Emissions from Wood Pellet Combustion: Carbon Monoxide, Particulate Matter, and Polycyclic Aromatic Hydrocarbons

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Abstract

The global shift from fossil fuels to biomass fuels necessitates an analysis of harmful emissions from these biomass fuels. Biomass combustion generates products such as carbon monoxide (CO), particulate matter (PM), and polycyclic aromatic hydrocarbons (PAH). This paper surveys various experiments measuring harmful emissions from wood pellet appliances. It was found in most cases that low combustion temperatures cause an increase in harmful emissions. The studies showed that the greatest emissions occurred during the startup and burnout phase of wood pellet appliances due to non-optimal excess air ratios (λ). Ideal excess air ratios are specific to the individual appliance, pellet type, and combustion condition, so a universal number cannot be generated. CO emissions were well-tested and still proved to be a potential concern in poorly ventilated areas, but CO is backed by a much greater bed of research and understanding. PM and PAH were also demonstrated to have concerning emission levels; however, these emissions lack a base of conclusive, uniform research and understanding. Additionally, this work explores various mitigation solutions for pellet-fuel appliance companies should they be faced with pressure to reduce harmful emissions.

1. Introduction

Biofuels, such as wood logs and wood pellets, offer numerous advantages over traditional hydrocarbon fuels when it comes to residential heating and cooking. Many regulatory bodies have implemented restrictions on fossil fuels, encouraging the use of biomass combustion appliances. For example, the EU has enacted a goal that by 2030, 32% of residential energy usage will come from renewable sources, such as wood pellets [1]. Currently, agencies have been focused on reducing greenhouse gases, like CO2, due to their tie with ozone depletion [2]. Biomass combustion typically is regarded as "carbon-neutral" since CO2 emissions can be offset by planting more trees. While such regulations show improvements in relation to ozone, they often result in unintended side effects and neglect to assess exposure of the harmful emissions from a personal health perspective. The research group Corsini et al. asserts that biomass combustion contributes to at least 10% of outdoor harmful

particulate matter in select areas [3]. Given that biomass combustion introduces higher levels of harmful emissions than those associated with fossil fuel combustion it is particularly important to understand the health effects of high exposure appliances like pellet stoves, boilers and grills [2].

While the base technology underlying wood pellet stoves, boilers, and grills has existed for decades, recent work has explored methods to limit harmful emissions into the atmosphere and into our bodies. This review characterizes some of these harmful emissions often found in biofuel home appliances and describes several ways to limit them.

Wood pellet appliances typically consist of a pellet hopper, auger, burn pot, and combustion chamber. The auger usually supplies the pellets from the hopper to the burn pot at a fixed rate for continuous mass flow, although some appliances are user-controlled or have a closed-loop feedback system that monitors temperature to determine mass flow rate. Once the pellets fall into the burn pot, a primary air source and ignitor create the combustion reaction. Figure 1 is a schematic of a typical pellet stove.



Figure 1: Schematic of a Pellet Stove [4]

2. Measurement Systems

2.1 Gas

Gas measurements, particularly concentrations of CO, are done with gas analyzers. One example is the Siemens Ultramat series. Before flowing into the Ultramat, the flue gas is cooled and diluted to avoid damaging the device. The device contains an infrared light and operates on the push-pull alternating light principle. When the gases move through the infrared light, different frequencies of IR are absorbed. Each

species absorbs a specific wavelength, allowing the device to calculate concentrations. One issue with this type of device is that with a gas mixture, some spectra may overlap [4,5].

2.2 Particulate Matter

One of the most common particle measurement tools is the Dekati Electrical Low-Pressure Impactor Plus (ELPI+), shown in Figure 2. It can capture particles from 6 nm to 10 μ m [5]. The device collects aerosols via a sampling probe, and then it dilutes them with heated air to avoid condensation [6]. Condensation would skew the results due to potential electrical shorting or conglomeration of individual particles [6]. Once diluted, particles are then electrically charged to a known charge level and passed through a low-pressure cascade impactor where they are separated based on their aerodynamic diameter. The ELPI+ has 14 stages of particle size classifications, where larger particles are collected in the first stages and smaller particles are collected in later stages [5]. Each stage is equipped with electrometers that can then detect the current of the charged particles [5]. The electrical current of each impactor stage is proportional to the concentration of particles, and thus the concentration for each size range can be calculated by the ELPI+. Dekati makes several similar instruments, such as the DLPI and original ELPI. These instruments work in a similar manner to the ELPI+ but have slightly different targeted size ranges. However, the ELPI+ seems to be the most common for biomass combustion experiments [7].



Figure 2: Dekati ELPI+ [5]

3. Experimental

3.1 CO

Carbon monoxide emissions are associated with many types of combustion and have been studied for quite some time. CO's toxicity is a function of both exposure time and concentration [8]. OSHA recommends a maximum of 50 ppm (or 573 mg/Nm3) for "continuous exposure," however, an increase in PPM has an exponential effect on toxicity to the human body [8]. Certain levels of CO, particularly above 400 ppm (458 mg/Nm3), can be life-threatening [8]. In regard to pellet appliances, CO tends to form from incomplete combustion with low combustion temperatures; higher combustion temperatures allow intermediate reactions to complete and eliminate the CO.

It's been shown that pellet appliances can be run in a variety of conditions by changing the operating parameters. While pellet stoves and boilers tend to favor lean combustion conditions, it's difficult to pin down an "ideal" excess air ratio (λ) for pellet appliances. The excess air helps increase combustion temperature up to a certain point [7]. However, beyond that point, the excess air simply absorbs energy, reducing the combustion temperature again [7]. For example, Obaidullah and De Ruyck found that their ideal excess ratio for a specific 5 kW pellet stove was roughly λ =2.5. They used varied fan speeds to create different excess air ratios and found that higher fan speeds generally yielded lower CO concentrations [7]. However, due to variations in appliance type, pellet quantity/type, humidity, etc., it can be difficult to reduce the ideal ratio to a single number.

In another experiment, Atkins et al. found that running a pellet boiler at part load drastically reduced the combustion temperature [9]. They ran a 50 kW pellet boiler at both partial and full load conditions; the partial load combustion temperatures ranged from 500-700C, while the full load combustion temperatures measured 900-1100C [9]. Obaidullah and De Ruyck confirmed why this phenomenon occurs, namely because the part-load combustion experiments had higher air excess ratios past the "tipping point" described above. These extremely high excess air ratios actually caused the air to absorb energy from the reaction, leading to an increase in CO concentration [7]. The part-load scenario was created by reducing auger speed, meaning mass flow rate was lower and pellets were dumped more slowly into the burn pot [7]. However, the fan speed was constant, so the excess air ratio increased (roughly λ =4.35) as the mass flow rate decreased. This relationship between CO emissions and load held for other types of appliances, as Schmidl et al. confirmed with a 6 kW pellet stove and a 40 kW pellet boiler [10].

In Figures 3 and 4 below, it can be seen that fan speed has a dramatic effect on CO emissions. The only factor changed between experiments C/D and E/F was fan speed, and the CO emissions dropped by roughly 50% when the fan was increased from 1250 rpm to 1400 rpm [7].

Experiment	Stove Load (kW)	Fan Speed (rpm)
А	2.5	900
В	2.5	900
С	5	1250

D	5	1250
E	5	1400
F	5	1400

Figure 3: Summary of Obaidullah and De Ruyck's Experiments [7]



Figure 4: CO Emissions for an Entire Cycle [7]

In addition to varied load conditions, researchers have shown that cycle phase has a dramatic effect on CO production [7]. A full cycle contains 3 phases: startup, combustion, and burnout. Each phase has slightly different combustion conditions and therefore produces different levels of emissions.

In the startup phase, the combustion temperature begins rising, as the pellets are igniting and the flame is spreading. As expected, Obaidullah and De Ruyck found this phase to yield high CO concentrations, higher than those measured in the combustion phase. Concentrations in their experiments reached roughly 2300 mg/Nm³ during startup, even in a full-load condition [7]. Schmidl et al. confirmed this result, demonstrating that the startup phase generated up to 40X more CO than the combustion phase [10].

With constant combustion temperature, mass flow rate, and heat generation, steady-state conditions are achieved, and the combustion phase begins. Combustion is typically the longest of the three stages. Some stoves may have an adjustable feed rate (mass flow rate), but the experiments examined here kept the auger at a constant speed. In experiments by Obaidullah and De Ruyck, the combustion phase produced the lowest CO emissions, maximizing at 1450 mg/Nm³ for part-load and 145 mg/Nm³ for full load [7]. This is to be expected, since the stove/boiler performance is designed primarily for the longest phase of use. The excess air ratio (and therefore mass/air flow rates) are designed to produce maximum efficiency during this phase, so it's expected that emissions are relatively low.

Once the combustion phase completes, the burnout phase starts. This phase creates the highest CO concentrations in any wood pellet stove/boiler. The auger generally stops feeding pellets to let the flame "burn out," increasing the excess air ratio higher from the combustion phase. This generally creates the case where λ is too high, and the excess air incidentally cools the combustion area [7]. As mentioned earlier, lower combustion temperatures lead to incomplete combustion, the perfect breeding ground for CO [7].

These aggregate levels of CO pose a health concern, especially when run for two hours or more in a poorly-ventilated area. When properly ventilated, it's likely that manufactured pellet stoves/boilers do not pose a significant CO threat to human health. However, many homes use hand-built stoves that may experience irregular combustion conditions and lack proper ventilation, so this concern cannot be written off without indoor air testing near such appliances.

3.2 PM

Particulate matter (PM) poses a significant threat to human health due to the fact that the smallest particles can enter tiny capillaries in the human lungs. Particulate matter is classified based on aerodynamic diameter, or an approximate diameter of the particle based on a 1 g/cm^3 droplet with similar properties as the flue gas [6]. Since particles do not hold a perfect spherical shape while in transit, the aerodynamic diameter is a best estimation. Three of the most common particulate matter classes are $PM_{0.1}$ (<0.1 µm), PM_1 (<1 µm), and $PM_{2.5}$ (<2.5µm) [7]. Compared with traditional hydrocarbon fuels like natural gas, wood pellet combustion produces exceedingly high concentrations of PM emissions [7]. These PM emissions, particularly PM_1 and smaller, greatly increase the risk of many adverse health conditions [11]. Studies have shown that particulate matter from wood fuel causes dramatic increases in cardiac disease, lung cancer, chronic obstructive pulmonary disease, and birth defects [11].

While many regulatory agencies have traditionally written standards in the context of a mass concentration limit for particulate matter, recent work has switched to a molar concentration. Since particulate matter encompasses a variety of species, a molar concentration is more useful when evaluating risk level, especially for small classifications like PM_{0.1} [3]. Another trend is to report particulate matter classes as a proportion of total particulate matter.

PM_{0.1}, also known as "ultrafine" particles, pose a unique concern due to their low weight; this "weightlessness" allows them to travel further in the air than other particulate matter. Therefore, location/placement of a pellet stove/boiler is not as effective in reducing these emissions, increasing the risk of indoor inhalation [3]. Most of these ultrafine particles are small bits of ash, containing large amounts of zinc, potassium, and sulfur [3]. PM_{0.1} has an added risk that larger particulate matter does not: the ability to be absorbed into the skin. This pathway provides even more of a chance for toxic/carcinogenic substances to enter the body, compared with larger particulate matter.

While $PM_{0.1}$ particles do not constitute a significant portion of particulate mass, they dominate in terms of molar concentrations [3]. Corsini et al. found that $PM_{0.1}$ constitutes 77% of the total number particle concentration in emissions from residential wood burning [3]. While exact percentages vary, Obaidullah and De Ruyck confirmed this finding. In Figure 5, it can be seen that the curves peak right

around 0.1 μ m, indicating this number dominance. This trend was also found in the startup and combustion phases [7].



Figure 5: Particle Size Distribution [7]

Despite this significant presence, there is very little research on $PM_{0.1}$, likely due to the difficulty and cost of measuring it compared with PM_1 and $PM_{2.5}$. However, the high health risk associated with $PM_{0.1}$ justifies the need for more research.

As with the CO concentration studies, particle mass concentrations varied with different phases of the combustion cycle, heat loads, and fan speeds of the pellet stoves. As shown in Figure 6, Obaidullah and De Ruyck found the highest PM concentrations generally in the steady-state combustion phase, followed by the startup phase when the stove was running at full load. Greater fan speeds had a beneficial effect to PM containment, possibly due to higher combustion temperatures from excess air and better mixing [7].



Figure 6: Mass Concentration of All Phases [7]

Figure 6 documents Obaidullah and De Ruyck's study results for PM_{2.5} emissions; the PM₁ results followed the exact same pattern. The least favorable conditions for PM₁ and PM_{2.5} emissions were full load with a slower fan speed, while higher fan speeds showed improvements in both CO and PM emissions. Partial loads actually proved beneficial for PM emissions despite increasing CO presence. Obaidullah and De Ruyck suspect that this is due to lower fuel consumption [10].

This presents an interesting decision, asking the question of whether running stoves at partial loads is a viable solution for reduced PM emissions. However, not all pellet stoves have the option to be run at a limited load, so other methods must be employed to address the issue. Additionally, running the appliances at partial load may introduce new problems not discussed here.

Earlier research by Obaidullah et al. produced similar results, with PM₁ mass concentrations exceeding 300 mg/Nm³ for the startup and combustion phases [12]. Consistent with other research, the startup phase gave way to a large spike in PM₁[12]. While there are too many factors to give a definitive PM average (wood type, pellet quality, fan speed, load, etc.), pellet stoves are capable of producing PM₁ emissions orders of magnitude higher than PM₁ found in the ambient [13]. Even densely-populated areas of China, a country notorious for pollution, recorded PM₁ concentrations of 71.7 µg/Nm3 [13]. Very few PM₁ regulations exist in the world, but Chen et al. found that PM₁ concentrations around 20 µg/Nm³ still have associated risk of hospitalization [13]. It's clear that with levels that are orders of magnitude above "ambient" PM₁ concentrations, wood pellet stoves should be scrutinized and engineered to reduce PM₁ concentrations.

While $PM_{2.5}$ is not as damaging to the human body as PM_1 or $PM_{0.1}$, $PM_{2.5}$ emissions still represent a material health hazard. There are many more standards and regulations related to $PM_{2.5}$; for example, the EPA recommends no more than 35 µg/Nm³ for a 24-hour period [14]. Similar to PM1, wood pellet combustion produces concentrations multiple orders of magnitude above recommended levels. However, researchers observed $PM_{2.5}$ concentrations of nearly 400 mg/Nm3, more than 10X the recommended limit [7].

While it's difficult to characterize PM emissions from wood pellets with a single number, researchers have found general trends across various experiments. Factors such as equivalence ratio, residence time, pellet quality, fuel-air mixing, and appliance structure all influence the mass concentrations of PM. These concentrations, particularly for PM₁ and PM_{2.5}, appear to be orders of magnitude larger than baseline/ambient values (and guidelines for PM_{2.5}), even in crowded/polluted cities [13]. It's difficult to estimate how well ambient air will dilute these emissions, and it depends on ventilation [12]. Therefore, it is still worthwhile to explore reduction methods and technologies.

3.3 PAH

Polycyclic aromatic hydrocarbons (PAH) are a group of hydrocarbons characterized by five or sixmember carbon rings fused together, such as in Figure 7 [9]. They contain known carcinogens and toxins, such as benzo[a]pyrene and napthalene [15].



Figure 7: Common PAH Species [16]

PAH can take the form of either particulate matter or gas, so some of these emissions are included in discussions above. Atkins et al. estimate that up to 98% of PAH mass exists in the gas phase, but this is dependent on appliance type and operating conditions [9]. However, it's important to note that not all PM emissions are classified as PAH.

OSHA recommends combined exposure limits of no more than 0.2 mg/m³ combined for 5 of the most harmful PAH species: benzo[a]pyrene, anthracene, pyrene, chrysene, and phenanthrene [17]. This applies to PAH in both the gaseous and solid phases.

Atkins et al. confirmed that PAH emissions follow the relationship between heat load and concentrations. They found that PAH emissions, like CO and PM, increased when the boiler/stove ran at a partial load, increasing the likelihood of incomplete combustion [9]. Since PAH refers to various species and sizes, it's not as straightforward to compare raw data. PAH measurements from the same experiment can be characterized differently based on measurement device or species observed. For example, some PAH particulates may fall outside the size range of the measurement device and will therefore be excluded.

Despite this logistical challenge, Hays et al. compiled data from 4 different studies, each using a wood pellet boiler and observing 8 of the exact same PAH species [18]. These 8 species represent half of the EPA's 16 "high-priority" PAH pollutants [14, 16] While the boilers and pellets used were not exactly the same, Hays et al. found that these 8 species comprised up to 10% of particulate matter emitted [15]. While the studies produced dramatically varying results that are hard to characterize, the preliminary findings imply the need for further investigation. Referencing the particulate matter research by Sippula et al. discussed above, even a PAH/PM fraction of 1/100 would exceed the recommended OSHA levels for PAH exposure [15]. Vincente et al. found that wood pellet stoves can generate over 100 mg/m³ of pyrene alone, one of the 5 PAH species on OSHA's list to avoid [18].

Due to this inconsistency in species examined and appliance type, trends should be observed rather than raw numbers. Consistent with the CO and PM research described above, PAH species form mostly from incomplete combustion at lower combustion temperatures [9]. The ideal combustion temperature for PAH formation is between 700 and 900C, as PAHs lose stability at higher temperatures [9]. As with CO and PM, PAH species are likely to form in higher quantities at startup and burnout, as the combustion temperature is lower compared with the main combustion phase. Additionally, Atkins et al. demonstrated that the part-load experiments generate the highest concentrations [9]. This is to be expected, as previous discussion has shown that partial loads often have lower combustion temperatures.

4. Reducing Emissions

Recent work has been done on the subject of reducing emissions from biomass fuels, particularly wood pellet stoves and boilers. The challenge in implementation is typically associated with cost, as the cost of the solution may be higher than the cost of the appliance itself [19]. Primary reduction techniques, such as changing the burn pot shape, refer to ways in which emissions are reduced from the initial combustion stage. Secondary measures, such as adding a catalytic combustor, refer to methods that reduce emissions after the primary combustion but before the smoke exits the appliance.

4.1 Burn Pot Shape

Polonini et al. found that the shape of the burn pot can have a profound effect on CO and PM emissions [20]. They fabricated and tested 2 versions of their innovative pot: a shallow version and a deep version, shown in Figure 8. Instead of passing air through the bottom via holes like most traditional burn pots, this design passes air along the sidewalls [20]. Secondary airflow travelled through the holes at the top of the burn pot. For the shorter burn pot, CO emissions dropped as low as 8 mg/Nm³ for a 30-minute test [20]. The average CO measure over 19 tests was 20 mg/Nm3, but the data was skewed by a few outliers, yielding a standard deviation of 27 mg/Nm3. Polonini et al. attribute this to improperly cleaning the burn pot between cycles and having residual ash combust [20]. The deeper burn pot produced a similar mean (28 mg/Nm3), but the standard deviation was still rather large at 15 mg/Nm3. These experiments should be repeated with proper cleaning to gather more data points; regardless, these levels of CO represent a monumental improvement over traditional pellet burn pots.

In contrast with CO, PM saw a slight improvement with increased depth, averaging 15 mg/Nm³ for the shallow pot and 11 mg/Nm³ for the deeper pot [20]. Both standard deviations for the PM are ~3.0 mg/Nm3, indicating cleaner data than we saw with the CO measurements. The CO levels for both pots are so low (compared to other pellet appliances) that it's fair to say the innovative shape and airflow may be good enough at either CO level. However, the PM results are more significant; the drop in PM may indicate the benefit of a larger distance between primary and secondary airflow. Polonini et al. suspect that the secondary airflow interferes with the primary airflow in the shallow burn pot, generating locally-rich conditions [20]. Additionally, Polonini et al. suspect that the deeper burn pot generated a straighter airflow, preventing entrapment of PM within the flowing air [20].



Figure 8: Innovative Burn Pots [20]

4.2 Catalytic Combustors

Catalytic combustors are another tool to reduce emissions from biomass appliances. Catalysts operate at high temperatures, which can be a challenge to produce; the flue gas temperature for biomass fuels is relatively low compared with hydrocarbon fuels [19]. Placing the combustor close to the burn pot helps keep the operation temperature high, but the startup phase may still be too cold for the catalyst to take effect [19].

As seen in Figure 9, A typical catalytic combustor is a wire mesh made of platinum and palladium. As the hot flue gas passes through the mesh grate, the platinum and palladium react with the combustion products, removing them from the gas [19]. Hukkanen et al. found that CO emissions were reduced by 21% for the overall cycle, with the most significant reductions coming in the burnout cycle [19]. PM₁ emissions were reduced by 30% [19]. Due to their simplicity, catalytic combustors are a good option for small-scale appliances.



Figure 9: Example Catalytic Combustor [19]

4.3 Porous Material

Rico et al. found that certain porous materials can greatly reduce particulate matter in biomass emissions. The ceramic porous inert material (PIM), seen in Figure 10, allows reactions to occur in small cavities at higher-than-normal combustion temperatures due to the conductivity and emissivity of the PIM [2]. The researchers placed zirconium oxide ceramic foam at 2 different distances from the combustion bed, searching for the optimal location [2]. As seen in Figure 11, they also tested both single-and double-layered PIM, with each layer being 30 mm thick [2]. Particulate matter from .03 to 10 μ m was collected and analyzed [2].



Figure 10: Porous Inert Material (PIM) [2]



Figure 11: Cross-section of PIM placement [2]

Rico et al. achieved the greatest PM reductions with the single-layered PIM placed 380 mm above the burn pot [2]. Three different air flow rates were tested, and reductions with this single-layer configuration ranged from 57% to 64% [2]. Despite these great reductions, CO levels remained the same

with single-layer PIM and actually increased with double-layer PIM [2]. Single-layer PIM yielded hotter gas temperatures, while double-layer PIM actually produced lower flue gas temperatures. This coincides with the PM results, as CO and PM form in cooler temperatures [2].

4.4 Staging

Air staging has been shown to reduce harmful emissions in biomass combustion appliances. Air staging occurs when a secondary air source causes additional combustion reactions beyond the initial burning of the pellets in the burn pot. As discussed above, pellet appliances can suffer from poor air-fuel mixing due to the irregular shape and distribution of pellets [17]. This poor mixing leads to incomplete combustion and low combustion temperatures. Since many harmful emissions like CO form from incomplete combustion, a second "stage" gives these intermediate products another chance to combust and form more stable products, such as CO2 [17]. Figure 12 shows a typical setup of a two-stage reactor. Secondary air enters at Zone III and allows additional reactions to take place outside of the primary combustion zone, Zone II [17].



Figure 12: Air Staging Schematic [17]

The addition of secondary air increased the flue gas temperature by up to 93C in Zone IV, indicating the additional combustion reactions [17]. Both CO and PM emissions dropped by over 50% with a second stage added, showing variation with secondary air speed, location of secondary air injection, and air inlet shape [17].

Deng et al. tested several different air ratios, or the flow rate of primary air (PA) to secondary air (SA) [21]. They found that relatively-even ratios of PA to SA produce the lowest CO and PM_{2.5} emissions,

suggesting the range from 4:6 to 6:4 [21]. They hypothesized that too much SA inhibited the secondary reactions that eliminate CO and other radicals, while too little SA would not do enough to alleviate poor mixing [21]. However, based on other literature reviewed in their study, this ratio is dependent on appliance and runtime and should be verified for each combustion condition [21]. Depending on appliance shape and style, air staging could provide a relatively simple and cost-effective solution for reducing harmful emissions.

5. Summary and Conclusion

In summary, it's clear that CO, PM, and PAH emissions follow similar trends in combustion conditions. They form at lower combustion temperatures, which can happen for a variety of reasons. Partial loads, lower fan speeds, and poor mixing all yield low combustion temperatures. Air-fuel ratios also have an effect; pellet appliances generally yield the lowest emissions when lean, but there is a "critical point" where extra excess air increases emissions again. This critical point varies by appliance and cannot be pinned down to a single number. Based on the work of Obaidullah and De Ruyck, λ =2.5 would be a good place to start if a researcher wanted to investigate this further.

Phase of combustion is also important to harmful emissions. The startup and burnout phases typically have lower combustion temperatures and therefore higher concentrations of harmful emissions. The air excess ratio of these phases is higher than the combustion phase due to lower mass content. In startup, the fuel mass is still increasing, while the fuel mass is dwindling in burnout. The steady-state combustion phase generally has the lowest emissions, which makes sense considering that the operating parameters (air-fuel ratio, temperature, etc.) are designed for this lengthy phase. However, the harmful emissions in startup and burnout were so large compared to the middle phase (up to 10X) that they had a significant impact on total/overall emissions.

The research presented here shows that wood pellet appliances can produce CO, PM, and PAH at levels well above regulatory limits. CO emissions in particular are heavily researched and regulated, but PM emissions (particularly PM_{0.1} and smaller) are not. Additionally, many PM regulations are written in terms of mass concentration, but this approach undermines the fact that smaller particles tend to do more damage. The US and the EU governments do not have any regulations around PM_{0.1}, which is considered the most harmful to the human body. There is not as much literature around PM_{0.1} compared with larger classifications, which is surprising given its toxicity. This alone justifies the need for further research and understanding around ultrafine particulate matter. Experiments need to be conducted that measure molar concentrations of PM_{0.1} alone, and those results need to be compared with current medical research using the same measurement system.

PAH is implicated as well, since PAH can take the form of particulate matter and is very toxic in small quantities. PAH needs more research examining total amounts of both the gaseous and particulate phases, as regulations tend to focus on just the PM form.

In addition to continuing research in CO, PM, and PAH emissions, experiments should be conducted to address both primary and secondary forms of emissions reduction. Many appliances lack these additional techniques due to added cost or complication, which may implicate the target market segment, margins, or consumer. However, the solutions presented here can be implemented relatively easily and should be examined in the context of a real-life product. Pellet appliances should be outfitted with one or multiple of the solutions described, and emissions measurements (both molar and mass) should be taken at various proximities from the grill over a period of time. This will indicate the true impact of the pellet fuel and the impact it has on human health.

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Appendix A: Research Proposal

This proposal is seeking funding to expand and advance testing of emissions from wood pellet appliances, specifically stoves, boilers, and grills. The demand for wood pellets has grown by roughly 10% per year for over the past decade, and the trajectory is expected to continue to grow [22]. The increasing regulations around carbon footprint are driving this growth, as regulators are heavily focused on recapturing emitted carbon dioxide (CO₂) and protecting the ozone layer. Biomass fuels, like wood pellets, excel at this; however, a lack of thorough research raises the question of whether additional problems are being introduced at the same time.

I discovered that wood pellet combustion actually generates higher amounts of particulate matter (PM) than fossil fuel combustion produces [2]. In addition, the nomenclature and regulations around PM tend to use mass concentrations rather than molar concentrations. Why is this a problem, you might ask? The smallest classifications of PM, such as PM_{0.1} are proven to be the most dangerous to human health due to the particulate's ability to penetrate deep into the lungs and become trapped [3]. Given their small size, PM_{0.1} tends to have low mass as well; however, research has shown that PM_{0.1} can dominate total PM in terms of number concentrations, comprising up to 77% of particulate matter [3]. If these small, lightweight particles dominate by number concentration, but only the mass concentrations are considered, then it's likely that exceedingly high amounts of PM_{0.1} are not being detected.

This project will capture PM_{0.1} and PM₁ concentrations from various wood pellet appliances, reported in both mass and molar concentrations. The appliances will be run at various operating conditions, such as partial/full load, high/low excess air ratios, and varying pellet types. Additionally, readings will be taken at various proximities from the appliances, since PM_{0.1} is virtually "weightless" in the air and can stay suspended for quite some time [3]. This will allow a greater understanding of the true impact of using these appliances on an everyday basis.

After the baseline tests are completed, this project will add various PM mitigation solutions to the appliances and repeat the trials. Solutions proposed include adding a catalytic combustor, a porous ceramic material, a uniquely-shaped burn pot, and a secondary airflow.

At the end of the project, I will have concrete data to share a better understanding of how safe pellet fuel truly is and if we are accomplishing the goal of a reduced carbon footprint without creating significant health hazards at the same time.

Appendix B: Homework Problem

a. Assume wood pellets are made entirely of cellulose. Write out the stoichiometric equation for combustion of cellulose.

$$C_{6}H_{10}O_{5} + a(O_{2} + 3.76N_{2}) \rightarrow xCO_{2} + yH_{2}O + 3.76aN_{2}$$

$$C:6 = x$$

$$H:10 = 2y \rightarrow y = 5$$

$$O:5 + 2a = 2x + y \rightarrow a_{s} = 6$$

$$C_{6}H_{10}O_{5} + 6(O_{2} + 3.76N_{2}) \rightarrow 6CO_{2} + 5H_{2}O + 22.56N_{2}$$

b. Obaidullah and De Ruyck found the ideal air excess ratio for their 5 kW stove was λ =2.5. Find the moles of air needed for this equation.

$$\lambda = \frac{1}{\phi} = \frac{a}{a_s}$$
(B.1)
$$2.5 = \frac{a}{6}$$
$$a = 15 \text{ moles}$$

c. Find the balanced chemical reaction, assuming λ =2.5. Write the full stoichiometric equation.

fuel\$ = 'C6H10O5'; phi = 1/2.5; Treac = 298.15

"Options - Unit System - Molar Basis and Kelvin degrees" "Main Reaction: C6H10O5 + a(O2 + 3.76N2) --> xCO2 + yH2O + fO2 + 3.76aN2"

5 + 2*a = 2*x + 2*f 10 = 2*y 6 = x 2.5 = a/6

Results:

a = 15 f = 11.5 x = 6 y = 5

$$C_6H_{10}O_5 + 15(O_2 + 3.76N_2) \rightarrow 6CO_2 + 5H_2O + 11.5O_2 + 3.76(15)N_2$$

d. Calculate the mass flow rate of pellets into the pellet stove. Assume enthalpy of formation of cellulose to be 1413.2 cal/g and the stove to have an efficiency of 92%. Assume cellulose has an HHV of 19,000 kJ/kg. All reactants are at 298K and 1 atm.

$$\frac{1413.2 \ cal}{g} * \frac{4.184 \ J}{cal} * \frac{162 \ g}{mol} = 957742.7 \frac{J}{mol}$$

$$\eta = \frac{Q_{actual}}{Q_{max}} = \frac{Q_{actual}}{\dot{m}_F HHV}$$

$$\dot{m}_F = \frac{Q_{actual}}{\eta HHV}$$

$$\dot{m}_F = \frac{5 \ kW}{.92 * 19,000 \frac{kJ}{kg}}$$

$$\dot{m}_F = .000286 \frac{kg}{s} = 1.029 \frac{kg}{hr}$$

e. Use HPFLAME to find T_{ad}

Input:

- 06 /CARBON ATOMS IN FUEL
- 10 /HYDROGEN ATOMS IN FUEL
- 05 /OXYGEN ATOMS IN FUEL
- 00 /NITROGEN ATOMS IN FUEL
- 0.4 /EQUIVALENCE RATIO
- 2000. /TEMPERATURE (K) (Initial Guess)

101325.0 / PRESSURE (Pa)

957742.3 /ENTHALPY OF REACTANTS PER KMOL FUEL (kJ/kmol-fuel)

Output:

CARBON ATOMS	6.0	
HYDROGEN ATOMS	10.0	
OXYGEN ATOMS	5.0	
NITROGEN ATOMS	.0	
EQUIVALENCE RATIO	.400	
TEMPERATURE (K) guess	2000.0	
PRESSURE (Pa)	101325.0	
ENTHALPY OF REACTANTS	(kJ/kmol fuel)	957742.3

FLAME TEMP. & COMBUSTION PRODUCTS PROPERTIES

Flame Temperature [K] = 1952.78

Mixture Enthalpy [J/kg] = .4310E+06 Mixture Specific Heat, Cp [J/kg-K] = .144835E+04 Specific Heat Ratio, Cp/Cv = 1.2479 Mixture Molecular Weight [kg/kmol] = 29.0709 Moles of Fuel per Mole of Products = .01308272

The mole fractions of the product species are:

H: .00000721 O: .00015525 N: .00000000 H2: .00003824 OH: .00110938 CO: .00020115 NO: .00509974 O2: .11496115 H2O: .06481708 CO2: .07829518 N2: .73531563

f. Use the mass flow rate found in d to estimate the LHV. Does this seem right?

$$P = LHV * \dot{m}$$

$$5000 \frac{kJ}{s} = LHV.000286 \frac{kg}{s}$$

$$LHV = 17,482.5 \ kJ/kg$$

Yes, this seems correct. Check:

$$HHV = LHV + \frac{n_{H2O}}{n_{fuel}} * h_{fg,H2O}$$
$$HHV = 17,482,517 + \frac{5}{1} * 407,000 = 19,522,517$$

Yes, this seems about right since it's close to our initial HHV (19,000,000 kJ/kg)

Appendix C: Literature Search

My literature search started with Google Scholar, which I accessed using our Purdue library link. The repeated keywords I found myself using were as follows: "biomass," "emissions," "pellet," "PM," "particulate", "CO," and "PAH." I did not expect to search with "biomass" so much, but it turns out that many publications referred to wood pellets and woodchips collectively as "biomass." I know that other true biomass fuels like straw or bark can fall into this category, but the majority of authors I stumbled upon had a strong focus on wood pellets. One paper that provided the backbone for most of my research and referencing was the work done by Obaidullah and De Ruyck [6]. This paper provided exactly the type of content I was looking for, so the I have at least 3 sources from forward referencing, and 3 from backward referencing. This is why my sources' date ranges span quite a range. Additionally, the Elsevier "Recommended articles" sidebar on the right provided several additional references that made it into my final paper. I would estimate that roughly 65% of my sources came from a raw search, and the remaining 35% came from forward/backward referencing or a "recommended article" feature.