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HOLISTIC ANALYSIS OF EMISSIONS FROM A MODULAR FAST PYROLYSIS SYSTEM FOR CONVERSION OF BIOMASS AND MIXED WASTE

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ABSTRACT: Fast pyrolysis of municipal solid waste (MSW) provides an alternative to open burn pits and MSW incinerators, which emit harmful gaseous emissions and particulate matter. Fast pyrolysis converts MSW to a primarily liquid product (i.e., bio-oil), which burns cleaner than solid MSW, while still providing heat/power. Pyrolysis reactors can also operate without external power by burning pyrolysis byproducts (i.e., bio-oil, char, and gas) to generate process heat needed to dry the feedstock and operate the reactor. To demonstrate the benefits of processing MSW using pyrolysis, we determined the total emissions for a 45 kg/h, modular fast pyrolysis system with on-site utilization of pyrolysis products. Emissions were measured during the operation of three different burners fueled by pyrolysis bio-oil, char, and gas. Measured emissions were compared to standards set by the EPA regarding nine pollutants emitted by municipal solid and other solid waste incinerators. Five of the EPA regulated pollutants (dioxins/furans, Cd, Pb, Hg, and HCl) were not observed during testing as they typically depend on feedstock composition (Cd, Pb, Hg, HCl) or are greatly reduced by burning liquid fuels (dioxins/furans). The pyrolysis system also met carbon monoxide, nitrogen oxide, and sulfur dioxide emissions standards at 10.6 ppm, 16.8 ppm, and 2.3 ppm respectively. PM emissions exceeded EPA limits at 60 mg/m^3 versus 30 mg/m^3. However, PM emissions are easily reduced by using a baghouse filter adjusting the bio-oil burner

air-to-fuel ratio. These results clearly demonstrated that pyrolysis can convert MSW to power/heat while also meeting EPA emissions standards without using air pollution control systems.

I. INTRODUCTION

Municipal solid waste (MSW) disposal is a significant issue everywhere. In 2015, the United States produced 262 million tons of MSW, of which 68 million tons were recycled, 23 million tons were composted, 33 million tons were incinerated for energy, and 137 million tons were landfilled.[1] Globally, MSW totaled 1.3 billion metric tons in 2013, and is expected to increase to 2.2 billion metric tons by 2025.[2] MSW is often landfilled or incinerated to produce energy, with landfilling more prominent in the U.S. and incineration more prominent in Europe.[2] Landfilling is a large source of methane (a powerful greenhouse gas), while incineration releases toxic heavy metals and volatile organic compounds (VOCs). The problems associated with MSW are also felt acutely within the U.S. military. Military installations in remote locations often lack suitable MSW disposal methods. Composting and landfilling are often unavailable due to lack of space, while on-site recycling requires too much labor resources and specialized equipment. Currently, MSW produced by remote military installations is incinerated, often in open burn pits.

Open burn pits are extremely dirty and emit pollutants that are a direct health hazard to soldiers and are environmentally hazardous. Specifically, open pit burning can produce VOCs, semivolatile organic compounds (SVOCs), polycyclic aromatic hydrocarbons (PAHs), chlorobenzenes, and carbonyls. At the small-scale (i.e., backyard barrel burning), open burning was demonstrated to generate VOCs at a rate of 2.46 g/kg-fuel, SVOCs at a rate of 0.26 g/kg-fuel, carbonyls at a rate of 1.47 g/kg-fuel.[3] Additionally, small-scale open burning generates PM at approximately 20 g/kg-fuel, CO at 43 g/kg-fuel, NO_x at 14 g/kg-fuel, and CH_4 at 15 g/kg-fuel.[4] Larger scale open burning of MSW occurs regularly at landfills as surface and subsurface fires, while crops are regularly

burned openly in fields. At these larger scales, air toxics (VOC, SVOC, etc.) are emitted at concentrations one order of magnitude less than small-scale open burning. In contrast, the magnitude PM, CO, NOx, and CH_4 emissions is comparable between small- and large-scale open fires. Oanh et al. found that open burning of rice straw generated PM at 9.4±3.5 g/kg-fuel.[5] Similarly, Li et al. found open burning wheat straw generated CO at 50 g/kg-fuel, CH_4 at 3.4 g/kg-fuel, SO_2 at 0.85 g/kg-fuel, and NO_x at 3.3 g/kg-fuel.[6]

Incinerators are a significant improvement upon open burning and are a common solution for MSW remediation in population dense cities or countries. Most modern municipal incinerators are equipped to generate steam, heat, or power.[2] Incinerators are more efficient than open burning and produce fewer emissions, such that the EPA does not regulate total VOCs, SVOCs, PAHs, or PCBs emitted by incinerators. However, air pollution control (APC) systems (i.e., scrubbers) are required to meet other EPA emission standards.[2] In the U.S., there are three sets of EPA regulations that set limits on incinerators. The current EPA regulations include rules for small municipal solid waste incinerators (MSWIs, < 250 tons/day),[7] large municipal solid waste incinerators (MSWIs, > 250 tons/day),[8] and other solid waste incinerators (OSWIs).[9] The EPA regulations for incinerators place limits on PM, dioxin/furan, cadmium, lead, mercury, HCl, nitrogen oxides (NO_x), and sulfur dioxide (SO_2).

Replacing open pit burning with incinerators would allow the U.S. military to reduce harmful emissions and better protect soldier health. However, APC systems would be needed to meet EPA emission guidelines. Most APC systems use either wet or dry scrubbing to remove PM and gaseous acids, selective non-catalytic reduction of NO_x using urea, and adsorbents (e.g., activated carbon or zeolites) to remove dioxins.[2] Implementing the required emission control needed for an incinerator in forward military installations is difficult and costly. An alternative to MSW

incineration is pyrolysis that produces bio-oil, char, and pyrolysis gas. Using pyrolysis to produce combustible liquids, solids, and gases enables using burners that are typically cleaner than simple incinerators.

Pyrolysis is an oxygen-free, thermochemical reaction that converts large molecular weight solids (typically biomass or plastics) to a majority liquid oil product (60–80%), often referred to as bio-oil or pyrolysis oil.[10] Secondary products include solid carbonaceous char and a gas stream composed of CO_2, CO, CH_4, and other light hydrocarbons. Fast pyrolysis is generally considered to occur at temperatures above 500 °C with heating rates greater than 100 °C/s and residence times on the order of a few seconds,[11] which ultimately favors bio-oil production.[10, 12-14] In contrast, slow pyrolysis occurs at temperatures between 300–750 °C with heating rates between 1 and 30 °C/min and residence times on the order of minutes to hours, which ultimately favors char production.[15] In order to process large volumes of MSW, fast pyrolysis is preferred. The products of pyrolysis, especially bio-oil and char, can be used immediately to generate heat/power or stored for later use. Storage is made possible because pyrolysis products are denser than MSW, resulting in significantly less storage volume.

Fast pyrolysis conversion is typically greater than 99%,[16] which enables complete elimination of MSW. Fast pyrolysis reactors are typically designed as fluidized bed reactors, where an inert fluidizing gas (N_2) carries the feedstock into a heated reaction zone and carries products out of the reactor.[10, 17] Fast pyrolysis reactors can be completely self-sustaining by using process heat generated via combusting some of the pyrolysis products and recycling the fluidizing gas/pyrolysis product gas mixture.[18] Overall, pyrolysis is a net energy producer when using low-moisture feedstocks (< 30% water).[18] More importantly, pyrolysis produces fuels that can be utilized via

readily available burners/generators. This is particularly relevant in military installations, which could use pyrolysis oil in already existing fuel burners to provide heat and electricity.

A process flow diagram for Mainstream's pyrolysis reactor with integrated product burners is shown in Figure 1. The pyrolysis reactor can process MSW at 45 kg/h (nominally one ton/day). Based on mass and energy balances, the pyrolysis reactor can be a net-positive energy producer, generating 13.6 kg/h of bio-oil (energy equivalent to 73 kW). Pyrolysis gas and bio-oil are used to heat the reactor and fluidizing gas. Pyrolysis char is used to dry incoming feedstock and lower moisture content from 30% to 10%. The major process units, represented in Figure 1, have been built and tested, with MSW processing capacity of 45 kg/h. The reactor was capable of yielding 65 wt% bio-oil; specific product yields and heating values are listed in Table 1.

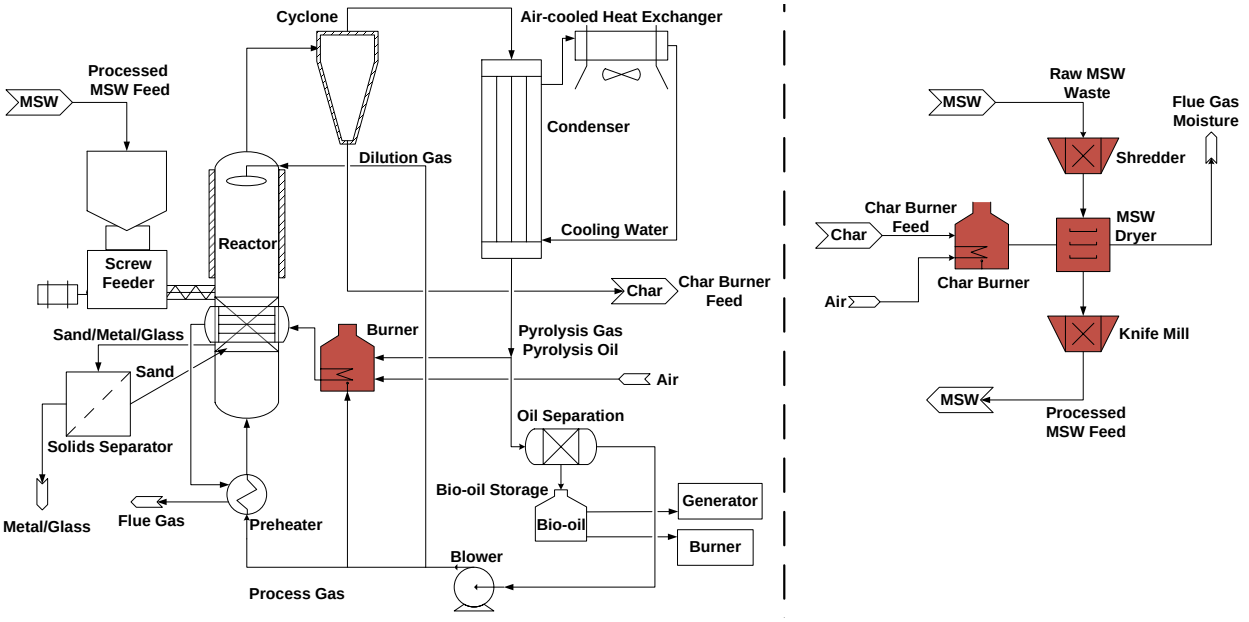

Figure 1. Process flow diagram for the 45 kg/h pyrolysis process with integrated burners.

Table 1. Heating Value and Dry Yield for Pyrolysis Products

Pyrolysis Product	Yield, dry [wt%]	Lower Heating Value [MJ/kg]
Bio-oil	65	17.5
Char	15	29.7
Non-condensable gas	20	7.3
Pine Feedstock	–	17.2

We evaluated whether fast pyrolysis can meet emissions guidelines by operating three individual burners with pyrolysis products and measuring the subsequent CO, PM, NO_x, SO_x and total hydrocarbon (THC) concentrations. THC concentration was measured as an analog for VOCs. The char, gas, and oil fuels were generated using a fast pyrolysis reactor as described in Figure 1. Based on pollutant concentrations and emission indices, we found that operating a fast pyrolysis reactor in sequence with gas, char, and oil burners can allow MSW remediators to meet EPA targets for common combustion pollutants.

II. EXPERIMENTAL METHODS

Pyrolysis

The 45 kg/h pyrolysis system was instrumented with numerous thermocouples, pressure transducers, flow meters, and power meters. The system was controlled using a programmable controller (Automation Direct DirectSOFT) and an operator interface terminal (C-More) to control automatic sequences, manual outputs, proportional-integral-derivate control loops, alarms, and interlocks. The 45 kg/h reactor was fed pine sawdust (d90 of 0.94 mm) at an average feed rate of 11.4 kg/h for 10 hours in order to ensure steady state operation and generate sufficient quantities of pyrolysis products subsequent burner testing. Pine was chosen as a surrogate for MSW because both have low sulfur content and the oil, gas, and char products are likely to have similar properties.[19-20] The fluidized bed as held at 500 °C using cartridge heater imbedded within a sand bed while the reactor was fluidized by pre-heated nitrogen fed into the bottom of the reactor at 60 m^3/h. Bio-oil was collected at a rate of 1.3 kg/h. This bio-oil will be referred to as MEC throughout the report. Specific properties of the bio-oils can be found in the Supplementary Information.

Emissions Measurement

A full suite of emissions analyzers were available to measure CO, CO_2, NO_x, PM, THC, and SO_x. For each burner, a probe sampled a portion of the exhaust gas and the corresponding sample line led directly to analyzers for CO, CO_2, NO_x, and THC. The sample line also included a cyclone (URG Corporation model 2000-30ED) and a particulate filter to protect the analytical instruments. CO and CO_2 concentrations were measured using a non-dispersive infrared analyzer (California Analytical Instruments 602 series). THC concentration was measured using a flame ionization detector analyzer (Thermo Scientific Model 51i-HT). A chemiluminescence analyzer (Thermo Scientific model 42i-HL) measured NO_x concentrations. Sample flow rate to the gas analyzers was set by a choked critical orifice backed by a diaphragm vacuum pump. SO_2 was measured offline by collecting the exhaust gas and using a colorimetric SO_2 glass detection tube (Sensidyne model # 103SB).

During bio-oil burner testing, PM was measured continuously using a particle mass analyzer (Pegasor PPS M analyzer). The particle mass analyzer had an internal cyclone that excluded particles > 1 μm and restricted the maximum detectable particle size to 1 μm. Previous research has shown that PM from oil combustion in a refractory-lined combuster is under 1 μm in diameter,[21] and therefore, the internal cyclone did not bias the measurements. The particle mass analyzer included its own heated sample lines, with an inlet and outlet separated by 7.5 cm. The inlet was located 48 cm upstream of the blower. During char burner testing, PM emissions were captured by a baghouse filter, which was weighed after operating the char burner for 16 h to calculate the rate of PM emissions. During gas burner testing, no PM emissions were measured as particulates are unlikely to form during combustion of non-condensable gases.[22]

Pyrolysis Bio-oil Burner Testing

A waste oil burner (Inov-8 International, S200) for high viscosity fuels was used for all bio-oil combustion testing. Bio-oil samples were purchased commercially (BTG BioLiquids BV, CAS # 94114-43-9) and produced on site using the 45 kg/h pyrolysis reactor. Because bio-oil is highly viscous, blended mixtures of bio-oil and ethanol/butanol were used to operate the bio-oil burner. Both bio-oil samples were blended with ethanol at a ratio of 20 wt% to 80 wt% (bio-oil:ethanol). In addition, a blend of 50 wt% butanol, 20 wt% BTG bio-oil, and 30 wt% jet fuel (Jet A) was created for testing. The bio-oil was blended with butanol because it is a renewable fuel and jet fuel is used industrially and by the military. Lastly, the burner was also operated with pure diesel fuel to ensure proper operation and provide a baseline comparison with traditional fossil fuels.

For each test of the bio-oil burner, flame temperature at four locations, exhaust gas emissions (CO, CO_2, NO_x, PM, and THC), and fuel feed rate were measured. SO_2 was measured offline by collecting the exhaust gas and using a colorimetric SO_2 glass detection tube. The bio-oil burner was mounted to a 30 cm diameter exhaust gas tube that was five meters long. Fuel and air were fed to the burner flame through atomizing nozzle, which mixes liquid fuel and air into an aerosol spray. The exhaust gas was vented directly to atmosphere to avoid backpressure within the exhaust gas tube. Exhaust gas was sampled five meters downstream of the flame to ensure complete mixing of the excess air and combustion products. Fuel feed rate was determined by measuring the mass lost in the fuel tank. Measurements were taken with an electronic balance that recorded the fuel mass to a computer every 30 seconds. The flame temperature was measured at 20, 40, 60, and 100 cm from the base of the flame at the nozzle.

Pyrolysis Char Burner Testing

Char burner experiments were conducted using an externally heated fluidized bed combustion reactor with 5020 silica sand bed material (Surface Prep Supply, d50 of 225 µm). Downstream of the reactor, a cyclone separator and a baghouse filter removed particulates from the exhaust gas. The reactor and cyclone temperatures were maintained with electric heaters. The reactor was 140 cm long with a 7.6 cm diameter with a bed depth between 30–38 cm. Pyrolysis char was produced on site using the 45 kg/h pyrolysis reactor under the same operating conditions described in the Experimental Methods section. The char contained 68.0 wt% fixed carbon, 15.5 wt% volatile matter, 20.4 wt% ash, and 2.3 wt% water. Carbon content was measured in accordance with ASTM 3176-15, ash content in accordance with ASTM 7582-12, and water content in accordance with ASTM 7582-12. The higher heating value of the char was 29.7 MJ/kg as measured by bomb calorimetry. Major components of the ash fraction were CaO, K_2O, Al_2O_3, MgO, and SiO_2. Additionally, the char particle size distribution (Figure S1 in Supplementary Information) was measured by mechanically sieving the char in accordance with ASTM 2862-10. The char was fed directly to the char burner without pre-processing.

During char burner operation, the exhaust gas emission concentrations, reactor temperature, char feed rate, and fluidizing combustion air flow rate were measured continuously. CO, CO_2, NO_x, THC, SO_2, and PM were measured using the suite of equipment described in the Emissions Measurement Section. The average char feed rate was determined with the screw feeder set-point and confirmed by measuring the mass of the char in the feed hopper before and after testing. The air flow rate was set with a mass flow controller, which relayed flow rates to the data acquisition and control system. The reactor temperature (790–900 °C), fluidizing air velocity (0.4–1.0 m/s), and excess air (15–35%) were varied during testing. Specific conditions can be found in the Supplementary Information.

Pyrolysis Gas Burner Testing

A high-velocity flue-gas burner (Fives North American Aardvark 4442A-2) designed specifically for low heating value fuels was used for all pyrolysis gas burner tests. The burner was capable of maintaining a 900 °C flame temperature. A cylindrical exhaust gas tube (4.5 cm diameter, 5 m length) was affixed to the gas burner to act as an outlet for exhaust gases, which were vented directly to atmosphere to avoid backpressure within the combustion chamber. The flow rate of the fuel was set by a pressure regulator on the fuel gas supply cylinder and the air flow rate was set by a variable-frequency drive-controlled blower. The air feed was preheated to 100 °C before being fed into the burner. The exhaust gas temperature was measured and the exhaust emissions were sampled one meter downstream of the flame to ensure complete mixing of the excess air and combustion products. The air-to-fuel equivalence ratio (λ) was calculated based on the fuel and air flow rates. The flame temperature was measured 30 cm downstream of the flame.

A surrogate pyrolysis gas (Airgas, North America) as a product of the pyrolysis process and propane as a simple heating supplement were tested with the burner. Propane has a lower heating value (LHV) of 47 MJ/kg, while the surrogate pyrolysis gas has a LHV of 8 MJ/kg as calculated using reported LHV values of constituent gases. The surrogate pyrolysis gas was composed of 16% CH_4, 39% CO_2, and 45% CO (volume percent, ±1% error) to match values reported by Bridgwater et al.[23] The gas burner was operated with the air flow rate ranging from 200–1,250 L/min. Fuel flow rates were set to reach a desired air-to-fuel ratio. For propane, this corresponded to a flow rate between 5–25 L/min (air-to-fuel ratio = 15.6 to 1), while the surrogate pyrolysis gas flow rate varied between 40–150 L/min (air-to-fuel ratio = 2.6 to 1). In addition to varying the air-to-fuel ratio, the total gas flow was varied to determine if emissions stayed constant

over the burner's operating range. The gas burner was stable, regardless of fuel, under the reported air-to-fuel ratios and with an air flow rate ranging between 240–2,200 L/min.

III. RESULTS AND DISCUSSIONS

Bio-oil Burner Emissions

Using a waste oil burner, we measured the CO, CO_2, NO_X, and THC emissions produced by burning three different bio-oil blends and a diesel fuel. Bio-oil blends used ethanol and butanol because the bio-oil was largely immiscible with diesel Jet-A. In general, results were similar to previous fuel-oil burner experiments,[24-25] while emissions were lower than typical diesel and spark ignition engines.[26-27] Summaries of emission factors (g-pollutant/kg-fuel) and concentrations are reported in Figure 2. Figure 2A shows that the CO emission factor was largely independent of the specific fuel blend. This was mirrored in Figure 2B, which shows that CO concentration was also unchanged by fuel choice. In contrast, the NO_X emission factor and concentration was higher for the diesel fuel and the bio-oil/butanol/Jet-A blend. Specifically, NO_X concentrations were 16% lower for the EtOH:MEC blend and 20% lower for the EtOH:BTG blend when compared with diesel fuel. Generally, NO_X emissions increase with increasing temperature and excess air.[25, 28] Additionally, ethanol has a relatively low adiabatic flame temperature compared to fuels with larger carbon-to-oxygen ratio.[29] Because the diesel fuel and the bio-oil/butanol/Jet-A blend had no ethanol and the blend had a lower proportion of alcohol (50% vs 80%), the flame temperature may have been elevated, which would have caused the NO_X emission factor to increase. EXPERIMENTAL TEMP DATA. The opposite trend was observed for THC emissions, where the EtOH:MEC and EtOH:BTG blends generated THC emissions approximately 70% higher than diesel fuel.

Since the ethanol:bio-oil mixture performed similarly to other fuel mixtures and ethanol is inexpensive, we chose to measure the PM and SO_2 emissions generated by operating a waste oil burner with an ethanol:bio-oil mixture. We used the MEC bio-oil for these tests. The PM concentration was measured to be 43 mg/m^3 at a λ of 2.65. PM emissions could be reduced further by improving combustion efficiency (e.g., decreasing λ). Interestingly, no SO_2 emissions were detected. We attributed this to the relatively low amount of sulfur present in the bio-oil and the limits of our detection method. Typically, most sulfur present in the MSW feedstock ends up in the char or gas product.[30] In order to estimate the maximum possible concentration of bio-oil burner SO_2 emissions, we determined the bio-oil sulfur content using elemental analysis and assumed that all sulfur was converted to SO_2. At a fuel feed rate of 5.8 kg/h and λ of 2.65, the SO_2 concentration was estimated to be 1.8 ppm.

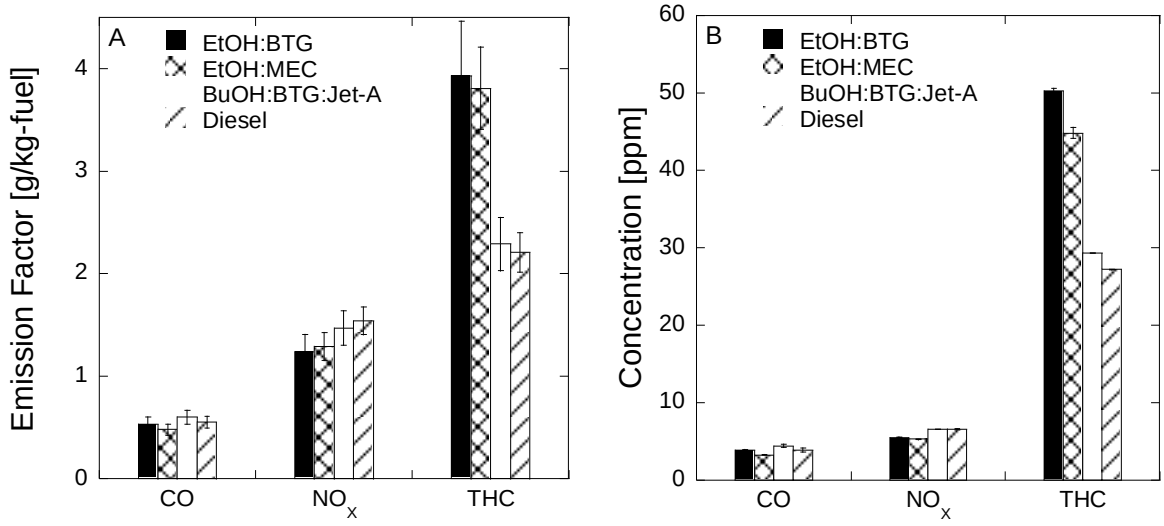

Figure 2. CO, NO_X, and THC emissions produced by operating a waste oil burner fueled by different fuel mixtures A) emission factors and B) emission concentrations. Error bars indicate 95% confidence intervals.

Char Burner Emissions

When burning solid carbonaceous fuels, CO emissions are a good indicator of combustion efficiency, which can be controlled by the reactor bed temperature, percent of excess air, and fluidizing gas velocity. Figure 3 shows the concentration of CO and NO_x as a function of bed temperature, percent of excess air, and gas velocity. Typically, increasing the combustion temperature reduces CO emissions and improves combustion efficiency.[31] We observed similar results using pyrolysis char (Figure 3A), where increasing temperature from 78 °C to 900 °C, decreased CO concentration from 605 ppm to 48 ppm. Increasing the fluidizing gas velocity from 0.4 m/s to 0.7 m/s had little effect on CO emissions, which increased from 83 ppm to 168 ppm. However, increasing the gas velocity to 1.0 m/s sharply increased CO emissions to 758 ppm. This suggests that the dependence of CO concentration on gas velocity is nonlinear. While bed temperature and gas velocity strongly influenced char burner CO emissions, the percent of excess air had relatively little effect. CO concentrations decreased slightly from 131 ppm at low excess air (11%) to 96 ppm at high excess air (36%).

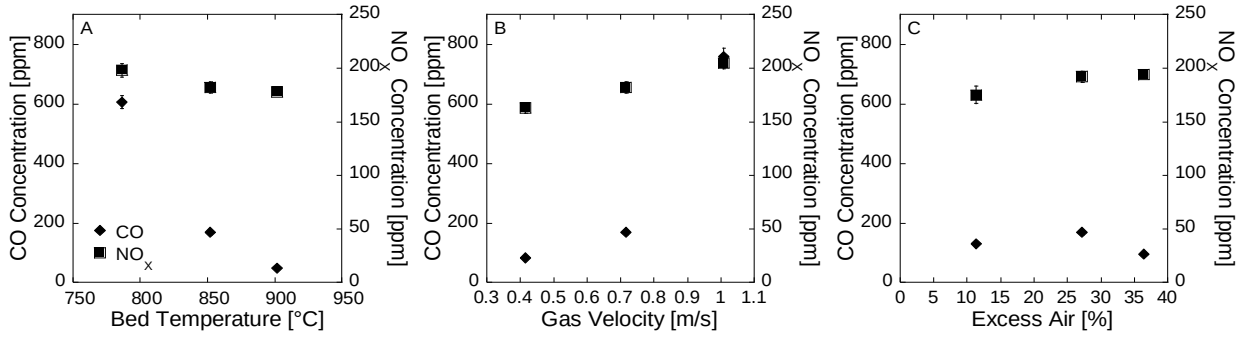

Figure 3. CO and NO_x emissions produced by a char burner fueled with pyrolysis char and varying the A) bed temperature, B) gas velocity, and C) excess air. Unless specified otherwise temperature was 840 °C, gas velocity was 0.7 m/s, and excess air was 25%. Error bars indicate 95% confidence intervals.

Reactor bed temperature, fluidizing gas velocity, and excess air level all had an impact on NO_x emissions. NO_x emissions decreased slightly with increasing temperature; NO_x concentration was highest at low temperatures (198 ppm at 780 °C) and lowest at high temperatures (178 ppm at 900 °C). Fluidizing gas bed velocity, which is inversely proportional to gas residence time, had an even greater impact on NO_x emissions (Figure 3B). As velocity increased from 0.4–1.0 m/s, the residence time decreased from approximately three seconds to one second, and NO_x increased from 163 ppm to 205 ppm. NO_x emissions increased with higher levels of excess air, as expected,[25] increasing from 175 ppm at an excess air level of 11% to 194 ppm at 36% excess air (Figure 3C). To minimize emissions, the reactor should be operated at 900 °C and 0.4 m/s gas velocity, while the percent excess air has a trade-off between NO_x and CO concentrations.

The sulfur content of the char was low, which resulted in near-zero SO_2 emissions detected. Average SO_2 emission across all reactor conditions was 3.40 ± 0.03 ppm. PM emissions were averaged over a 16 hour operating period, which included the full range of char burner operating conditions. Solids collected by the baghouse filter (17 g total) were taken to be the totality of PM emissions since baghouse filters have typical efficiencies of 99.99% and the average gaseous flow rate was 0.0425 m^3/min.[32] This corresponded to an average PM emission concentration 417 mg/m^3.

Flue-gas Burner Emissions

CO, NO_X, and THC emissions were measured during flue-gas burner testing. SO_2 and PM emissions were not observed because propane gas and the surrogate pyrolysis gas were sulfur-free and burning flammable gases does not emit solid particulates.[33] While the surrogate pyrolysis gas did not contain any sulfur, approximately 35% of sulfur in biomass is volatilized during pyrolysis.[30] This would correspond to the burner releasing SO_2 at 4.5 ppm based on elemental

analysis of the feedstock. The burner produced a stable flame between 700 °C and 900 °C. With respect to emissions, the pyrolysis gas performed similarly to propane. However, a significantly greater feed rate was required to operate the burner with pyrolysis gas than with propane (10 vs 100 L/min). This is because the LHV of the pyrolysis gas was low compared to propane (8 MJ/kg vs 47 MJ/kg). Despite the difference in fuel feed rate, both the pyrolysis gas and propane showed similar emission trends when the fuel feed rate was varied (Figure 4). Regardless of fuel, CO and THC emissions decreased as the fuel feed rate increased, while NO_X emissions remained unaffected.

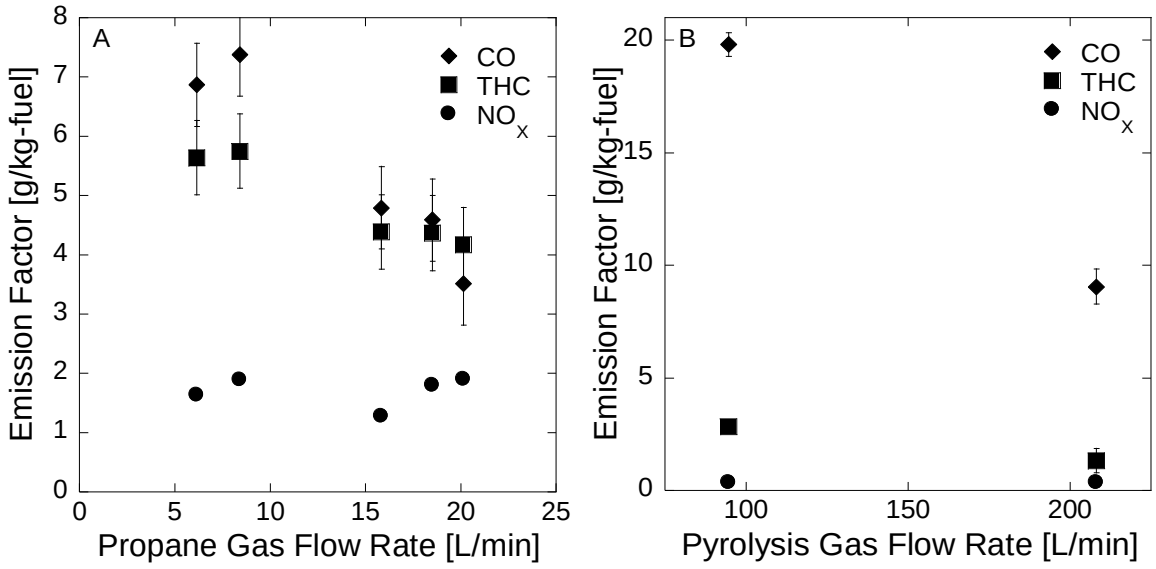

Figure 4. Emission data at a fuel-to-air equivalence ratio of A) 2.0 for propane and B) 1.1 for process gas for the Aardvark gas burner with heated air feed at 100 °C, error is within the bounds of the marker where not visible.

When varying the air-to-fuel ratio, CO, THC, and NO_X emissions followed similar trends for both the pyrolysis gas and propane. At fuel-rich conditions ($1 < \lambda < 2.25$), the emissions remained relatively unchanged. Both fuels produced fewer than 20 g CO/kg-fuel, fewer than 20 g THC/kg-fuel, and 5 g NO_X/kg-fuel. Notably, the pyrolysis gas produced fewer THC and NO_X

emissions (4.5 g THC/kg-fuel and 0.25 g NO_X/kg-fuel) compared to the propane fuel. At fuel-lean conditions (λ > 2.25), CO, THC, and NO_X emissions spiked; the emissions spike was more prominent with the pyrolysis gas fuel. The raw, undiluted point-source emissions for the burner were similar or less than the ambient air quality standards for incinerators.[34]

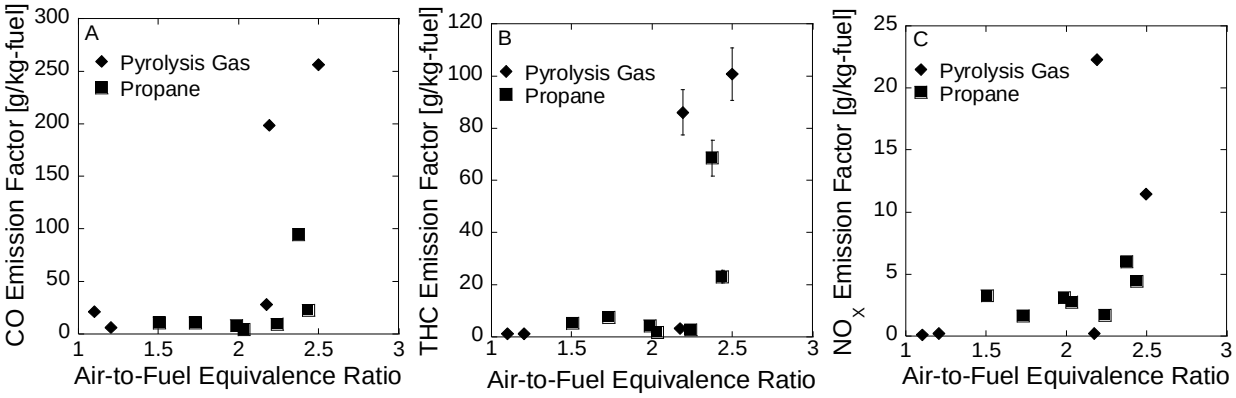

Figure 5. Emission data over the air-to-fuel equivalence ratios for the gas burner for propane and process gas with heated air feed at 100 °C, error is within the bounds of the marker where not visible. Fuel feed rate was adjusted to ensure burner light-off, but was approximately 15 L/min for propane and 100 L/min for pyrolysis gas.

Meeting Incinerator Emissions Standards using Fast Pyrolysis

There are three sets of EPA regulations that are relevant for determining if pyrolysis of MSW can meet current emissions standards. The current EPA regulations include rules for small MSWIs (< 250 tons/day),[7] large MSWIs (> 250 tons/day),[8] and other solid waste incinerators (OSWI).[9] Each set of emissions rules stipulates that emission concentrations are calculated on a dry basis at 7% oxygen. Table 2 shows the emissions (CO, NO_X, SO_2, and PM) produced by each individual burner. The char burner was the dirtiest of the three burners and only met EPA standards for SO_2. The gas burner met all of the EPA standards, while the bio-oil burner only failed to meet PM standards. However, since each of these burners are subcomponents of the overall pyrolysis unit and oil, char, and gas yields are significantly different, determining the total pyrolysis

emissions required weighting individual burner emissions with the yield of the pyrolysis products. Table 2 does not include other regulated pollutants (HCl, Cd, Pb, Hg, and dioxins/furans) as they were not detected. HCl, Cd, Pb, and Hg were not present in our feedstock, but could be an issue for other feedstocks (e.g., PVC plastics produce HCl and coal produces Hg). Dioxins/furans were likely absent due to the efficiency of oil and gas burners compared to solid waste incinerators; dioxin and furan emissions have been found to be 20 times lower for pyrolysis compared to combustion.[35]

Table 2. Bio-oil, Char, and Flue-gas Burner Emissions

Burner	CO* [ppm]	NO_X* [ppm]	SO_2* [ppm]	PM* [mg/m^3]
Bio-oil	3.3	5.3	1.8‡	39.4
Char	68	160	3.4	360
Gas	27.3	1.4	4.5‡	0.0

* Emissions calculated on a dry basis at 7% oxygen per EPA standards
‡ Estimated based on sulfur content of the burner fuel

To compare pyrolysis emissions with the EPA standards, we took the emissions concentrations from each burner (Table 2) and calculated the concentration of each pollutant assuming that each burner released its exhaust through a single stack. A detailed explanation of this calculation can be found in the Supplementary Information. Table 3 compares EPA emissions standards with the total emissions of a 45 kg/h pyrolysis system that uses standard oil, char, and gas burners to provide process heat and generate electricity. The pyrolysis system easily meets CO, NO_X, and SO_2 EPA emissions standards without using an air pollution control system. We expect these results to remain relatively feedstock-agnostic because the combustion efficiencies realized here were the result of converting solid wastes to clean-burning liquid/gas products. An exception would be any feedstocks rich in chlorine, lead, mercury, or cadmium. Further testing is required to determine whether pyrolysis can effectively handle these types of feedstocks without using an air pollution control system.

Despite meeting most EPA emissions standards, the pyrolysis system did not meet PM standards (20 to 30 mg/m^3, depending on the EPA ruleset). While the gas burner generated no PM emissions, the bio-oil and char burners produced PM at 39.4 mg/m^3 and 360 mg/m^3 respectively. For the pyrolysis system as a whole, this corresponded to PM concentrations of 60 mg/m^3, approximately double current EPA standards. Because the bio-oil burner λ was not optimized here, lowering λ below 2.65 could significantly reduce PM emissions. Additionally, implementing a baghouse filter on the bio-oil and char burners would reduce the PM below the target concentration. Baghouse filter efficiencies are typically 99.99% on a mass basis, even with continuous use.[32] Table 3 shows that simply adding a baghouse filter to the char burner would reduce PM emissions by nearly 50% and fail to meet OSWI PM emission standards by 2.6 ppm. We expect that subsequent tuning of the bio-oil burner to decrease PM emissions would allow the pyrolysis system to meet EPA standards.

Table 3. Combined Emissions for 45 kg/h Pyrolysis System with Integrated Burners

Emission	Total Pyrolysis Emissions*	Total Pyrolysis Emissions with PM Filter*	EPA OSWI	EPA Large MSWI	EPA Small MSWI
CO (ppm)	10.6	10.6	40	50	50
NO_x (ppm)	16.8	16.8	103	180	500
SO_2 (ppm)	2.3‡	2.3‡	3.1	30	30
PM (mg/m^3)	60.0	32.6	30	20	24

*Emissions calculated on a dry basis at 7% oxygen per EPA standards

‡ Sulfur emissions predicted to be 1.3 ppm assuming all sulfur in the feedstock was released as SO_2

IV. CONCLUSIONS

Disposing of municipal solid waste is burdensome. Landfills are unpopular and use valuable land. Incinerators reduce waste volume and can generate power, but require air pollution control systems. Pyrolysis is an alternative waste disposal method capable of converting MSW to energy dense liquid, solid, and gas products. Combustion of pyrolysis byproducts was shown to be possible using unmodified burners. Burning pyrolysis products generates enough energy to

make MSW pyrolysis self-sustaining, while also producing energy for the grid. Additionally, converting MSW to liquid and gaseous fuels enables cleaner combustion compared to incinerating MSW. MSW pyrolysis met CO, NO_X, and SO_2 emissions standards at 10.6 ppm, 16.8 ppm, and 2.3 ppm respectively. PM emissions exceeded EPA standards, but adopting a baghouse filter on the char burner and increasing the bio-oil burner air-to-fuel ratio would likely enable the pyrolysis system to meet EPA regulations.

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HOLISTIC ANALYSIS OF EMISSIONS FROM A SMALL, MODULAR FAST PYROLYSIS SYSTEM FOR CONVERSION OF BIOMASS AND MIXED WASTE

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Properties of Pyrolysis Products. The following tables list the yield and properties of pyrolysis products. Table 1 lists the yield and lower heating value of bio-oil, char, and non-condensable gases produced by pyrolyzing pine sawdust at 500 °C. Bio-oil was the majority product, while char had the largest heating value. Table 2 lists several properties of the bio-oil product in comparison with Grade D crude oil. Compared to Grade D oil, our bio-oil had less water and solids, but more ash and sulfur. Density was approximately the same, while the gross heat of combustion was higher for bio-oil.

Table 1. Heating Value and Yields for Pyrolysis Products

Pyrolysis Product	Lower Heating Value [MJ/kg]	Dry Yield [wt%]
Bio-oil	17.5	65
Char	29.7	15
Non-condensable Gases	7.3	20

Table 2. Properties of Pyrolysis Bio-oil

	Grade D	Bio-oil
Gross Heat of Combustion* [MJ/kg]	15	19.2
Water Content‡ [wt%]	30	24.4
Solids Content‡ [wt%]	0.25	0.042
Density at 20 °C [kg/dm^3]	1.1–1.3	1.2
Sulfur Content‡ [wt%]	0.05	0.016
Ash Content‡ [wt%]	0.15	0.064

*Minimum measured value
‡Maximum value measured

Table 3. Properties of Pyrolysis Char

Property	Value
Heating Value, dry [MJ/kg]	29.7
Moisture Content [wt%]	6.03
Ash content [wt%]	5.26
Fixed Carbon [wt%]	82.7

Particle Distribution of Pyrolysis Char. The char particle size distribution was determined by sieving pyrolysis char through sixteen different sieves ranging a No. 6 sieve (3.36 mm) to a No. 400 sieve (0.037 mm). Approximately 78 g of char was placed in the No. 6 sieve. The sieves were stacked one upon another, with the largest mesh size on top. The stack of sieve was then agitated using a vibratory sieve shaker. After shaking, each sieve was weighed to determine the mass of char present in each sieve. The particle distribution is shown in Figure 1. The particle size distribution peaked at 0.25 mm and approximately 99% of the char was smaller than 1 mm.

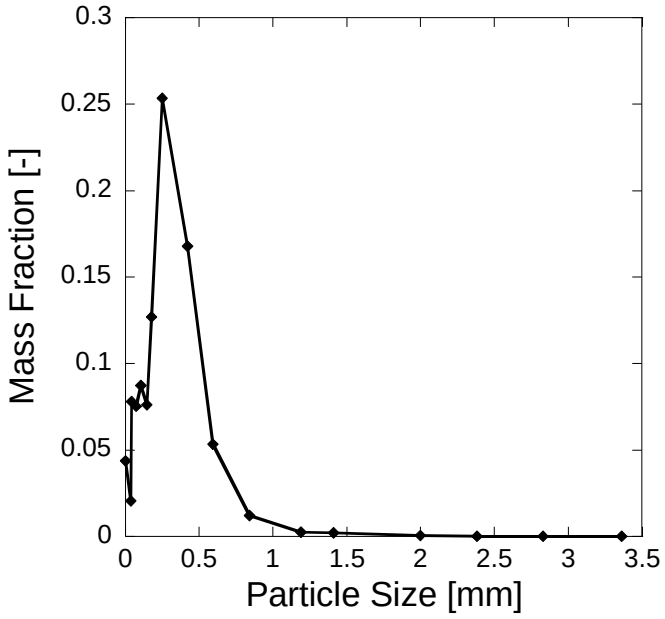

Figure 1. Particle size distribution of pyrolysis char.

Burners for Pyrolysis Products. Figure 2 shows pictures of the bio-oil burner, char burner, and gas burner. The bio-oil burner and gas burner are shown operating as received and without the combustion tube that was used during testing.

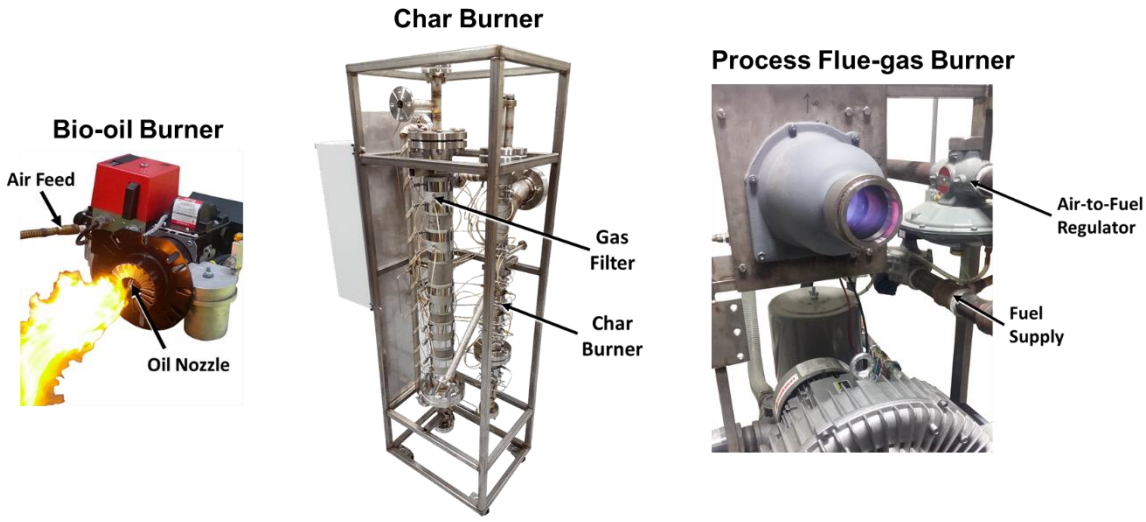

Figure 2. Bio-oil, char, and flue-gas burners

Test Conditions for Char Burner Testing. Table 4 shows the test conditions used for char burner testing. The preheater was maintained at 360 °C while the gas velocity, excess air, and reactor temperature were changed. Conditions were chosen to minimize the number of tests.

Table 4. Test Matrix for Combustion of Pyrolysis Char with a Fluidized Bed Reactor

Test	Preheater Temperature [°C]	Gas Velocity [m/s]	Excess Air [%]	Reactor [°C]
1	200	0.7	25	840
2	360	0.7	25	840
3	360	0.7	35	840
4	360	0.7	15	840
5	360	0.7	25	900
6	360	0.7	25	780
7	360	0.4	25	840
8	360	1.0	25	840

Calculations for Total Emissions. To calculate the total CO, NO_X, SO_2, and PM emissions produced by a MSW pyrolysis reactor the emissions concentrations measured during testing were first corrected to a 7% oxygen basis and then a dry basis, as prescribed by EPA standards. Equation 1 was used to correct emissions to a 7% oxygen basis. The equation uses PM as an example, but

was also used for CO, NO_X, and SO_2. In Equation 1, $O_{2,\ ambient}$ was 20.9%, $O_{2,\ 7\%}$ was, and $O_{2,\ exhaust}$ was calculated for each burner test and based on stoichiometry.

Equation 1 $$C_{PM,\ Oxygen\ Corrected} = C_{PM}\frac{O_{2,\ ambient} - O_{2,\ 7\%}}{O_{2,\ ambient} - O_{2,\ exhaust}}$$

After correcting for 7% oxygen, Equation 2 was used to correct emission concentrations to a dry basis. Once again, $H_2O_{exhaust}$ was calculated for each burner test and based on stoichiometry.

Equation 2 $$C_{PM,\ Dry\ Basis} = C_{PM,\ Oxygen\ Corrected}\frac{100}{100 - H_2O_{exhaust}}$$

Calculating the total emission concentration produced by a MSW pyrolysis reactor begins with Equation 3 where the mass of PM produced by the bio-oil, char, and gas burners is divided by the mass of the exhaust gas released by the bio-oil, char, and gas burners. The mass of the exhaust gas from each burner ($m_{exhaust,\ bio\text{-}oil}$; $m_{exhaust,\ char}$; and $m_{exhaust,\ gas}$) is known from measurements conducted during burner testing. Because the concentration of PM ($C_{PM,\ bio\text{-}oil}$; $C_{PM,\ char}$; and $C_{PM,\ gas}$) for each burner is also known, the mass of PM can be determined using Equation 4. Then substituting for $m_{PM,\ bio\text{-}oil}$, $m_{PM,\ char}$, and $m_{PM,\ gas}$ from Equation 4 into Equation 3 leads to Equation 5, which can be used to calculate the total concentration of a specific pollutant produced by a MSW pyrolysis reactor.

Equation 3 $$C_{PM,\ Total} = \frac{m_{PM,\ bio\text{-}oil} + m_{PM,\ char} + m_{PM,\ gas}}{m_{exhaust,\ bio\text{-}oil} + m_{exhaust,\ char} + m_{exhaust,\ gas}}$$

Equation 4 $$C_{PM,\ gas} = {m_{PM,\ gas}}/{m_{exhaust,\ gas}} \rightarrow m_{PM,\ gas} = C_{PM,\ gas} * m_{exhaust,\ gas}$$

Equation 5 $$C_{PM,\ Total} = \frac{{C_{PM,\ bio\text{-}oil}}/{m_{t,\ bio\text{-}oil}} + {C_{PM,\ char}}/{m_{t,\ char}} + {C_{PM,\ gas}}/{m_{t,\ gas}}}{m_{exhaust,\ bio\text{-}oil} + m_{exhaust,\ char} + m_{exhaust,\ gas}}$$

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14. ABSTRACT

Fast pyrolysis of municipal solid waste (MSW) provides an alternative to open burn pits and MSW incinerators, which emit harmful gaseous emissions and particulate matter. Fast pyrolysis converts MSW to a primarily liquid product (i.e., bio-oil), which burns cleaner than solid MSW. To demonstrate the benefits of processing MSW using pyrolysis, we measured emissions from three different burners fueled by pyrolysis bio-oil, char, and gas and compared the results to standards set by the EPA regarding emissions from waste incinerators. Five of the EPA regulated pollutants (dioxins/furans, Cd, Pb, Hg, and HCl) were not observed during testing as they typically depend on feedstock composition (Cd, Pb, Hg, HCl) or are greatly reduced by burning liquid fuels (dioxins/furans). The pyrolysis system also met carbon monoxide, nitrogen oxide, and sulfur dioxide emissions standards at 10.6 ppm, 16.8 ppm, and 2.3 ppm respectively. PM emissions exceeded EPA limits at 60 mg/m^3 versus 30 mg/m^3. However, PM emissions are easily reduced by using a baghouse filter adjusting the bio-oil burner air-to-fuel ratio. These results clearly demonstrated that pyrolysis can convert MSW to power/heat while also meeting EPA emissions standards without using air pollution control systems.

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