rates. Based on these results for the wood chip boiler, we conclude that current U.S. regulatory testing requirements, which only capture CO emissions at high load for a 60 min time period, are not likely to present a realistic picture of typical performance.

The SO$_2$ emission rates based on the CEMS data for the all three fuels were fairly consistent across various sampling periods. Mill end chip had the lowest emission rate (5.6 mg MJ^{-1}). The SO$_2$ emission rate for bole chip was 60% higher (15.0 mg MJ^{-1}). In contrast, the oil-fired boiler had a much higher SO$_2$ emission rate of 92.9 mg MJ^{-1}. By 2018, most northeastern U.S. states will limit distillate fuel oil sulfur content to 15 mg kg^{-1}, which should significantly lower SO$_2$ emissions from oil boilers.

The VOC emission rates based on the CEMS data for all three fuel-technology combinations were fairly variable across the different sampling periods. In general, bole chip tended to have the highest emission rate (21.1 mg MJ^{-1}), followed by mill end chip (6.4 mg MJ^{-1}). The VOC emission rate for the oil boiler was significantly lower at 0.4 mg MJ^{-1}.

Table 7
Summary of metals in PM emission results for three fuel types (ND = non-detect; mg MJ^{-1} energy output basis).

	Metric	Bole Chip	Mill End Chip	Heating Oil
Arsenic	g h^{-1}	ND	ND	ND
	g MJ^{-1}	ND	ND	0.0003
Barium	g h^{-1}	0.272	0.363	0.136
	g MJ^{-1}	0.030	0.032	0.012
Beryllium	g h^{-1}	ND	ND	ND
	g MJ^{-1}	ND	ND	ND
Cadmium	g h^{-1}	0.045	0.045	ND
	g MJ^{-1}	0.003	0.002	0.000
Chromium	g h^{-1}	0.045	0.045	0.045
	g MJ^{-1}	0.005	0.004	0.005
Copper	g h^{-1}	0.181	0.136	0.045
	g MJ^{-1}	0.016	0.012	0.003
Lead	g h^{-1}	0.136	0.136	0.045
	g MJ^{-1}	0.014	0.010	0.004
Manganese	g h^{-1}	1.32	2.31	0.045
	g MJ^{-1}	0.124	0.193	0.006
Nickel	g h^{-1}	0.045	0.045	0.045
	g MJ^{-1}	0.003	0.003	0.037
Potassium	g h^{-1}	170.1	157.9	0.862
	g MJ^{-1}	16.9	13.4	0.085
Selenium	g h^{-1}	ND	ND	ND
	g MJ^{-1}	ND	ND	ND
Silver	g h^{-1}	ND	ND	ND
	g MJ^{-1}	ND	ND	0.0002
Zinc	g h^{-1}	5.1	2.6	3.6
	g MJ^{-1}	0.537	0.219	0.365

3.3. Trace element emission rates from the combustion of wood chips and heating oil

Results of the emissions testing for trace elements during combustion of bole chip, mill end chip, and distillate heating oil are presented in Table 7. Overall, there was significant variation in trace element emission rates across the three fuel-technology combinations. Stack analysis showed higher emission rates for heating oil than the wood chip fuels for arsenic, nickel and silver. In contrast, the wood chip fuels had higher emission rates for barium, cadmium, chromium, copper, lead, manganese, and potassium. Mercury was not tested, but a wood chip fuel elemental analysis found that commercially sold wood chips in New York State and Vermont purchased during the time of this study contained 2.5 µg kg^{-1} of fuel [19]. This can be compared to an average mercury content of 2.0 µg kg^{-1} in No. 2 distillate oil sold in the northeastern U.S. [18], and noting that No. 2 distillate oil has about three times higher energy content than wood on an equivalent mass basis. This suggests the distillate oil likely emitted lower mercury than the wood chip fuels on an equivalent delivered energy basis for space heating.

When comparing emissions among the two wood chip fuels, bole chips overall tended to have higher emission rates than mill end chips for cadmium, chromium, copper, lead, nickel, and zinc. We note that Sippula et al. [23] measured metal emissions in wood chip-fired boiler stacks before and after installed controls, one with a scrubber and one with an ESP, and reported decreases in the measured metals after the controls. While mercury emissions were not measured, use of advanced pollution controls, such as scrubbers, ESPs, and fabric filters, would likely also capture some portion of the mercury emissions (e.g., particle-bound), as has been seen with coal combustion controls [27].

4. Conclusions

Our results suggest that wood chip stoker boilers with multi-cyclone emission controls (the most common chip boiler type in the northeastern U.S.) installed to replace No. 2 distillate oil boilers will increase local exposure to elevated PM concentrations, which may be of particular concern at institutional settings with sensitive populations, such as schools and hospitals.

CO emissions from the wood chip boiler were highly variable and dependent on load, state of operation, fuel moisture content, and air to fuel ratios. Current U.S. regulatory testing requirements only capture CO emissions at high load for 60 min, therefore do not fully capture CO emissions performance under typical woodchip operating conditions.

In general for PM, NOx, CO, SO$_2$, and VOC emissions, only SO$_2$ had higher emissions from the oil combustion compared to the wood chip fuels. We note that sulfur content in No. 2 distillate oil will be reduced to 15 mg kg^{-1} or less by 2018 due to state regulations in the northeastern U.S. Therefore, we expect SO$_2$ emissions from conventional oil-fired boilers and furnaces to decrease significantly from what was measured in this study using No. 2 distillate oil with 2.2 g kg^{-1} sulfur content.

In sum, these results provide for increased awareness of potential human exposures to higher concentrations of air pollutants from wood combustion in settings with sensitive populations, such as schools and hospitals. This can help shape considerations of local health implications from heating technology and fuel use choices, and the additional measures, such as advanced pollution controls, that can help mitigate their impacts.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.biombioe.2017.10.017.

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