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Experimental Investigations of the Effects of Secondary Air Injection on Gaseous Emission Profiles (NO_x, NO, NO_2, CO) and Hydrocarbons (C_xH_x) in Cookstoves Using Charcoal from *Eucalyptus glandis*

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Abstract

Use of biomass in domestic cookstoves leads to the release of oxides of nitrogen (NO_x), nitric oxide (NO), nitrogen dioxide (NO_2), carbon monoxide (CO) and hydrocarbons C_xH_x that can be detrimental to health of the public and the environment. Attainment of complete combustion is the best strategy for mitigating the release of these emissions. This study sought to experimentally determine the effects of secondary air injection on the emission profiles of NO_x (NO & NO_2), CO and C_xH_x in a charcoal operated cookstove. Charcoal from *Eucalyptus glandis* was bought from Kakuzi PLC. Composites from three batches were analyzed for chemical composition and the stoichiometric air equivalent. Proximate analysis data show that the charcoal composed 58.72% ± 3.3% C, 15.95% ± 1.2% Volatile Matter, 4.69% ± 0.55% Moisture, 20.7% ± 0.8% Ash, High heat value (HHV) of 30.5 ± 1.1 and 29.3 ± 1.3 Low heat value (LHV) (MJ/kg) with a chemical formula of $C_{18}H_2O$ and a stoichiometric air requirement of 5.28 ± 0.6 $\mathrm{m_N^3\ air/h}$ with a fuel flow rate of 1 kg fuel/hr. Emission profiles for CO and C_xH_x reduced significantly by 70% and 80% respectively with secondary air injection whereas those of NO_x increased by between 15% and 20% for NO_2 and NO. The study reveals that secondary air injection has potential to mitigate on emission release, however other measures are required to mitigate NO_x emissions.

Keywords

Biomass, Stoichiometric, Emissions, Combustion, Pollution, HAP, Cookstoves

1. Introduction

Biomass is a major form of fuel for approximately 80% of the Kenyan population and is expected to remain the main source of energy for the foreseeable future [1]. More than 34.3 million tons of biomass is used annually of which 15.1 million tons is in the form of fuelwood while 16.5 million tons is charcoal [1]. Fuelwood supplied 89% of rural energy with a per capita annual consumption of 741 kg and 7% urban household energy with a per capita annual consumption of 691 kg and cottage industry, 1.3 million tons. Charcoal supplies 82% of urban household energy with a per capita annual consumption of 152 kg, while for rural households, 34% with a per capita consumption of 156 kg [2].

1.1. Combustion

Combustion is widely used for energy production both in industry and in household applications. The combustion conditions in the household stoves have a crucial influence on the emissions [3]. The traditional stove for cooking has a very low energy efficiency of 10% - 15% and high emissions, while the traditional stoves used for space heating in rural areas have an energy efficiency of 25% - 30% [4]. Low combustion efficiencies result in energy wastage and pollutant emissions due to insufficient air supply and poor mixing between air and fuel, air is the common source of oxygen [5].

1.2. Combustion Phases

Combustion consists of three relatively distinct but overlapping phases: Preheating phase, when the unburned fuel is heated up to its flash point and then fire point. Flammable gases start being evolved in a process similar to dry distillation. Distillation phase or gaseous phase, when the mix of evolved flammable gases with oxygen is ignited. Energy is produced in the form of heat and light. Flames are often visible. Heat transfer from the combustion to the solid maintains the evolution of flammable vapors. Charcoal phase or solid phase, when the output of flammable gases from the material is too low for persistent presence of flame and the charred fuel does not burn rapidly and just glows and later only smolders [6].

1.3. Biomass Burning Related Pollution

Despite its wide use, biomass burning is associated with household air pollution (HAP) where users are continually exposed to compounds of incomplete combustion known to have detrimental effects to human health [7] [8]. Combustion of wood and other biomass is a significant contributor to poor air quality in many developing countries [9]. Emissions of particulates and Polycyclic Aromatic Hydrocarbons (PAH) are a major health hazard, particularly in Africa where the use of domestic cookstoves has increased alongside population expansion [10]. HAP is associated with significant global morbidity and mortality. Carbon monoxide and polycyclic aromatic hydrocarbons (PAH) e.g., Benzo[a]pyrene (**Figure 1**) are the major HAP pollutants from combustion [11].

PAHs are known carcinogens suspected to be responsible for the growing cancer cases in Kenya [9]. Carbon monoxide binds with hemoglobin to form carboxyhemoglobin thus displacing oxygen leading to asphyxiation [11]. Emissions adversely affect the occupational health of workers thus reducing their productivity in workplaces [12]. Other effects associated with HAP include; increased blood pressure (BP), dyspnea, childhood pneumonia, lung cancer, low birthweight and cardiovascular diseases, causing diseases that lead to approximately 2 million deaths annually [7].

Emissions from cookstoves depend on the cookstove, fuel type, cooking practices, ventilation, tree species and season of the year. Biomass harvesting has led to forest degradation and loss of forested area which has reached less than world accepted coverage of 10% [1].

2. Materials and Methods

2.1. Feed Stock Characterization

Charcoal was bought from Kakuzi PLC, a company that produces charcoal for sale using *Eucalyptus glandis* as the main plant species. Experimental sample was prepared by forming composites from three batches bought at different times during the months of January to April 2022. This allowed for sampling during the dry and wet season. Samples were analyzed in triplicates. The experiments were conducted with the charcoal as received; no further treatments were done.

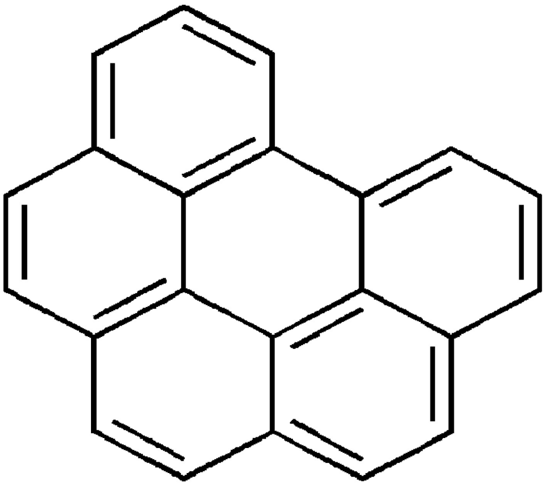

Figure 1. Benzo[a]pyrene.

2.2. Proximate Analysis

Proximate analysis gives the gross composition of the biomass. ASTM standards are applicable for determination of the individual components of biomass:

- Volatile matter: E-872 for wood fuels
- Ash: D-1102 for wood fuels
- Moisture: E-871 for wood fuels
- Fixed carbon: determined by difference

Analysis was done at the Kenya Industrial Research Institute (KIRDI) to determine the chemical composition of the composite sample. The analysis determined the; moisture content (M), volatile matter content (VM), fixed carbon content (FC), and the ash content using a proximate analyzer.

Moisture was driven off at ~105˚C - 110˚C, Volatile matter is driven off in an inert atmosphere at 950˚C, using a slow heating rate. The ash content was determined by taking the residue and burning at above 700˚C in oxygen [13]. The fixed carbon is then determined by mass difference:

$$FC = 1 - M - Ash - VM \tag{1}$$

where;

FC is fixed carbon

M is fixed carbon

VM is fixed carbon

On a dry basis determination, moisture content is removed from the VM and ash contents first [11].

$$FC = 100 - M\% - A\%dry - VM\%dry \tag{2}$$

where;

A is ash content

2.3. Stoichiometric Air Equivalent/Theoretical Air

The stoichiometric volume of oxygen required for complete combustion of fuel mass unit is obtained by summing volumes of oxygen required for combustion of fuel components, from the preceding equations. It can be noticed the presence of oxygen in fuel composition, having a mass participation of (O^i) and this quantity should be no longer introduced from the outside into the furnace [14]. According to Avogadro's law, at normal state (pressure of 760 Torr and temperature of 0˚C), the volume occupied by one kmol of ideal gas is 22.4 N·m^3/kmol. The stoichiometric volume of oxygen required was calculated using Equation (3) [15];

$$V_{O_2}^0 = \frac{22.41}{100}\left(\frac{C^i}{12} + \frac{H^i}{4} + \frac{S^i + O^i}{32}\right)\left[\frac{\mathrm{m_N^3\,O_2}}{\mathrm{kg\ fuel}}\right] \tag{3}$$

The oxygen flow rate;

$$V_{O_2}^0 = B \cdot V_{O_2}^0 \left[\frac{\mathrm{m_N^3\,O_2}}{\mathrm{h}}\right] \tag{4}$$

where B is the fuel flow rate, [kg fuel/h]

2.4. Stoichiometric Volume of Dry Air

Cookstoves use atmospheric air and not oxygen; with a volume of 21% in air v/v. The stoichiometric volume, V_a^0 of dry air required to burn 1 kg of fuel [14]:

$$V_a^0 = \frac{V_{\mathrm{O}_2}^0}{0.21}\left[\frac{\mathrm{m_N^3\ air}}{\mathrm{kgfuel}}\right] \tag{5}$$

The air flow rate;

$$V_a^0 = B \cdot V_a^0\left[\frac{\mathrm{m_N^3\ air}}{\mathrm{h}}\right] \tag{6}$$

where;

V_a^0 is the volume of air

$V_{\mathrm{O}_2}^0$ is the volume of oxygen

$\mathrm{m_N^3}$ is normal air volume in m^3

2.5. Moist Air Stoichiometric Volume

If the air introduced into the combustor is moist, the stoichiometric air volume, V_{ma}^0 is higher, $V_{ma}^0 > V_a^0$, due to water vapor. To compensate for the moisture the absolute humidity, *x*, is determined using, air temperature, t_a [˚C] and its relative humidity, φ_a [%] [14].

The volume of water vapor in moist air is:

$$V_{\mathrm{H_2O}}^0 = \frac{x\rho_a V_a^0}{\rho_{\mathrm{H_2O}}}\left[\frac{\mathrm{m_N^3\ H_2O}}{\mathrm{kg\ fuel}}\right] \tag{7}$$

where;

ρ_a is the density of air, normal state = 1.2925 kg/Nm^3

$\rho_{\mathrm{H_2O}}$, density of water vapor, normal state = 0.804 kg/Nm^3

t_a is ambient air temperature

φ_a is relative humidity

The stoichiometric moist air volume (V_{ma}^0) is calculated using equation [15];

$$V_{ma}^0 = V_a^0 + \frac{x\rho_a V_a^0}{\rho_{\mathrm{H_2O}}} = \left(1 + \frac{x\rho_a}{\rho_{\mathrm{H_2O}}}\right)V_a^0 = \left(1 + 1.61x\right)V_a^0\left[\frac{\mathrm{m_N^3\ air}}{\mathrm{kg\ fuel}}\right] \tag{8}$$

where is

$$x = \frac{0.622\varphi_a\rho_s}{\rho_b - \varphi_a\rho_s}\left[\frac{\mathrm{kg\ H_2O}}{\mathrm{kg\ dry\ air}}\right] \tag{9}$$

The value of *x* is assumed to be 10 g $\mathrm{H_2O}$/kg which corresponds to: t_a = 25˚C and φ_a = 50%. The value of *x* can also be determined from the Molliere diagram in **Figure 2** for moist air and the abscissa corresponding to intersection of the isotherm with Isohigra, ϕ_a, gives precisely the value of absolute humidity, *x* [g·$\mathrm{H_2O}$/kg dry air] [14].

2.6. Combustion Processes with Excess Air

Cookstoves and other combustion devices imperfection makes it difficult to

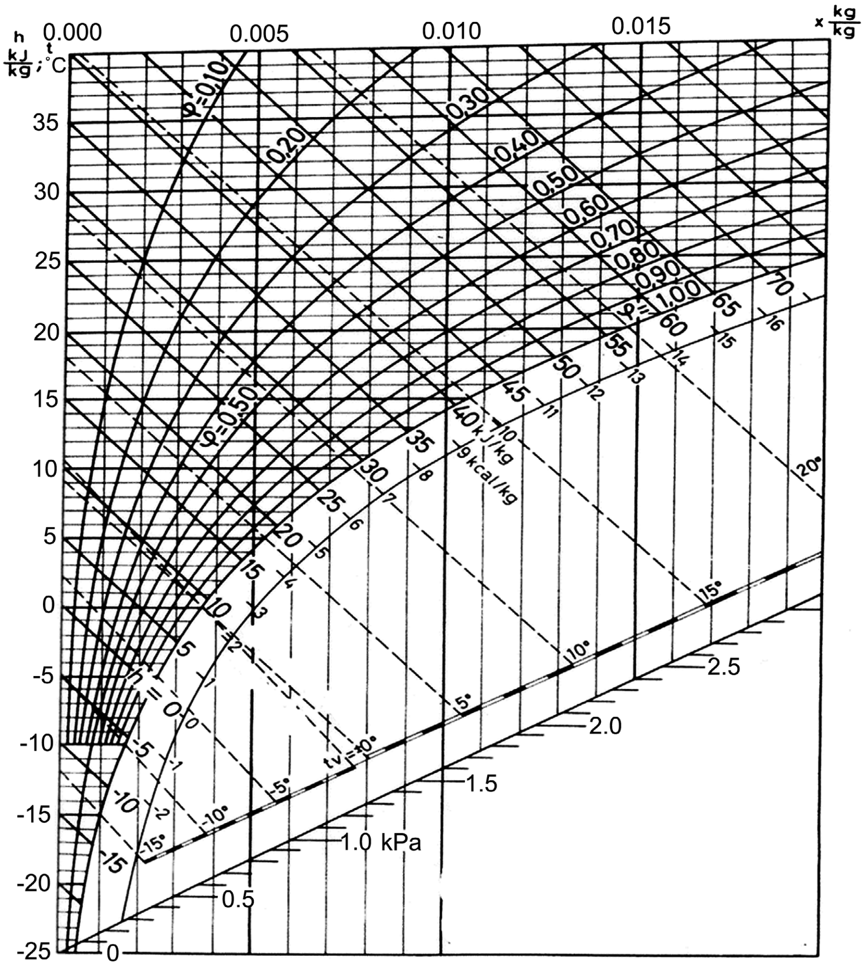

Figure 2. Molliere diagram [12].

attain fuel and oxidant homogeneous mixtures [15]. Thus, in combustion processes, more air is used than the stoichiometric value to favor complete combustion. The ratio between the actual air volumes, the excess air coefficient, λ, injected in combustion chambers, V_a and the stoichiometric air volume, V_a^0 is calculated using Equation (10) [14];

$$\lambda = \frac{V_a}{V_a^0} \tag{10}$$

λ depends on a number of factors such as; fuel properties, combustion mode and cookstove design. For combustion furnace the excess air coefficient varies between 1.05 to 1.7 [14]. The volume of combustion air with excess air is calculated using Equation (11),

$$V_a = \lambda_f V_a^0 \tag{11}$$

2.7. Experimental Set up

The experimental set-up is as illustrated in the **Figure 3**. The ceramic cookstove, a widely used fuel-efficient stove designed for use with charcoal was used in the

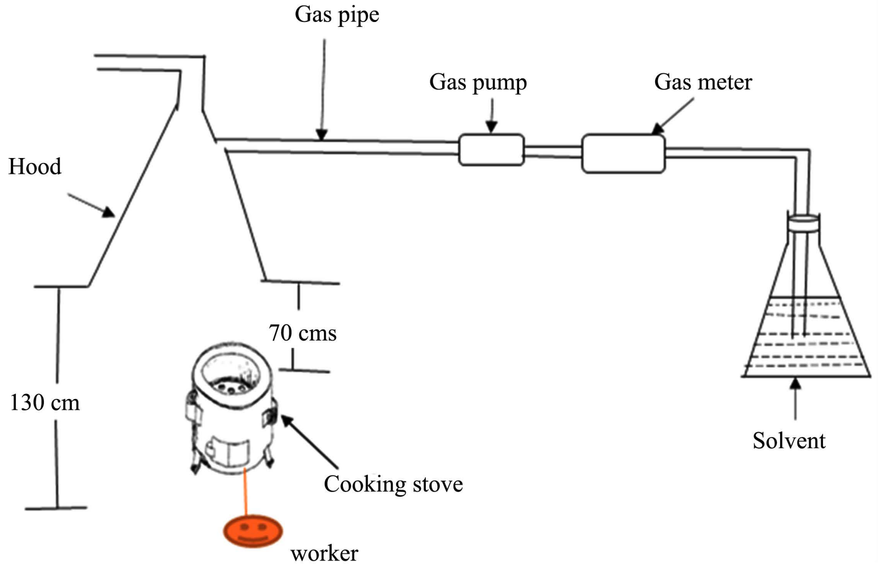

Figure 3. Experimental set up.

study. Almost 500,000 units have been sold across Kenya [1]. It consists of a metal casing with a 60 mm thick clay liner for heat conservation, and a grate with twenty-five 20 mm perforations and a door for air inflow control (**Figure 3**). This was loaded with one kg charcoal and ignited normally. The stove was allowed to light in the open outdoors for 10 minutes. The stove was then set up below the hood after the pre-heating phase. The measurements commenced at the start of the distillation phase and went on through the distillation phase for 30 minutes.

The experiments were repeated using the same analytical procedure, with secondary air injection. The secondary air injection was done using a pre-calibrated air pump with a delivery pipe leading to the combustion area. Emission data was collected using a calibrated Testo model 350 emission analyzer. Emission data was saved into the computer memory and experiments repeated in triplicates.

2.8. Emission Measurements

Gaseous emissions were measured with a calibrated Testo emission analyzer model 350. The analyzer was warmed-up of 10 minutes to produce reliable, stable readings. The analyzer was coupled with CO, C_xH_x, NO_x, NO and NO_2 gas sensors and a probe. The measurements were done as described in the instrument manual using a computer operated operation software. Data was saved in computer memory and later retrieved after analysis.

3. Results and Discussion

3.1. Feed Stock Characterization

Charcoal samples were bought from Kakuzi PLC, a company that makes charcoal for sale. Charcoal from tree species *E. glandis* which is commonly used in the study area was used in this study. Three batches of charcoal were sampled during the wet and the dry season. Composite samples were prepared for analysis. Analysis data is presented in **Table 1**.

The charcoal had a good HHV and LHV values making it appropriate for experimentation. The ash content was high at 20.7% ± 0.8% on a wet weight basis.

3.2. Proximate Analysis

The data for chemical/elemental composition, ash content and volatile matter is presented in **Table 2**. The elemental compositions of charcoal give an empirical formular of $C_{18}H_2O_1$, this shows that the charcoal from the present study had a high carbon content than that reported in [9] of $C_{6.5}H_2O$. This can be attributed to tree species and extent of pyrolysis. However, the carbon contents agree with the findings reported in selected trees species which ranges from 49.54% in *A. sieberiana* to 50.98% obtained in *A. leiocarpus* [15].

3.3. Stoichiometric Air Equivalent/Theoretical Air

The stoichiometric air equivalent was determined using method described in [15]. The experimentally determined chemical formular of charcoal ($C_{18}H_2O$) was used to calculate the stoichiometric air equivalent. The parameters are presented in **Table 3**.

A fuel flow rate of 1 kg fuel/hr was used in the experiments and an excess air coefficient of 1.05 to ensure that complete combustion is attained. An air volume of 5.2 $\text{m}_\text{N}^3\ \text{air}/\text{kg fuel}$ and an air flow rate of 5.55 $\text{m}_\text{N}^3\ \text{air}/\text{hr}$ was used in the experiments. The experimental conditions gave a clean smoke free burn.

Table 1. Charcoal chemical characteristics (n = 3).

	Present study	**[9]**
C (% w/w)	58.72 ± 3.3	76.62
Volatile Matter (% w/w)	15.95 ± 1.2	16
Moisture (% w/w)	4.69 ± 0.55	3.1
Ash (% w/w)	20.7 ± 0.8	13.4
HHV (MJ/kg)	30.5 ± 1.1	
LHV (MJ/kg)	29.3 ± 1.3	

Table 2. Proximate analysis data (n = 3).

	Present study	**Present study**	**[15]**	**[9]**
Element	Range (%)	Mean ± Std dev	Range (%)	Mean
Carbon	55.3 - 58.7	55.34 ± 3.3	49.54 - 50.98	76.72
Oxygen	4.14 - 4.88	4.1 ± 0.25	-	-
Hydrogen	0.54 - 0.58	0.52 ± 0.02	-	1.73

Table 3. Stoichiometric air parameters.

Parameter	Mean	Units
$V_{O_2}^0$	1.09 ± 0.1	$m_N^3\,O_2/h$
B	1.0 ± 0.01	kg fuel/hr
O_2 flow rate	1.09 ± 0.3	$m_N^3\,O_2/kg$ fuel
V_a^0	5.2 ± 0.5	m_N^3 air$/$h
Air flow rate	5.2 ± 0.5	m_N^3 air$/$h
t_a	22.45 ± 1.2	˚C
φ_a	64.5 ± 2.2	%
x	11 ± 0.9	kgH_2O/kg dry air
V_{am}^0	5.28 ± 0.6	m_N^3 air$/$h
λ_f	1.05	Unitless
V_a^0	5.55 ± 0.5	m_N^3 air$/$h

3.4. Emission Profiles

The combustion of solid fuels such as wood and charcoal can be summarized [16] by the reactions Equations (12)-(16);

$$\text{Fuels} \rightarrow \text{volatiles} + \text{tar} + \text{char} \tag{12}$$

$$\text{Volatiles} + O_2 \rightarrow CO + H_2O + \text{smoke} \tag{13}$$

$$\text{Char} + 0.5O_2 \rightarrow CO \tag{14}$$

$$CO + 0.5O_2 \rightarrow CO_2 \tag{15}$$

$$\text{Volatiles and tar} \rightarrow \text{smoke} \tag{16}$$

In charcoal, the devolatilization products include C_xH_x, CO, NO_x, SO_x, particulates, PAHs among other. Their release and quantities depend on the combustion conditions and fuel-oxidant ratios as well as cookstove device design and operation protocols.

3.4.1. CxHx Emission Profiles

Figure 4 presents the emission profiles for C_xH_x with and without secondary injection. The profile show that the hydrocarbon emissions exponentially from the start of the process to reach a plateau at around 2000 seconds of runtime. The study observed that the spread was across the three phases of combustion with low emission at preheat phase, highest at the distillation phase and plateau reached at the solid phase.

Secondary air injection of 5.2 m_N^3 air$/$kg fuel and a flow rate of 5.55 m_N^3 air$/$hr reveal a significant reduction in hydrocarbons by close to 80% for all experiments conducted. This can be attributed to the complete combustion of the

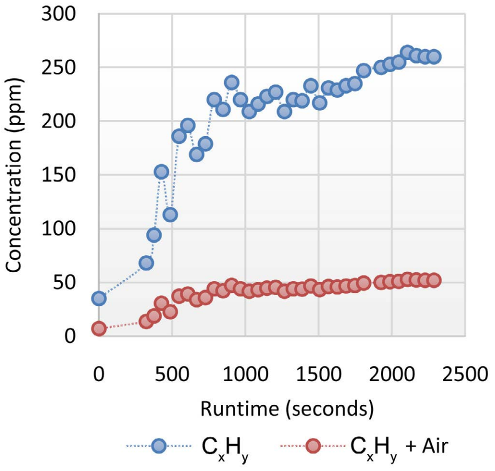

Figure 4. Emission profile for C_xH_x.

hydrocarbons and conversion thereof to carbon dioxide and water vapor. This study shows that emission of the hydrocarbons can be mitigated using low-cost air injection at the stoichiometric requirement.

3.4.2. CO Emission Profiles

The emission profiles of CO are presented in **Figure 5**. The profile shows a marked variation of the CO levels at all stages of combustion. The data for CO emissions obtained are comparable with previous experimental data [10]. The concentration of CO has been widely used as an indicator of the level of poor mixing in combustion systems.

The highest CO concentrations were reached in the final phase. This can be attributed to the formation of an ash layer on the surface of the smoldering charcoal which limits the interaction with atmospheric oxygen. [9] reports a concentration of 168 ppm after the 45 minutes, though the present study didn't capture the 45th minutes the trends agree with this finding due to the rapid decline in emissions after the 35th minute.

Injection of secondary air shows a very significant reduction of CO concentrations by >70%. This can be attributed to supply of enough oxygen and increased turbulence and mixing of fuel and oxidant. Air injection also served to break the ash layer that form on the glowing charcoal surface ensuring increased oxidation carbonaceous matter to CO_2.

3.4.3. NO_x Emission Profiles

NO_2 is a NO_x produced as a result of the oxidation of Nitrogen in air which is approximately 78%. Emissions of NO_x from charcoal depend on the fuel–N content of the fuel. However, emission factors on a thermal basis for NO_x using wood is slightly higher than for charcoal [9]. The generation of the NO_x during combustion is not desirable due to their environmental impacts. **Figure 6** depicts the emissions profile of NO_2.

Trends show that the release of NO_2 increases to reach a peak of 0.8 ppm at the 500th, this then falls gradually to stabilize at later rises to stabilize at 0.24 ppm at the distillation phase of the combustion (**Figure 6**). NO is another member of the NO_x family also formed due to the presence of nitrogen in air. The emission profile of NO is presented in **Figure 7**. The profile follows similar profile.

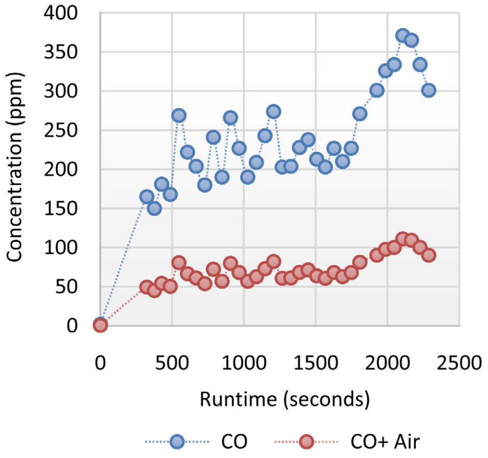

Figure 5. Emission profile for CO.

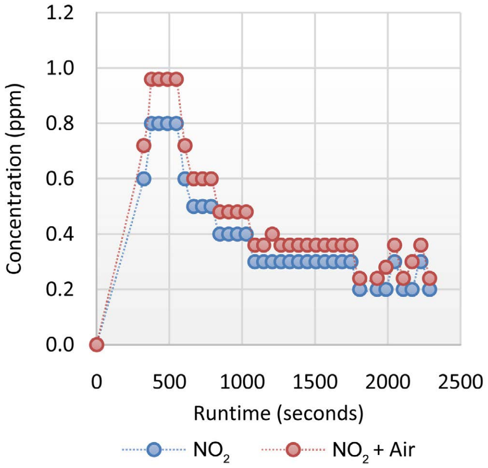

Figure 6. Emission profile for NO_2.

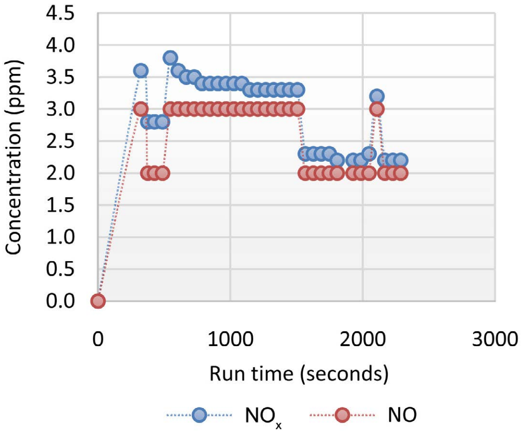

Figure 7. Emission profile for NO.

NO increases to reach a peak at 3.0 ppm to reach a plateau at and later drop to 2.0 ppm in the last phase of combustion, this could be as a result of temperature variations in the combustion chamber.

The data for NO_x emissions obtained are comparable with previous experimental data [16]. [9] report trace concentrations of NO after the 45th minute.

4. Conclusion

The study makes significant contribution to combustion studies especially control of pollutant emission. It's the conclusion of this study that the secondary air injection can improve the combustibility of fuel. This is also a climate plus approach as it removes precursors of serious atmospheric pollutants in the atmosphere. However more studies are required to investigate how air injection affects the efficiency of heat recovery systems.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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