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Repeatability in Particulate and Gaseous Emissions from Pellet Stoves for Space Heating

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ABSTRACT: The discussion of emission performance test method repeatability has been a long-standing problem. In March of 2016 the U.S. EPA released a discussion paper on processes for developing improved cordwood test methods for wood heaters. This paper discussed the need to improve repeatability of cordwood fueling protocols and emission measurements to improve confidence in certification processes; however, repeatability for pellet stoves was not addressed. In 2016 the Alliance for Green Heat held the Pellet Stove Design Challenge which was created to test, study, and assess high-efficiency, low-emission pellet stoves. During this workshop, three pellet stoves were selected for extensive testing which provided results on their repeatability to one single test method in the U.S. in terms of emission rates, emission indices, and efficiency. The goal of this study was to assess the repeatability of a single certification test method for pellet stoves and the emission data used for certification purposes. Overall, the repeatability of the test method was favorable with variation in the overall burn rate (kg/h) of 4%, 2%, and 3% for stoves 1, 2, and 3, respectively. The variability in emission rate (g/h) for stoves 1, 2, and 3 was 4%, 17%, and 65%, respectively, which raised concerns for regulatory purposes. However, the concern was met with suggestions for test method development which may include sampling during higher emission periods such as start-up or burn out, sampling at higher flow rates, and moving toward higher resolution scales. Specifically, including start-up or burn out periods which are typically "poorer combustion periods" is more reflective of in-field use and a better measure of the actual stove's performance. This work also provided support for EPA's New Source Performance Standard requirement for manufacturers to report CO emissions and efficiency.

1. INTRODUCTION

Pellet stoves are an increasingly popular way for homeowners to heat their households. In the period 2005−2014, the use of cordwood and wood pellets for primary residential space heating rose by 33% according to the United States Energy Information Administration.[1] This is in part due to the historically cheaper cost of pellet fuel but also because the technology offers a renewable alternative to fossil fuel heating. Even with home heating oil prices falling to a low of $2.24 in February 2016 in New York State,[2] pellet stoves are still considered an integral part in the state's energy picture. Figure 1 shows the average monthly home heating oil prices (given in cents) in New York State from September 2005 to January 2018, with an increasing trend in the last six months. Strauss points out the increase in heating oil prices in the 2017-2018 heating season should increase the demand for pellets and new pellet appliances.[3] Pellet stoves are space-heating devices that can heat less than an entire home, helping offset significant amounts of home heating oil or propane without the larger investment of replacing a boiler or using more labor intensive cordwood.

The U.S. Environmental Protection Agency (EPA) estimates 88 000 pellet stoves will be sold this year.[4] Residential wood combustion is a major source of ambient air pollution. Wood smoke has become one of the main sources of particulate matter (PM) emissions in ambient air.[5] McDonald measured PM emissions from a wood pellet stove to be 24.9 kg/MJ or approximately 12 times greater than oil-fired boilers using high-sulfur level fuel, 400 times greater than those using ultra-low sulfur (ULS) and 1800 times higher than PM emissions from natural-gas fired boilers.[6] Pellet stoves however are considered to have the lowest particulate emissions of all wood fueled heating systems in the U.S. and the highest efficiency.[6,7] Residential wood-heating devices are now the largest source of PM in New York State, greater than the PM from all diesel vehicles or the electric power sectors.[8] Modern appliances that use pellet fuel can provide a cleaner option than traditional cordwood appliances;[9] however, all wood firing can suffer from nonoptimal conditions, resulting in incomplete combustion and poor emissions.[10]

In 2013 the Alliance for Green Heat (AGH) held a Wood Stove Design Challenge in Washington DC followed by a 2014 workshop which sought to identify and provide recognition for advanced cordwood stove technologies and the development of more representative testing protocols for cordwood stoves.[11] As a sequel to 2013 and 2014 events, AGH created the 2016 Pellet Stove Challenge Design to test, study, and assess high-efficiency, low-emission pellet stoves used for heating. The project had the following objectives: (1) to test pellet stoves and quantify efficiency and emissions performance; (2) to showcase affordable, clean, and efficient pellet stoves at the 2016 Pellet Stove Design Challenge event; (3) to study and assess "next-generation" features that may help with wider deployment of clean and efficient pellet stoves; and (4) to bring together a wide variety of stakeholders to share and disseminate knowledge about pellet stove technology and deployment.

The discussion of test method repeatability has been a long-standing problem.[12−14] In March of 2016 the EPA released a discussion paper on processes for developing improved cordwood test methods for wood heaters.[15] This paper discussed the

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Figure 1. Average monthly heating oil prices in NYS from September 2005 to January 2018.

need to improve repeatability of the cordwood fueling protocol and emissions measurements to improve confidence in certification processes; however, repeatability for pellet stoves was not addressed. One report details the repeatability and reproducibility of cordwood stoves from an EPA accredited laboratory, finding that the repeatability and reproducibility of a wood heater during emission testing is poor. Curkeet and Ferguson found, at the 95% confidence level, that repeatability for the EPA test methods using a catalytic stove with an EPA certification value of 3.1 g/h had at best ±2.9 g/h but ranged to as high as ±5.4 g/h, and the reproducibility for these methods was at best ±4.5 g/h with a high of ±6.4 g/h, method 5G-3 demonstrating the least amount of error.[16] Error in repeatability may stem from the operator, fuel type, the appliance design and controls on combustion conditions, and laboratory measurements. The goal of this study was to assess the repeatability of a single certification test method for pellet stoves and the emission data used for certification purposes. Detailed in this report is the technical evaluation of the efficiency and PM emissions performance of three pellet stoves and the repeatability of one certification test method at within one laboratory.

2. METHODS

2.1. Stoves. The three pellet stoves in this study were selected from an applicant pool based on the Alliance for Green Heat's call for stove manufacturers to compete in the 2016 Pellet Stove Design Challenge. The main characteristics of each of the stoves are presented in Table 1; the reported output is based on the maximum burn rate observed at Brookhaven National Laboratory.

Stove 1 was a Midwestern U.S. multifuel fireplace insert and/or standalone pellet stove. The stove sought a new burn box design to widen the flame to provide a more appealing flame as opposed to the typical intermittent pellet flames. Stove 1 also employed the use of ash reduction filters to reduce particulate matter further. The stove did have a pressure gauge to indicate when the filter needed to be changed.

Stove 2 was a top-feed pellet stove with a unique pellet feeder arrangement.[17] The stove was manufactured in the Northwestern U.S.; therefore, the manufacturer recommended softwood pellets or torrefied pellets for optimal performance.

Stove 3 was a European stove with a dual-fuel system, combining a down draft cordwood stove and a newly developed two-stage pellet burner. The stove was intended to use the pellet burner for days when a homeowner would not be enjoying the aesthetics of a log-wood fire and could operate as a cordwood stove during a power outage or again for the simple aesthetics. The pellet burner pregasifies off on the side of the stove, so there were no visible flames if only the pellet burn was utilized. For comparative purposes, only the pellet burner was operated for this study. Stove 3 also had a proprietary catalyst on the basis of metal oxide foam integrated along its sides so the combustion gas passes through the two-stage combustion design of the stove.

2.2. Fuel. The fuel used in during testing was a Pellet Fuels Institute (PFI) certified premium hardwood–softwood blend pellet purchased by AGH for Brookhaven National Laboratory from Curran Renewable Energy, LLC. Samples were collected from the two pallets delivered to Brookhaven and sent off for analysis by TwinPorts (Superior, WI) test facility for moisture, ash, sulfur, chlorine, and calorific values. Boman et al. reported that studies have shown fuel properties can significantly affect the emissions; therefore, the same brand and batch of pellets was used in all testing.[10] A summary of the higher heating value (HHV), moisture content, and percentage of carbon, oxygen, hydrogen, and ash of the pellets can be seen in Table 2.

Table 2. Summary of Fuel Data

description	value
HHV (kJ/kg)	19 197
%C	48.76
%H	6.87
%O	43.76
%ash	0.64
wood moisture (% wet)	3.77

2.3. Operating Procedure. In all cases the stoves were run in accordance with ASTM E2779-10[18] (with the exception of one modification described below). This method is used for determining average emission

Table 1. Summary Description of Tested Stoves

stove characteristics	stove 1	stove 2	stove 3
output (kW)	4.65	10.2	6.15
control principle	thermostat but can override to manual 1–5 setting	unmarked dial	high-on/off
fuel-feeding principle	horizontal	top	horizontal
preferred fuel type	multi	softwood pellet	dual fuel
ignition type	electrical	electrical	match-lite (electrical anticipated)

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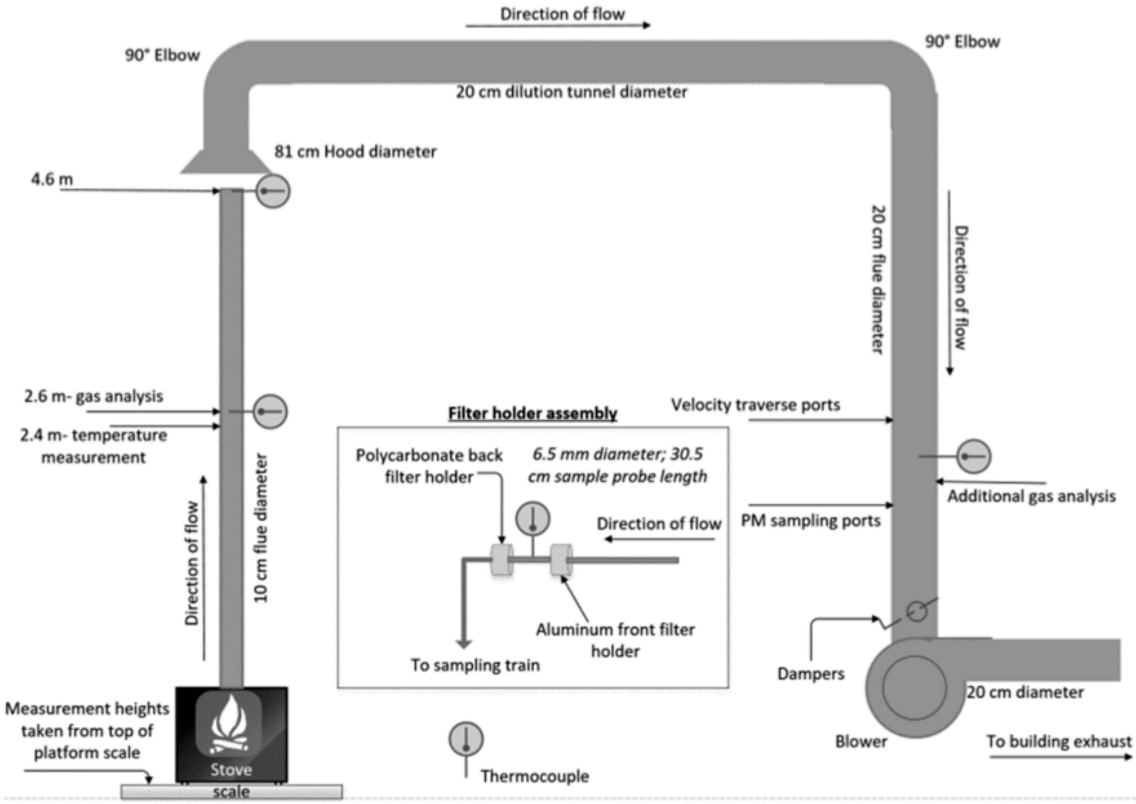

Figure 2. Schematic of the measurement setup from the dilution tunnel.

rates and average emission factors for pellet heaters which is useful for U.S. regulatory communities and determining compliance. This method comprises three categories: maximum burn rate (the highest burn rate a stove can achieve), less than 50% of maximum (medium), and the minimum burn rate the stove can achieve (low). The duration of emission sampling is 60, 120, and 180 min for high, medium, and low, respectively. Prior to emission sampling, the stove must be run for 1 h at its maximum burn rate.

The method specifies sampling particulate emissions in accordance with ASTM E2515-11[19] which uses dual-filter trains sampling at a constant volume in a dilution tunnel. The laboratory setup may be seen in Figure 2. In all cases the exhaust gas was diluted by a factor of 3−12 (based on CO in stack to CO in dilution tunnel) to maintain a flow rate in the dilution tunnel between 150 and 500 SCFM and a sampling temperature between the probes of 32 °C. Sampling in the dilution tunnel allows for the condensation of organic particles in the hot flue gas stream during the dilution with room temperature air. The method uses a composite filter over the entire test, capturing the high, medium, and low burns on one filter.

Many studies have reported different emission characteristics based upon different loads[5,10,20−23] and stressed the importance of including start-up, low load, and burn-out phases when determining a system's emission rates. The interest of this project laid not only on the performance of the stove in different output loads but also on test method repeatability; therefore, start-up and burn-out (or shutdown) phases were not measured since they are not included by the test procedure used in this study. To accurately determine the stove's performance in individual burn phases, a separate filter was used to capture the individual burn phases. Because there were two sampling ports, a dual filter train was set up; one filter train captured a composite of all the burn phases (high, medium, and low) while another had filter changes during the test to capture each individual burn phase. Each pellet stove was run to the prescribed procedure three times resulting in triplicate measurements to allow for a measure of method precision. Each manufacturer was present during testing to ensure their stove was operating correctly; however, once the test started no manufacturer adjustments were allowed in the Pellet Stove Design Challenge event rules.

2.4. Measurement Techniques. Particulate matter (PM) measurements were performed in accordance with ASTM E2515[19] as mentioned earlier. The filters used were 47 mm Teflon-coated glass fiber filers, Emfab filters. (Pallflex Emfab TX40 filters are borosilicate microfibers reinforced with woven glass cloth and bonded with PTFE.) Each sampling train consisted of a front and back filter. Filters were conditioned prior to testing in a desiccator and weighed twice over a two-day period on a gravimetric balance prior to sampling. At the conclusion of each day, each filter used was weighed until its weight stabilized (there was no change in weight within a 6 h period); this value was used as the final weight to determine PM emission values. The gravimetric balance used for stoves 2 and 3 had a range up to 120 g ± 0.02 mg, whereas stove 1 used a gravimetric balance that had a range up to 220 g ± 0.1 mg. The decision to move to a more accurate balance for stoves 2 and 3 was only to improve the accuracy of PM measured and was not owed to stove 1 exceeding the 120 g range. After observation of such a low particulate mass catch on the filters during the 1 h sample period at the high firing rate, a more sensitive balance was needed to accurately quantify the mass loadings. The flow rate of gas was determined using EPA Method 5 trains, one manual instrument model 511 made by Apex Instruments (measurement values were collected every 10 min) and another automatic instrument (logging flow, temperature, ΔP, ΔH, and total volume sampled every minute) model XC-5000. Both samplers used a type K thermocouple for temperature analysis which had an associated error of 2.2° or 0.75%. The automatic sampler and manual sampler had flow rate measurement errors of 1.00×10^{-6} and 3.54×10^{-5} cubic meters and ΔH measurement errors of 0.005 and 0.005, respectively.

Flue gas samples for analysis were taken from the dilution tunnel and directly from the stack, as seen in Figure 2. For sampling from the dilution tunnel, water vapor (7.2 °C dew point) was removed using a thermoelectric cooler/drier and a diaphragm sampling pump. Gas analysis in the dilution tunnel included oxygen and carbon monoxide. Analysis of samples from the flue gas included oxygen, carbon monoxide, hydrocarbons, and carbon dioxide sampled at 2.6 m, in accordance with

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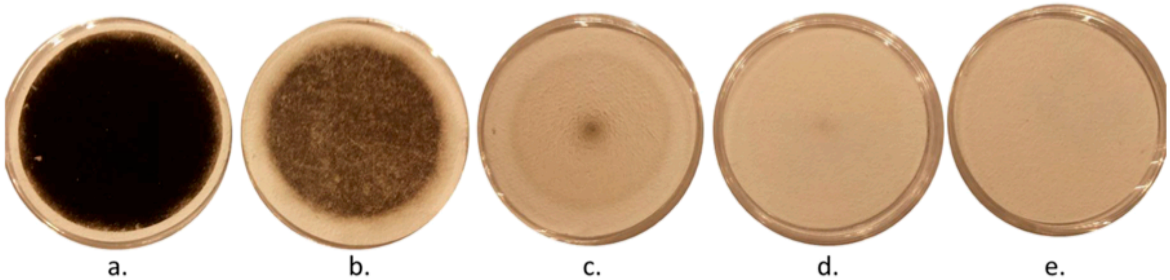

Figure 3. Filters: (a) from a stove with an emission rate of 2.50 g/h, (b) from a stove with an emission rate of 1.4 g/h, (c) from a stove with an emission rate of 0.9 g/h, and (d) and (e) from a stove with an emission rate of 0.56 g/h.

Table 3. Burn Rate, Emission, Efficiency, and Carbon Monoxide Data from All Three Tests Done for Stove 1

first round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
entire	364	0.86	0.92	1.04	62.1	994
high	60	1.32	1.18	0.87	66.8	116
medium	120	0.82	0.97	1.12	60.7	323
low	180	0.73	0.85	1.13	61.0	544
second round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
entire	363	0.82	0.90	1.03	63.9	1562
high	60	1.13	1.35	1.14	64.2	326
medium	120	0.86	0.78	0.86	64.4	560
low	180	0.73	0.79	1.05	63.3	662
third round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
entire	368	0.77	0.85	1.04	62.3	1421
high	60	1.04	2.13	1.95	63.5	254
medium	120	0.82	0.88	1.02	62.0	475
low	180	0.68	0.78	1.11	61.5	659

CSA B415.1-10[24] for stack loss efficiency calculations from the top of the platform scale where the stove was resting. Each gas analyzer was calibrated prior to each test with both nitrogen and the specified calibration gas. All calibration gases were certification grade from Matheson Tri-Gas Co. and provide an accuracy of 2%.

Carbon monoxide was measured in the dilution tunnel using a Rosemount Analytical model 880 NDIR carbon monoxide analyzer. Oxygen was measured in both the dilution tunnel and the hot stack via a Beckman model 755 paramagnetic oxygen analyzer. Both oxygen analyzers have a set of four ranges: 10%, 25%, 50%, and 100%. Carbon monoxide, carbon dioxide, and hydrocarbons were measured directly in the stack using a CAI ZRE infrared analyzer. The signals from all analyzers were logged at 5 s intervals. Each gas analyzer had a resolution of 0.1%. All gas analyzers were recorded on the lab Data Acquisition (DAQ) system using Visual Basic (VB).

Type K thermocouples were also logged at a 5 s interval for all temperature measurements which included the following: stack temperature (2.4 m from stove base; CSA B415.1-10[24] for stack loss efficiency calculations), top of stack (4.6 m), dilution tunnel, ambient air (laboratory), and in-between the front and back filters in each PM sample probe. The thermocouples were purchased from Omega and have an associated error of 2.2° or 0.75%. Thermocouples were logged on a separate computer via Pico Logger software. Velocity in the dilution tunnel was measured with a Pitot tube and digital pressure gauge made by the Pressure Conservatory. A precision Baratron pressure transducer was used to measure a voltage proportional to the pitot tube ΔP and recorded on the lab DAQ system.

Efficiency was calculated for each of the stoves following CSA B415.1-10.[24] In summary, the values required to calculate efficiency include the following: fuel data (moisture on a wet basis, HHV (kJ/kg), % carbon (C), % hydrogen (H), % oxygen (O), and % ash), gaseous emission data in percentages (CO, CO_2, O_2), temperature data (stack and ambient), total particulate emissions captured in the dilution tunnel (grams), the elapsed time of each data entry, and the weight of fuel remaining of each data entry. Each stove sat on a platform scale to measure its weight every five seconds; this provided a direct measure of mass consumption to allow the burn rate of each stove and burn phase to be calculated. The scale was a 5′ X 6.5′ Sartorius Combies model floor scale with a resolution of 0.09 kg and therefore associated error of ±0.045 kg.

3. RESULTS

3.1. Stove 1. Stove 1 had a maximum burn rate of 1.32 kg/h and a minimum burn rate of 0.68 kg/h, on a dry basis. Its overall emission rate (the value reported to EPA for certification purposes) ranged from 0.85 to 0.92 g/h, significantly lower than the current 4.5 g/h limit and 2020 2.0 g/h limit. (In February of 2015, the U.S. Environmental Protection Agency (EPA) released its New Source Performance Standard (NSPS) in order to strengthen its clean air standards. The ruling sought to lower emission standards for residential wood heaters to promote cleaner combustion technologies and improve air quality.[27]). Normalizing the emission data for comparison purposes based on the burn rate provided a value of 1.03−1.04 g/kg, indicating that the stove was very consistent in terms of emission profiles based upon its burn rate. Overall the high burn rate saw the most variability ranging from 1.04 to 1.32 kg/h, while the gap in variability tightened in the medium and low burn rates to 0.05 kg/h difference. Carbon monoxide emission data showed the stove operated in a range of 254−1562 g and an oxygen value in the range of 17.9−18.9%. Further, the overall efficiency of the stove ranged from 62.1 to 63.9%. As mentioned above in the operating procedure, each individual burn rate was also measured to provide detailed emission profile information. A summary from each of the three tests may be seen in Table 3 below, and the averaged data can be seen in Table 4.

3.2. Stove 2. Stove 2 had a maximum burn rate of 2.45 kg/h and a minimum burn rate of 1.09 kg/h, on a dry basis. Its overall

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Table 4. Average Values of Emissions and Efficiency Data for Stove 1

	average of three tests				
burn period	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
overall	0.82	0.89	1.04	62.8	1326
high	1.18	1.55	1.32	64.8	232
medium	0.86	0.88	1.00	62.4	453
low	0.73	0.81	1.10	61.9	622

emission rate (the value reported to EPA for certification purposes) ranged from 2.02 to 2.86 g/h, surely passing the current 4.5 g/h limit but problematic for the 2020 limits. Normalizing the emission data for comparison purposes based on the burn rate provided a value of 1.36−1.98 g/kg, indicating some variation in terms of emissions, even while the burn rate remained very consistent (3.1 vs 1.45 kg/h). Overall the high burn rate saw the most variability ranging from 2.27 to 2.45 kg/h, while the gap in variability tightened in the medium and low burn rates to no difference and 0.09 kg/h difference, respectively. Carbon monoxide data showed the stove operated in a range of 823−7441 g and oxygen values in the range of 16.5−16.8%. Further, the overall efficiency of the stove ranged from 71.1 to 76.5%. A summary from each of the three tests may be seen in Table 5 below, and the averaged data can be seen in Table 6.

3.3. Stove 3. Being in its early prototype phase, stove 3 only had one burn rate, high. Therefore, the medium and low burn rate categories for testing were disregarded, and no entire burn period can be reported. Data from stove 3 were collected during its high burn with both sampling trains at the test method's specified 60 min period but also over an extended time period of approximately 200 min, but with only one sampling train. It was decided to sample for an extended period of time to ensure a sufficient amount of PM was collected on the filter for accurate measurement. The two filter trains each sampled the 60 min period, followed by the automatic sampler sampling the extended time and the manual sampler sampling ambient laboratory air to provide a sense of background PM values.

Stove 3's single, maximum burn rate ranged from 1.27 to 1.59 kg/h on a dry basis. Its overall emission rate (the value reported to EPA for certification purposes) ranged from 0.21 to 1.11 g/h, surely passing both the current EPA limit and 2020 limit. Normalizing the emission data for comparison purposes based on the burn rate provided a value of 0.14 to 0.68 g/kg,

Table 6. Average Values of Emissions and Efficiency Data for Stove 2

	average of three tests				
burn period	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
overall	1.41	2.50	1.69	72.4	6616
high	2.36	4.43	1.80	77.5	1041
medium	1.41	3.07	2.11	70.5	2182
low	1.13	2.47	2.10	69.7	2252

again indicating some variation in terms of emissions. Carbon monoxide data showed the stove operated in a range of 21−105 g and an oxygen value in the range of 11.7−13.1%. Further, the overall efficiency of the stove ranged from 71.6 to 77.3%. A summary from each of the three tests may be seen in Table 7 below, and the averaged data can be seen in Table 8.

4. DISCUSSION

It is common practice for laboratory methods to be evaluated in terms of accuracy (the closeness to the true or standard method value) and precision (the closeness of duplicate results) among other parameters of interest. Pellet fuel burning appliances should have less variability between runs due to the consistency of the fuel type and fuel feed controls compared to cordwood stoves.[25]

The burn rate of stove 1 has good agreement for the entire burn (composite), high, medium, and low burns, and they have coefficient of variation of 4, 11, 3, and 5% respectively as seen in Table 9. The PM emissions rate also shows good agreement with more variability for the high burn. The efficiency also shows good agreement for each of the low, medium, and high burns and low variability. The emissions index shows good agreement in the low and medium burns and more variability in the high burn.

Stove 2 results have more variability in terms of emission rate and emission index as seen in Table 10. Surprisingly, the burn rate had a low amount of variability, 2, 4, 0, and 5% for the entire burn, high, medium, and low burns, respectively, given that that the automatic feed system had failed at one point and the manufacturer was aware the stoves burn rate was never consistent. Stove 2's highest variability occurred during the medium and low burn rates, unlike stove 1 which had its larger variability during the high burn. However, stove 1 was much closer to marketability compared to stove 2 which resembled more of a prototype.

Table 5. Burn Rate, Emission, Efficiency and Carbon Monoxide Data from all Three Tests Done for Stove 2

first round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
entire	365	1.45	2.02	1.36	73.2	5771
high	60	2.45	5.02	1.98	78.5	1458
medium	120	1.41	2.31	1.59	70.9	1738
low	180	1.13	1.85	1.57	70.3	2486
second round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
entire	362	1.41	2.86	1.98	71.7	6636
high	60	2.27	4.44	1.88	76.5	823
medium	120	1.41	3.88	2.67	69.9	2415
low	180	1.09	2.95	2.63	69.0	3999
third round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
entire	364	1.45	2.61	1.73	72.4	7441
high	60	2.36	3.82	1.56	77.6	843
medium	120	1.41	3.03	2.09	70.7	2392
low	180	1.18	2.61	2.10	69.7	4169

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Table 7. Burn Rate, Emission, Efficiency, and Carbon Monoxide Data from all Three Tests Done for Stove 3

first round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
high, sampling train 1	60	1.54	0.21	0.14	75.0	22
high, sampling train 2	60	1.54	0.41	0.26		
extended	203	1.36	0.38	0.25	71.6	66
second round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
high, sampling train 1	61	1.50	0.40	0.26	77.3	21
high, sampling train 2	60	1.50	0.31	0.20		
extended	217	1.27	0.47	0.35	75.6	35
third round testing	duration, min	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
high, sampling train 1	60	1.59	1.11	0.68	73.2	22
high, sampling train 2	60	1.59	0.92	0.56		
extended	217	1.41	0.79	0.54	72.2	105

Table 8. Average Values of Emissions and Efficiency Data for Stove 3

	average of three tests				
burn period	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
high	1.54	0.56	0.35	75.2	21
extended	1.32	0.38	0.38	73.1	69

Table 9. Coefficient of Variation for Stove 1

burn period	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
overall	4%	4%	0%	2%	23%
high	11%	32%	43%	3%	46%
medium	3%	11%	13%	3%	27%
low	5%	5%	4%	2%	12%

Table 10. Coefficient of Variation for Stove 2

burn period	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency, %	CO, g
overall	2%	17%	19%	1%	13%
high	4%	14%	12%	1%	34%
medium	0%	26%	26%	1%	19%
low	5%	23%	25%	1%	25%

Stove 3 also had very good repeatability in terms of burn rate, with a 3% coefficient of variation for the high burn and 5% for the extended run (still at high fire but longer sampling time). The efficiency variation was 3% in each case as shown in Table 11.

Table 11. Coefficient of Variation for Stove 3

burn period	burn rate, dry kg/h	emission rate, g/h	emission index, g/kg	CSA B415.1 efficiency %	CO, g
high	3%	65%	62%	3%	49%
extended	5%	57%	39%	3%	2%

Stove 3's coefficient of variation was large for both the emission rate and emission index, in all cases over 35%. Individual results of pellet fuel burn rate, PM emissions rate, thermal efficiency, and PM emissions index are plotted for each of the triplicate analyses of the composite as well as the high, medium, and low burn tests for each pellet stove.

Carbon monoxide in all three stoves had the largest variation due to the nature of CO; however, the individual stove tests are within good range of each other and may be considered to have good agreement. Cases where the burn rate (kg/h) has a larger variation correspond to larger variation among emission values , such as the high burn period in stove 1. No changes were made to this stove so it is unclear why there was variation in burn rate. For stoves 1 and 2 this is a much tighter agreement than the cordwood test repeatability study done by Curkeet and Ferguson.[16] The high error in stove 3 was expected to be due to the amount of particulate matter collected, which was typically less than 1 mg.

It is important to discuss the large variation seen at the very low emissions levels for stove 3. It is expected the variation is larger due to the small amount of PM collected on the filter (less than 0.50 mg in most cases with the exception of approximately 1.50 mg during the extended burn time). Variability from filter to filter during the same run should be negligible; however, there is some known error in filter handling. As the amount of total mass collected approaches the resolution limit of the scale, more variation is seen. It is still unclear why the amount of PM collected from the first round of testing to the third round of testing nearly tripled.

Current U.S. EPA certification limits are 4.5 g/h for pellet stoves, which all the above stoves meet. However, the 2020 regulations become more stringent at 2.0 g/h which only stove 1 and stove 3 will meet with ease. Currently, pellet stoves on the EPA Burn Wise list range from 0.28 to 4.50 g/h having an average of 1.9 g/h and median of 1.7 g/h.[26]

As we progress toward cleaner stoves, the question of error becomes significant. The error may be associated with too little mass of a catch; sometimes the amount of PM collected is within the resolution of the scale or within the background PM collection (ambient concentration levels during stove 3 testing resulted in an average of 0.05 mg/m^3). Figure 3 below shows the progression of filters from the three stoves presented in this study with above average emission rates, average emission rates, and below average emission rates. It is important to note particle color does not correlate to weight. To eliminate the error of too little mass of PM collected for an accurate measurement, recommendations may include increased sample times to allow for a larger sample of mass to be collected which reduced the coefficient of variation in stove 3 from 65% to 57% and 62% to 39% for the emission rate and emission index, respectively.

One concern this may raise for policy makers and test method developers is the reliability of a single set of data for stove certification, given that a stove may operate cleanly for one run and poorly for another with no change in control settings, fuel type, or other testing conditions. Uncertainty exists with all measurements, and any uncertainty will propagate through calculations and cause some error within the final reported values. From an analysis of the measured uncertainty values in the lab, the largest uncertainty exists when measuring the amount of PM collected. This is owed to the low amount of collected mass as discussed above. It is understood that filter handling for gravimetric sampling

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Energy Fuels 2018, 32, 3543−3550

introduces some weighing artifacts that may be insignificant for heavy filter loadings but can significantly affect mass measurements of filters with low particulate loading. Current methods specify a scale of at least 0.1 mg resolution, but perhaps there needs to be a migration toward a higher resolution to distinguish between stoves. Additional considerations may also include increased sampling rate (greater than the 0.25 cfm allowed) or alternative methods. While sampling directly from the stack would allow for a greater catch, condensable volatiles would then be missed, and this would also introduce error.

Previous to the release of the U.S. EPA's NSPS release in 2015, wood stove manufacturers were not required to provide any efficiency data unless they chose to. If an efficiency value was not reported, the default value was given; for pellet stoves it was 78%.[27] From the testing reported in this work, it is evident each of the three stoves did not achieve even the default value of 78% and actually fell lower. This supports EPA's decision to require manufacturers to report efficiency values for their product.[27]

The emission index (g/kg) provides a direct comparison from stove to stove in terms of particulates captured given the amount of fuel burned as opposed to emission rate (g/h). The emission index value normalizes the data so a stove that has a high firing rate with low emissions may not be penalized in comparison to a stove which has a low firing rate but high emissions. Using the emission rate (g/h) of a stove does not distinguish well enough between a high firing stove with low emissions rate and low firing stove with high emissions rate since the high firing stove with low emissions could have a greater mass emission rate than the low firing rate stove with high emissions rate. The emission index (g/kg) better reflects the quality of the burn as well by taking the burn rate out of the equation. However, it is still important in terms of ambient air quality and exposure to report the emissions rate and therefore should not be dismissed.

Carbon monoxide is reported here for all three stoves because CO is a pollutant and must not be ignored in terms of health. High concentrations of CO can cause serious health issues for children, the elderly, and those with existing cardiovascular illnesses such as asthma or chronic obstructive pulmonary disease (COPD); yet still remains unregulated. It is an odorless and colorless gas that is typically emitted in much higher quantities from wood combustion devices in comparison to oil or gas heating devices. The current ANSI Z21.47 standard (1998), for natural gas fired units, requires that an air-free flue gas sample of CO not exceed an average of 400 ppm when a furnace vent is either blocked entirely or partially.[28] Kinsey et. al report CO readings from 10 000 ppm for a pellet hydronic-heater (40 kW) to as high as 180 000 ppm for an up-draft cord wood hydronic heater (38 kW).[7] Reporting CO allows state and federal regulators to better assess the local, short-term environmental impacts related to combustion. Further, the NSPS will also require manufacturers to report CO values.[27] Table 12 below shows the average CO emissions in terms of ppm for each stove and burn period, indicating only two of the three stoves fall below the 400 ppm ANSI standard.

Table 12. Average CO Emissions (ppm) for Each Stove and Burn Period

	average CO values (ppm)		
burn period	stove 1	stove 2	stove 3
overall	269	1362	69
high	285	1301	22
medium	279	1369	N/A
low	256	1385	N/A

5. CONCLUSION

Three prototype pellet stoves were tested to ASTM E2779-10, in triplicates. Stove 1, 2, and 3's performance averaged over all three tests was 0.9, 2.50, and 0.56 g/h, respectively. These values are all below the current NSPS for PM emissions of 4.5 g/h, and two of the stoves are less than the 2020 standard of 2.0 g/h.[26,27] In terms of PM emissions the coefficient of variation for emission rate (g/h) for stove 1, stove 2, and stove 3 was 4, 17, and 65%, respectively. This demonstrates the need to sample longer for stoves with low emissions rates and particle loadings on the filters to have good agreement and stay above the method detection limit.

Efficiency measurements of each of these pellet stoves were all below the default value of 78% which supports the need to actually measure this in certification testing as is now required by the NSPS. This is important consumer information that will help determine the affordability of this emerging renewable heating fuel and is essential information for state energy programs. Emissions index is also essential information for state air planners designing change out programs as it provides a direct comparison from stove to stove and a much better indication of combustion quality. Finally, while CO is not currently regulated by EPA, it is an important health and safety factor that should be reported.

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Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) Administration, U.S. Energy Information. Short-Term Energy Outlook—Projected Winter Fuel Expenditures by Fuel and Region. [Online] December 8, 2015. http://www.eia.gov/forecasts/steo/report/WinterFuels.cfm.

(2) NYSERDA. *Monthly Average Home Heat Oil Prices*; NYSERDA: Albany, NY, 2016.

(3) Strauss, W. *Weather, Heating Oil Prices, and Wood Pellet Sales.* Bethel: FutureMetrics LLC, 2018.

(4) United States Environmental Protection Agency. FACT SHEET: Summary of Requirements for Woodstoves and Pellet Stoves. [Online] February 4, 2015. http://www.epa.gov/residential-wood-heaters/fact-sheet-summary-requirements-woodstoves-and-pellet-stoves.

(5) Orasche, J.; et al. Comparison of Emissions from Wood Combustion. Part 1. *Energy Fuels* **2012**, *26*, 6695−6704.

(6) McDonald, R. *Evaluation of Gas, Oil and Wood Pellet Fueled Residential Heating System Emissions Characteristics*; Energy Sciences and

Technology Department/Energy Resources Division., Brookhaven National Laboratory: Upton, NY, 2009; p 75. BNL-91286–2009-IR.

(7) Kinsey, J. S.; et al. Emission characterization of residential wood-fired hydronic heater technologies. *Atmos. Environ.* **2012**, *63*, 239–249.

(8) Northeast States for Coordinated Air Use Management (NESCAUM). *New York State Wood Heat Report: An Energy, Environmental, and Market Assessment. Rep. No. 15-26*; New York State Energy Research and Development Authority: Albany, NY, 2016.

(9) Johansson, L. S.; et al. Emission characteristics of modern and old-type residential boilers fired with wood logs and wood pellets. *Atmos. Environ.* **2004**, *38*, 4183–4195.

(10) Boman, C.; et al. Stove Performance and Emissions Chacteristics in Residential Wood Log and Pellet Combustion, Part 1: Pellet Stoves. *Energy Fuels* **2011**, *25*, 307–314.

(11) Alliance for Green Heat. Pellet Stove Design Challenge. [Online] Alliance for Green Heat, 2016. http://www.forgreenheat.org/decathlon/announcement.html.

(12) State of Maine, Department of Environmental Protection. *RE: Docket ID: EPA-HQ-OAR-2009-0734: Proposed Updates to the Residential Wood Heater New Source Performance Standards (NSPS)*; 2014.

(13) U.S. Environmental Protection Agency. *Residential Wood Heaters New Source Performance Standards (NSPS) Draft Proposal*; 2013.

(14) Curkeet, R. *New Hearth Emission Standards-The Definitive Guide to the EPA's NSPS for New Wood Heaters. s.1.*; Intertek, 2015.

(15) U.S. Environmental Protection Agency. *Process for Developing Improved Cordwood Test Methods for Wood Heaters*; U.S. EPA Office of Air Quality Planning and Standards: Washington DC, 2016.

(16) Curkeet, R.; Ferguson, R. *EPA Wood Heater Test Method Variability Study—Analysis of Uncertainty, Repeatability and Reproducibility Based on the EPA Accredited Laboratory Proficiency Test Database*; 2010.

(17) Johnson, G. W. A. *Pellet Stove.* US D601,684 S, Redmond, WA, October 6, 2009.

(18) ASTM International. *E2779-10 Standard Test Method for Determining Particulate Matter Emissions from Pellet Heaters*; ASTM International: West Conshohocken, PA, 2010.

(19) *E2515-11 Standard Test Method for Determination of Particulate Matter Emissions Collected by a Dilution Tunnel*; ASTM International: West Conshohocken, PA, 2011.

(20) Venturini, E.; et al. Evaluation of non-steady state condition contribution to the total emissions of residential wood pellet stove. *Energy* **2015**, *88*, 650–657.

(21) Schmidl, C.; et al. Particulate and gaseous emissions from manually and automatically fired small scale combustion systems. *Atmos. Environ.* **2011**, *45*, 7443–7454.

(22) Verma, V. K.; et al. Performance of a domestic pellet boiler as a function of operational loads: Part 2. *Biomass Bioenergy* **2011**, *35*, 272–279.

(23) Win, K. M.; Persson, T. Emissions from Residential Wood Pellet Boilers and Stove Characterizated into Start-up, Steady Operation, and Stop Emissions. *Energy Fuels* **2014**, *28*, 2496–2505.

(24) Canadian Standards Association. *Performance testing of solid-fuel-burning heating appliances. s.l.*; Canadian Standards Association, 2010.

(25) Butcher, T.; Trojanowski, R.; Wei, G. *Cord Wood Testing in a Non-Catalytic Wood Stove*; Brookhaven National Laboratory: Upton, NY, 2014.

(26) U.S. Environmental Protection Agency. List of EPA Certified Wood Stoves. [Online] May 2016. https://www.epa.gov/compliance/list-epa-certified-wood-stoves.

(27) U.S. Environmental Protection Agency. Federal Register—The Daily Journal of the United States Government—Standards of Performance for New Residential Wood Heaters, New Residential Hydronic Heaters and Forced-Air Furnaces. [Online] March 16, 2015. https://www.federalregister.gov/articles/2015/03/16/2015-03733/standards-of-performance-for-new-residential-wood-heaters-new-residential-hydronic-heaters-and.

(28) Brown, C. J.; Jordan, R. A.; Tucholski, D. R. *Furnace CO emission under normal and comprised vent conditions*; The United States Consumer Product Saftey Comission: Washington, DC2000.

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Energy Fuels 2018, 32, 3543–3550