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Effect of Excess Air Ratio and Temperature on NO_x Emission from Grate Combustion of Biomass in the Staged Air Combustion Scenario

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ABSTRACT: The combustion of biomass, in this case demolition wood, has been investigated in a grate combustion multifuel reactor. In this work a temperature range of 850–1000 °C is applied both for staged air combustion and nonstaged combustion of biomass to investigate the effects of these parameters on the emission levels of NO_x, N₂O, CO, hydrocarbons (C_xH_y) and different other components. The composition of the flue gas is measured by four advanced continuous gas analyzers including gas chromatograph (GC), two Fourier transform infrared (FTIR) analyzers, and a conventional multispecies gas analyzer with fast response time. The experiments show the effects of staged air combustion, compared to nonstaged combustion, on the emission levels clearly. A NO_x reduction of up to 85% is reached with staged air combustion. An optimum primary excess air ratio of 0.8–0.95 is found as a minimizing parameter for the NO_x emissions for staged air combustion. Air staging has, however, a negative effect on N₂O emissions. Even though the trends show a very small reduction in the NO_x level as temperature increases in nonstaged combustion, the effect of temperature is not significant for NO_x and C_xH_y, neither in staged air combustion or nonstaged combustion, while it has a great influence on the N₂O and CO emissions, with decreasing levels with increasing temperature.

1. INTRODUCTION

Because of environmental problems and a significant decrease in the reserves of fossil fuels, the interest in using renewable energy carriers has become more and more widespread in recent decades. Figure 1 shows the share of different energy resources used by end users in the world in 2008.¹ Biomass, which is almost a CO₂-neutral carbon-based renewable energy source, is cheaper to utilize compared to the other sources of renewable energy, especially in the case of cofiring and combined heat and power (CHP) applications.² Thermal conversion of biomass, including combustion, gasification, and pyrolysis, is the most common method of extracting energy from biomass. Among the three mentioned technologies, combustion, which has been used for a long time as a source of heat for residential purposes, remains the leading technology also in recent years.

Life cycle analyses show that the most important environmental issue regarding emissions from wood combustion is NO_x, contributing to almost 40% of the total emissions, including NO_x, PM₁₀, CO₂, SO_x, NH₃, CH₄, nonmethane volatile organic compounds (NMVOC), residues, and others.³ The basis for this comparison is in relation to expected environmental impact points (EIP) of the specific emission according to the ecological scarcity method, applied for heating with wood chips. However, it is evident that the given data from any LCA analysis depends strongly on the valuation of the greenhouse gas effect since the ranking changes significantly as a result of the different CO₂ impacts of the fuels.⁴ In this respect, much effort has been done to characterize biomass and reduce NO_x emissions from the combustion of biomass and other solid fuels.^{5,6} Two major approaches exist for NO_x reduction, primary measures and secondary measures. The main difference is that primary measures prevent formation of NO_x emissions,

while secondary measures clean these emissions after their formation. Modifications to fuel properties and the combustion chamber and improvements to combustion technologies are examples of primary measures. One of the most important and widely used measures to reduce NO_x in solid fuels combustion is staged combustion. Staged combustion, including staged air combustion and staged fuel combustion, can give a NO_x reduction in the range of 50–80%.^{4,7–10} The purpose of staged air combustion is to add primary air at a less than stoichiometric ratio, in order to devolatilize the volatile fraction of the fuel, resulting in a fuel gas consisting mainly of CO, H₂, C_xH_y, H₂O, CO₂, and N₂ and also small amounts of NH₃, HCN, and NO_x from the fuel nitrogen content. If sufficient oxygen exists in the first stage, the fuel nitrogen intermediates will be converted to NO_x (mainly NO), but shortage of oxygen will cause NO to act as an oxidant for CO, CH₄, HCN, and NH_i (with $i = 0, 1, 2, 3$) in the reduction zone, hence to reduce the nitrogen in NO and NH_i to molecular nitrogen, i.e., N₂, in reactions such as NO + NH₂ = N₂ + H₂O or NO + CO = CO₂ + 0.5N₂.^{4,11–13} NH₃ and HCN form in the pyrolysis stage of combustion depending on the temperature and fuel type, where ammonia is believed to be the most important N-species in the combustion of biomass, while HCN is more important for high-rank coals.^{14–16} Ammonia that is formed in this stage is converted to radicals of NH_i. At high temperatures and fuel lean combustion, these radicals will be converted to NO, while at fuel rich conditions, N₂ will be the main product.¹⁷ Thereafter sufficient air is added in the second

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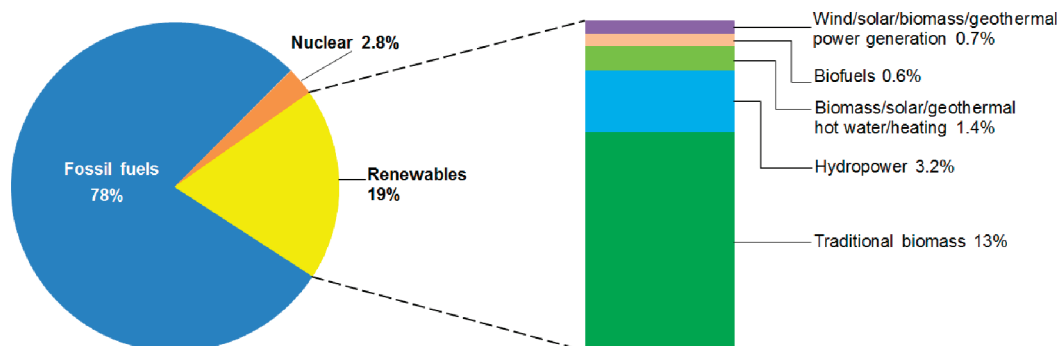


Figure 1. Renewable energy share of global final energy consumption, 2008.¹

stage to ensure a good burnout and low emission levels from incomplete combustion.

The effect of excess air ratio, residence time, and temperature has previously been studied for a single wood particle in batch combustion, showing dependency of the NO level with excess air ratio and temperature simultaneously.¹⁸ Also, previous work showed the effect of fuel type and cocombustion of biomass with natural gas on NO emissions when using two-stage combustion.¹⁹ Fuel-N content and the conversion rate of NOx precursors (NH₃ and HCN) to NOx have been experimentally and numerically investigated for different solid biomass.^{20–23}

Combined staged (CS) technology¹⁰ is a method of NOx reduction including reburning, staged air combustion (AS), staged fuel combustion (FS), and selective noncatalytic reduction (SNCR).^{24–26} Combined staged technology starts to reduce NOx emissions where the other methods are not sufficient. Studies showed that CS is more effective for NOx reduction, especially in the temperature range of 1000–1400 °C.¹⁰ For temperatures below 850 °C, higher N₂O, lower NOx, and slightly higher CO emissions are reported for cocombustion of coal and woody biomass.²⁷ Another technique of primary measures for NOx reduction in biomass combustion is flue gas recirculation (FGR) which basically increases the total mass flux of the gases and decreases the temperature and the partial pressure of oxygen in the mixture.²⁸ It is proposed that in fixed-bed combustion of straw, the effect of FGR on NO reduction could be considerable.²⁹

The present study aims to investigate the effect of temperature on the emission level, especially NOx, for a selected type of biomass, demolition wood (DW). Combustion of DW as a type of wood residue represents a large source of alternative energy, in addition to the fact that a waste disposal problem will also be solved. DW originates from many sources; therefore, it contains normally different undesirable components such as metals, paints, plastics, etc., which will raise the environmental impact of DW. Especially, the high nitrogen content, compared to wood, is the focus in this study. A series of experiments have been carried out for conventional combustion (nonstaged) and staged air combustion to study the effect of temperature variations, combustion technology, and excess air ratio. The work is a part of a wider study which has been carried out in the same reactor, investigating different operating parameters and fuels.³⁰

In the next section a brief explanation of the experimental setup and test procedures is presented and the sampling devices and methodologies are described. Thereafter, the results are presented and discussed with respect to different variables, and finally conclusions are given.

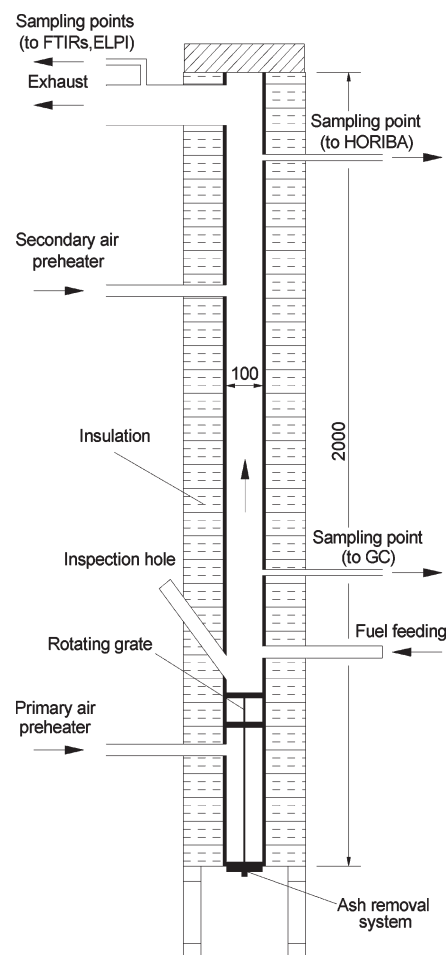


Figure 2. Schematic drawing of the reactor (the sizes are given in millimeters).

2. EXPERIMENTAL SECTION

2.1. Reactor Specifications. The reactor is shown in Figure 2. The reactor is installed vertically and has a total height of 2 m. The inner diameter is 100 mm. The reactor is made of ceramic material (alumina based) and has two reactor tube sections, each 1 m high. A rotating grate, with two grate levels, is placed 0.4 m from the bottom of the reactor, which means that the combustion zone is 1.6 m high. The two grate levels are 10 cm apart and there are rotating blades on each level that moves the unburnt fuel particles on the grates and from the upper grate to the second grate and from the second grate to the ash bin through a

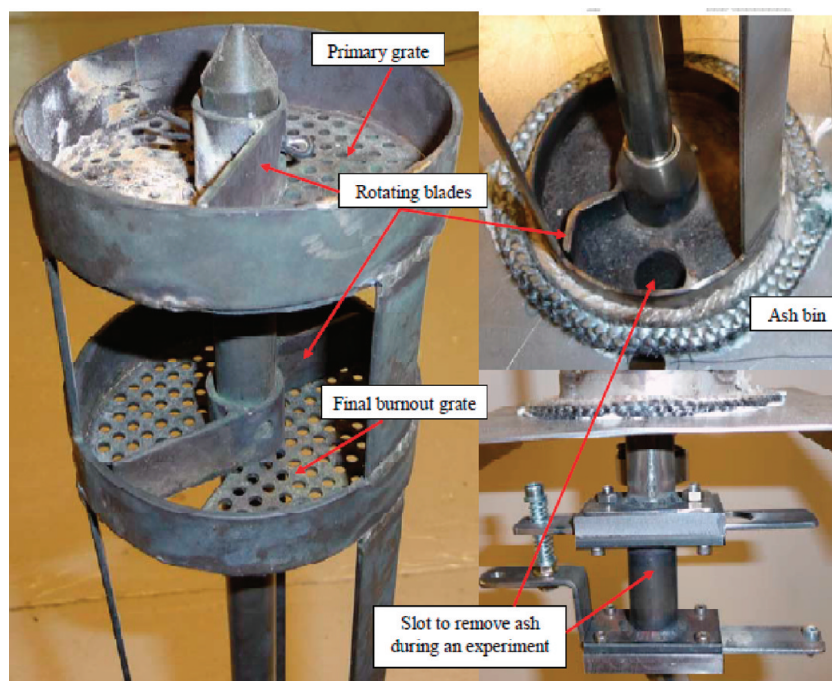


Figure 3. Design of two grates and ash bin.

slot in the grates. The rotational speed of the blades is about 3 min per revolution, which means that the residence time of each pellet on the grates is up to about 6 min, totally. This ensures complete burnout of the particles/char at the lower grate and that the ashes can be collected in the bottom of the reactor, i.e., in the ash bin. The grates and the ash bin are shown in Figure 3. A view glass is located just above of the upper grate to be able to see the combustion zone from the outside. The residence time of the fuel in the reactor is high, and the gas in the reactor has very low flow velocity (0.04–0.07 m/s). The secondary combustion zone ensures a residence time of several seconds and is hence in fact comparable to conventional combustion systems. According to the given reactor dimensions and velocity, the residence time for the primary zone (90 cm) is 13–23 s and for the secondary zone (70 cm) 10–18 s, giving a total residence time of 23–41 s. The outer surface of the reactor is insulated, and the reactor heating system has an effect of 16 kW, composed of four identical electric heaters with a height of 0.5 m.

The reactor can be heated up to 1300 °C, and the fuel feeding rate can be up to 0.5 kg/h. A pneumatic-vibration based feeding system is installed to ensure automatic fuel feeding at a set rate. Pellets are fed by a water-cooled moving piston into the reactor and immediately fall down on the upper grate. Air can be fed to the reactor in three stages (below the lower grate, above the upper grate (two inlets), and at one level higher up (two inlets)), up to 120 NL/min, totally. Five preheaters are installed, one for each air inlet stream, to heat the feeding air to the reactor temperature before entering the reactor. The air flow rate is controlled by mass flow controllers via a PC.

2.2. Sampling and Measurements. As shown in Figure 4, gas is extracted in four places to carry out analysis and further measurements. The flue gas composition is measured by means of three gas analyzers. In case of staged air combustion, a gas chromatograph (GC) is used to measure the gases in the primary section. The GC is sampling between the first and the second combustion stage and is a Varian CP-4900 Micro-GC. Sampling for the GC is made through an 8 mm diameter stainless steel probe, and the gas is passed through an ice bath to ensure removal of water, particles, and tars, in addition to reducing the temperature to an acceptable level for the GC. The sampling condition

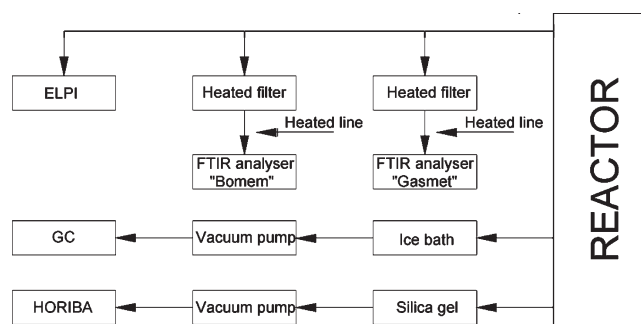


Figure 4. Schematic diagram of the sampling line.

is noncondensing gas of 0–40 °C and the maximum sample pressure is 200 kPa. The GC is equipped with two dual-channel micromachined thermal conductivity detectors (TCD), with a detection limit of 1 ppm for WCOT (wall-coated open-tubular) columns. The WCOT column is a column in which the liquid stationary phase is coated on the essentially unmodified smooth inner wall of the tube. The sample flow rate is 1 L/min and the sampling time interval is 2 min. Argon and helium are used in two columns, 10 and 20 m long, respectively. The first column measures CH_4 , CO_2 , $\text{C}_2\text{H}_2 + \text{C}_2\text{H}_4$, and C_2H_6 while the second column measures H_2 , O_2 , N_2 , CH_4 , and CO .

A Horiba multispecies gas analyzer PG-250 is sampling from the top of the reactor. It is capable of measuring five components, NO_x , SO_2 , CO , CO_2 , and O_2 , with the same methods used by a permanent continuous emissions monitoring system (CEMS). These include pneumatic nondispersive infrared (NDIR) for CO and SO_2 , pyrosensor NDIR for CO_2 , chemiluminescence (crossflow modulation) for NO_x , and a galvanic cell for O_2 measurements. A vacuum pump is used to extract the sample gas at a flow rate of 0.4 L/min from the top of the reactor and passes it through a silica gel box and a filter to remove moisture and particles. The response time (T_{90}) of the analyzer is less than 45 s for NO_x , CO , O_2 , and CO_2 and less than 240 s for SO_2 . The

Horiba analyzer is equipped with a drain separator unit (DS-200) and an electronic cooler unit.

Two Fourier transform infrared spectroscopy (FTIR) analyzers are also used to measure the gas composition in the exhaust gas. They do sampling at the same point, which allows for comparing different measurements for these species: H_2O , CO_2 , CO , NO , N_2O , NO_2 , SO_2 , NH_3 , HCl , HF , CH_4 , C_2H_6 , and C_3H_8 . The Gasmeter DX-4000 FTIR analyzer incorporates a spectrometer, a temperature controlled sample cell, and signal processing electronics. The sample cell is heated up to 180 °C which ensures that the sample stays in the gaseous phase even with high concentrations of condensable hydrocarbons. The measurement time is typically 60 s, and the spectrometer resolution is 4 cm^{-1} at a scan frequency of 10 scans/s. The sample cell has a multipass fixed path length of 5 m and a volume of 0.4 L. To measure O_2 , an optional oxygen sensor, based on a ZrO_2 cell, is attached to the analyzer. In total it can measure C_2H_4 , C_6H_{14} , CHOH , HCN , and O_2 in addition to the above-mentioned 13 gases.

A Bomem MB 9100 FTIR analyzer is also used to measure common mutual species and C_4H_{10} . The measurement time is typically 80 s. A large cell, operating at 176 °C and equipped with a 1 cm^{-1} spectral resolution detector, was handling the analyses. Generally FTIR measurements of NO may introduce significant uncertainties. However, the NO_x emissions reported in the results part are based on the Horiba analyzer, which is using the chemiluminescence technique. The N_2O measurements were made by the FTIR analyzers, and the reported values were measured by the Gasmeter FTIR. The Bomem FTIR showed N_2O values close to the values measured by the Gasmeter FTIR.

The temperatures at different levels in the reactor are measured by means of thermocouples and are monitored continuously to control and protect the furnace operation. These thermocouples are used to set and control the temperatures of the air preheaters and the reactor wall heaters during heat-up and during the experiment.

2.3. Experimental Procedures. Experiments are performed for nonstaged and staged air combustion. Four different temperatures are selected, 850, 900, 950, and 1000 °C. During each experiment, the temperature is kept constant for the reactor, primary air, and secondary air at the mentioned values. All the sampling devices are calibrated each day, before starting the experiments. Also the Gasmeter FTIR calibration was performed daily by means of background spectra.

Pellets with a diameter of 6 mm and a length of 10–15 mm are fed by the automatic feeding system. The fuel feeding rate is set to 400 g/h. To have a precise composition of the fuel, during each run, three different samples at three different times are taken from the fuel feeding system to analyze the moisture content.

The total excess air ratio for nonstaged combustion is set to 1.6; however, the variation in the fuel feeding rate allows capturing a total excess air ratio range of 1.2–3, making it possible to see emission trends as a function of total excess air ratio. Each experiment has been carried out for at least 2 h at stable operating conditions.

For the staged air combustion experiments, the total excess air ratio is also set to 1.6, while the primary excess air ratio is set to 0.8, which means that 50% of the total air is fed at each stage. The air flow has been held constant during the experiments. However the variations in the fuel feeding rate cause natural variations in primary and secondary excess air ratios, since the fuel was fed as pellets and, depending on the length of the pellets and the position of the pellets on the grate after feeding, natural variations in the excess air ratio occurred. Therefore the mentioned situation makes it possible to experimentally derive the effect of variations in the primary excess air ratio on the NO_x reduction potential by staged air combustion. Data treatment has been carried out in a cautious manner to avoid nonreliable data. Significant transient effects are effectively eliminated by a filtering procedure while treating the experimental data. This filtering procedure requires that the change in the excess air ratio per second is less than 0.01; otherwise, the

Table 1. Proximate Analysis of DW Pellets (wt %)

pellets	ash (dry basis)	volatile (dry basis)	fixed carbon (dry basis)	moisture (wet basis)
demolition wood (DW)	2.49	75.97	21.54	9.68–14.97

Table 2. Ultimate Analysis of DW Pellets (wt % Dry Ash Free Basis)

pellets	C	H	O	N	S	Cl
demolition wood (DW)	48.45	6.37	44.11	1.06	0.02	0.05

complete measured data set at the current time is omitted in the final results.

2.4. Fuel Characteristics. The biomass that has been used for the present work is classified as demolition wood (DW). The received biomass is pelletized with a pellet machine to get a similar shape and composition for all the experimental runs. To ensure a homogeneous distribution, a large volume of demolition wood was shredded to sawdust size, followed by thorough mixing. Pellets were then made from these fine pieces. Hence, variation in the N-content is not regarded as a challenge in these experiments. The type of biomass, the origin of it, and the pretreatment technology applied to it are important parameters influencing the ultimate and proximate analysis of the biomass. For example, drying can reduce the moisture content from 65% in virgin wood down to below 10% in wood pellets. The proximate analysis of the present fuel is shown in Table 1, where the moisture content is measured during each experimental run. Moisture, VM, and ash content are measured using ASTM E871 (50 g, 103 ± 2 °C, 24 h), ASTM E872 (1 g, 950 °C, 7 min), and ASTM D1102 (2 g, 580–600 °C, 4 h) standards, respectively, and the fixed carbon is calculated by the difference to 100%. Three samples are analyzed from different parts of the pellets to get repeatable analyses, showing that the fuel was homogeneous. Volatile matters for wood chips, bark, and straw are normally in the range of 76–86, 70–77, and 70–81 wt %, respectively,³¹ while for the DW it was 75.97%, as shown in Table 1.

Table 2 shows the DW ultimate analysis (dry ash free). All the samples have been dried in a vacuum exsiccator over phosphorus pentoxide prior to analysis. The determination of C/H/N/S is performed using the “EA 1108 CHNS-O” by Carlo Erba Instruments elemental analyzer. The method is adjusted for sample amounts of 2–10 mg and performs with an uncertainty within 0.3 wt % as required for confirmation of assumed chemical composition. The operation range covers the content from 100 to 0.1 wt %; sulfur determination is in the concentration range of 1.0 down to 0.01 wt % and the uncertainty is estimated to be 0.02 wt % for C/N/S/Cl. The nitrogen content, which is the most important element in this study is 1.06%, while for wood, straw, peat, sewage sludge, and coal it is 0.03–1, 0.3–1.5, 0.5–2.5, 2.5–6.5, and 0.5–2.5 wt %, respectively.³²

3. RESULTS AND DISCUSSION

3.1. Accuracy of the Results and Combustion Quality. In order to establish the accuracy of the measured results, the total carbon balance is needed to quantify the measured values compared to expected analytical values. The carbon balance for one of the experiments, at 1000 °C for staged air combustion, is shown in Figure 5. This figure shows the results from the start time until the end of the experiment. However, it should be noted that the first and very last section of the data has not been considered in the final data for further treatment, since these parts are typical transient periods, outside of the stable run

period. The calculated values for CO_2 , based on the carbon content in the fuel and the measured volume percentage of oxygen and CO in the flue gas, is very close to the measured values for CO_2 for each sampling interval and hence the deviation is sufficiently close to zero. In the case of a large deviation between measured and calculated values, the data set is removed from further treatment, since it could be from an unstable or transient period. Therefore the carbon balance for the presented

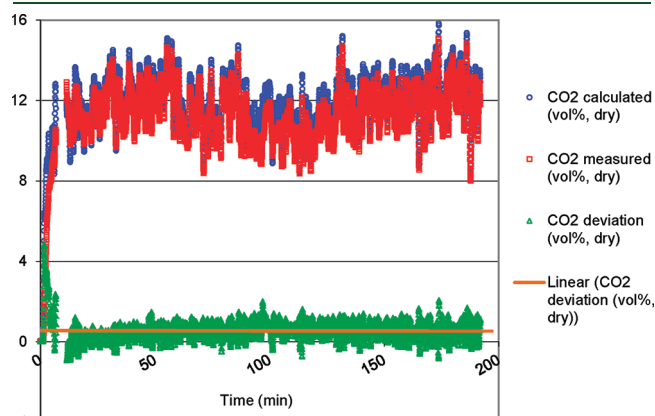


Figure 5. CO_2 deviation based on calculated and measured values from the Horiba gas analyzer for one set of the experiments; 1000 °C, staged air combustion.

set of results is in good order. All the experiments at the different temperatures, with and without staging air, show the same good results for the CO_2 deviation. Emission level of C_xH_y and CO for both staged and nonstaged combustion is shown in Figure 6. For C_xH_y , both staged air combustion and nonstaged combustion show the same emission level, typically below 4 ppm at 11% O_2 in dry flue gas. Here low values for C_xH_y and CO emissions, respectively, below 5 and 50 ppm shows good mixing conditions and residence times in line with modern industrial scale boilers.³³ Emissions of gaseous hydrocarbon compounds and CO are a result of incomplete combustion. Favorable combustion leads to small emissions of hydrocarbons because the organic material burns out. Large emissions of hydrocarbons indicate unsatisfactory combustion conditions and probably soot emissions. Large boilers are generally operated at an appropriate oxygen concentration, temperature, and residence time and consequently are under favorable combustion conditions. Particles originating from incomplete combustion are few or none and can be decreased in modern boiler types by 180 times,^{21,34,35} and in the advanced wood furnaces, CO emissions of 10–20 mg/m^3 can be reached, depending on the temperature and selection of the optimum excess air ratio.⁴ At 900 and 950 °C, some values up to 10 ppm C_xH_y are visible for nonstaged combustion, which originate from the sets of data from the period where the actual stable experiment had not been started, which means that in the stable run, the C_xH_y for both 900 and 950 °C still is 1 to 3 ppm. The C_xH_y emission level is slowly increasing with excess air ratio

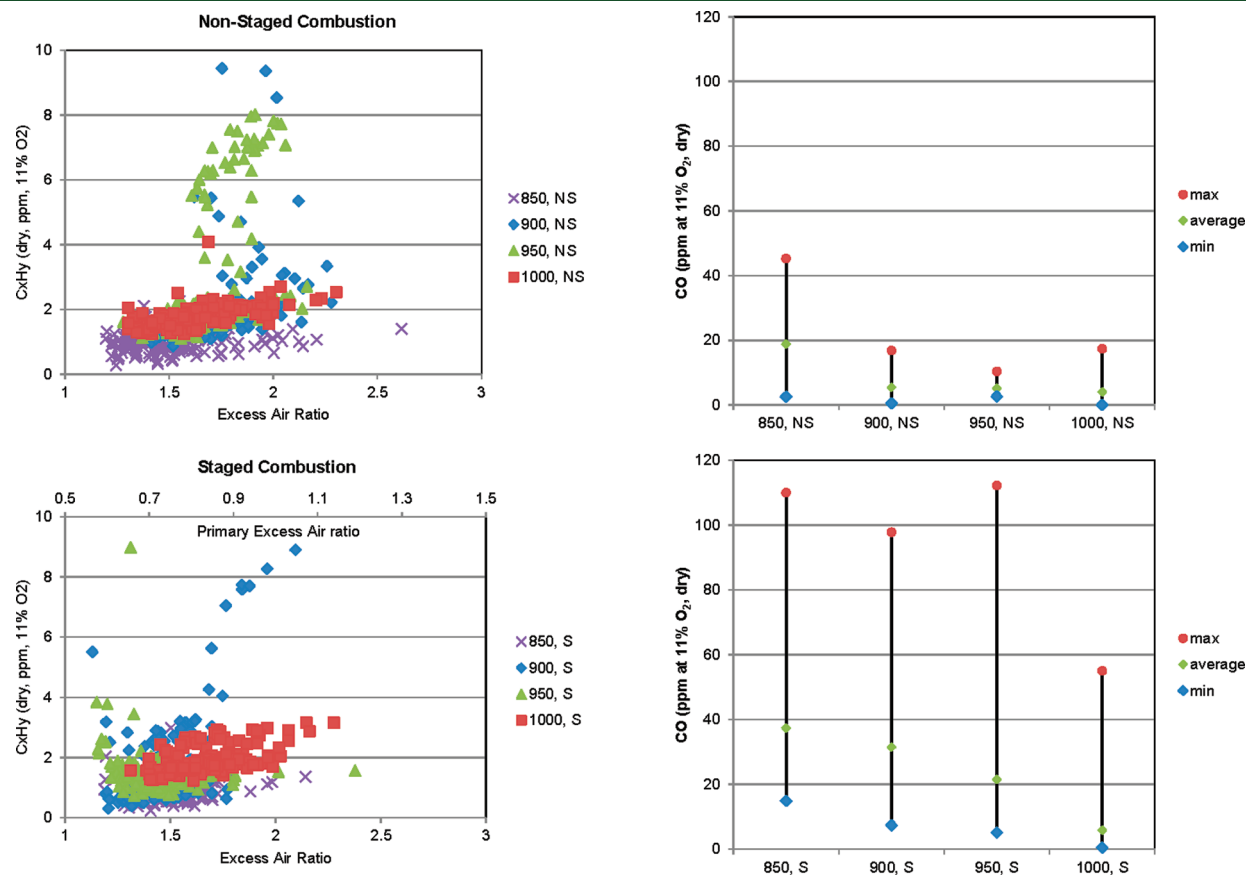


Figure 6. (Left) C_xH_y as ppm corrected to 11% O_2 in dry flue gas for staged air combustion and nonstaged combustion as a function of excess air ratio at the different temperature levels. (Right) Average, minimum, and maximum value of CO as ppm corrected to 11% O_2 in dry flue gas for staged air combustion (S) and nonstaged combustion (NS) at the different temperature levels.

for both staged air combustion and nonstaged combustion. The influence of excess air ratio is small, but the C_xH_y emission level is a direct function of the excess air ratio, and the minimum C_xH_y emission is taking place at the lowest total excess air ratio.

For nonstaged combustion, the temperature has an inverse effect on the C_xH_y emission levels. The lowest emissions occur at 850 °C, while at 900, 950, and 1000 °C the temperature has no effect. For staged air combustion, the effect of temperature is more visible than for nonstaged combustion. Here the effect of temperature is still low, but it is clearly possible to observe an increase in emission levels with increasing temperature in the lower graph. The observed increase of C_xH_y may be due to a higher release of gaseous hydrocarbons from the fuel at higher temperatures, where the pyrolysis process is intensified by temperature and the formation of hydrocarbons are increased.³⁶ Detailed experimental and thermodynamic studies on particulate emissions from this multifuel reactor to investigate particle size distribution and chemical composition of fly/bottom ash is extensively discussed recently.^{37,38} The effect of temperature on CO is the same as for N_2O . As shown in Figure 6, increasing temperature decreases the CO level, while staged air combustion has a negative effect on CO. The former effect is due to a more complete oxidation of CO, forming CO_2 . However, with air staging, the low stoichiometric ratio in the first stage cause the oxidation route to be less effective, so the emission of CO rises. The staged air combustion results indicate an increase of 99–482% for CO for staged air combustion, while the mean CO levels are about a factor of two increased.

Emissions of gaseous hydrocarbon compounds and CO are a result of incomplete combustion. Large emissions of CO indicate unsatisfactory combustion conditions. In the present case, higher excess air ratio (more oxygen available) will lead to an almost complete combustion (heated reactor, i.e., temperature not influenced by the excess air ratio). Consequently, the maximum CO level corresponds to the lowest excess air ratio or the more fuel rich condition. In the staged combustion experiments, because of the under-stoichiometric condition in the first stage, a large amount of unburnt species (including CO) will be formed in the primary stage. The burnout of the unburnt species should take place in the second stage. If necessary air is available in the burnout zone, the CO level will be very low; if not the CO level will be high. So the higher deviation for CO emission in the staged combustion is a result of the variations in secondary excess air ratio, where more fuel rich conditions in the secondary stage can, combined with not perfect mixing of fuel gas and secondary air, cause higher fluctuations in the CO level.

3.2. NO_x Emissions: Effect of Excess Air Ratio and Air Staging. Skreiberg et al.³⁹ showed that the fuel-N conversion to NO_x is strongly dependent on the excess air ratio and that it increases with increasing excess air. However, in this study we investigate further the effect of temperature on this dependency. Figure 7 shows the effect of temperature on the NO_x emission level (ppm at 11% O₂ in dry flue gas) for both staged air combustion and nonstaged combustion of DW at 850, 900, 950, and 1000 °C. NO₂ was typically below 1 ppm for the staged and nonstaged experiments which is within the measurement error ranges for the analyzers (Gasmeter and Bomem FTIRs) for NO₂, except for one case at 900 °C for staged-air combustion, where a NO₂ level of up to 3 ppm is measured. Consequently, we have not found any clear relation between NO₂ and temperature in the investigated temperature range. The upper graph for nonstaged combustion shows an increasing trend for NO_x when

the excess air ratio is increasing. This corresponds with the findings of Skreiberg et al.³⁹ The NO_x level for these cases, at all temperatures, varies from 75 to 200 ppm at 11% O₂ in dry flue gas. However, this graph also indicates that temperature variations have almost no effect on the NO_x emission level. All the data for the different temperatures are included, showing that they are in the same range for the same excess air ratio and temperature. This indicates that the temperatures are, as expected, too low for thermal NO_x formation. According to the literature, thermal NO_x formation in different biomass combustion systems starts at temperatures above 1400 °C.^{8,40}

The lower graph, which shows the same information for staged air combustion of DW, is distinctively different from that of nonstaged combustion. Here an optimum excess air ratio exists for each combustion temperature. The NO_x results are here plotted as a function of overall excess air ratio while the corresponding primary excess air ratio (half of the overall excess air ratio) is given on the upper *x*-axis for staged combustion. GC measurements were carried out to analyze the composition of the primary gas, but they can only provide an indicative value for the primary excess air ratio and not with a sufficient time resolution. The main point is to use total excess air ratio as an indicative value to show the effect of variations in the primary excess air ratio on the NO_x reduction degree, and this was effectively achieved with the experimental setup, the measurements carried out, and the data treatment procedures employed. The NO_x emission level is ranging from 25 to 120 ppm at 11% O₂ in dry flue gas. This range points out that the emission level by staged air combustion of DW decreases by a factor of 2–4, corresponding to 50–75% NO_x reduction compared to nonstaged combustion. In the optimum case, i.e., at an excess air ratio of 1.6–1.9, the NO_x reduction is 85%. This corresponds to an optimum primary excess air ratio of 0.8–0.95. Again, the effect of temperature is very low, as was the case for nonstaged combustion, and all temperatures show the same trend. First an increase occurs in the NO_x level with an increasing excess air ratio up to approximately 1.5, and from this point the NO_x level is decreasing until the excess air ratio approaches 2. A further increase in the excess air ratio will cause the NO_x level to start increasing again. This effect is due to the increase of available air both in the primary and secondary zone, which causes the first stage not to be in a fuel-rich condition, as was explained in the Introduction. This will increase the TFN/Fuel-N ratio, hence lowering the NO_x reduction potential, where total fixed nitrogen (TFN) is the total mass of nitrogen in the flue gas (all N containing gases coming out from the reactor except for N₂), and Fuel-N is the total mass of nitrogen in the fuel.⁴¹ The concentration of NH₃ and HCN was very low in the flue gas after a long residence time, typically below 1 ppm, which was within the measurement error range of the analyzers for these species.

Figure 8 shows the effect of temperature on N₂O emissions at different excess air ratios. The upper graph clearly shows that in the case of nonstaged combustion, the N₂O emission level is not a function of the excess air ratio. However, the effect of temperature on the N₂O emission level is significant in nonstaged combustion. The maximum N₂O emission level is seen at the lowest temperature, i.e., 850 °C. Increasing the temperature with just 50 °C will decrease N₂O from 4.8 to 0.9 ppm (mean value), which corresponds to 80% reduction. The same trend has been found for N₂O formation for temperatures above 750 °C in the literature, where reduction of NO has a significant effect on the production of N₂O.^{42,43} The production of N₂O is mainly

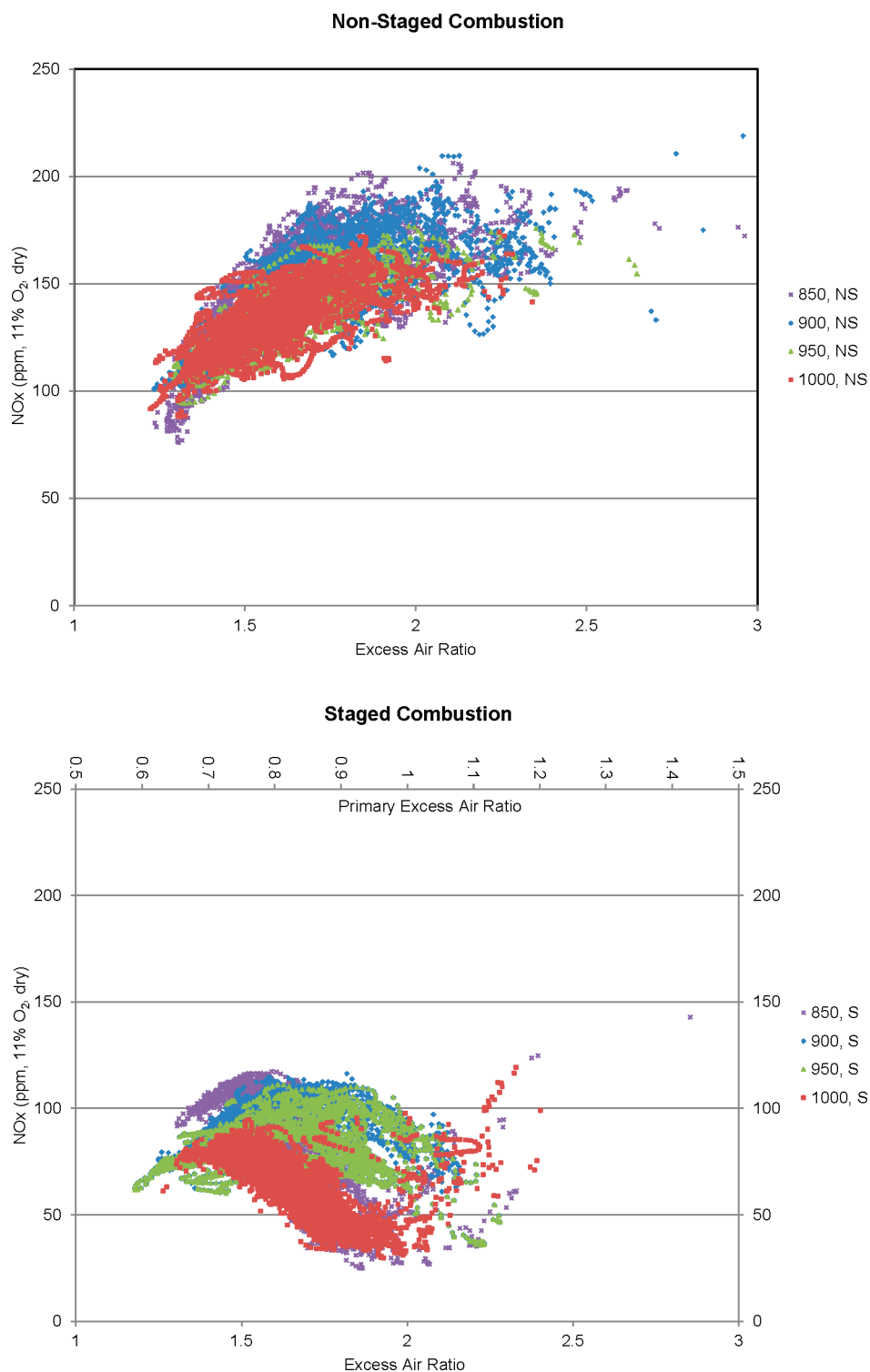


Figure 7. NOx as ppm corrected to 11% O₂ in the dry flue gas for staged air combustion and nonstaged combustion as a function of excess air ratio at the different temperature levels.

from the reaction of $\text{NO} + \text{NCO}$ and $\text{NO} + \text{NH}_3$, so N_2O is mainly reduced by thermal decomposition. HCN and NH_3 either can reduce to N_2O and N_2 or oxidize to NO . At the higher temperature, the oxidation route becomes more dominant; therefore, having a low amount of N_2O is predicted.^{11,42} The staged air combustion experiments show higher values of N_2O for the same conditions. At the optimum excess air ratio for NOx

reduction, the increase in N_2O emission compared to nonstaged combustion is almost 120%.

3.3. Temperature Influence on NOx and N₂O Emission Levels. As discussed in sections 3.2 and 3.3, the effect of temperature on the NOx emission level is small in the investigated temperature range of 850–1000 °C. However, with the extraction of the maximum and minimum levels for each

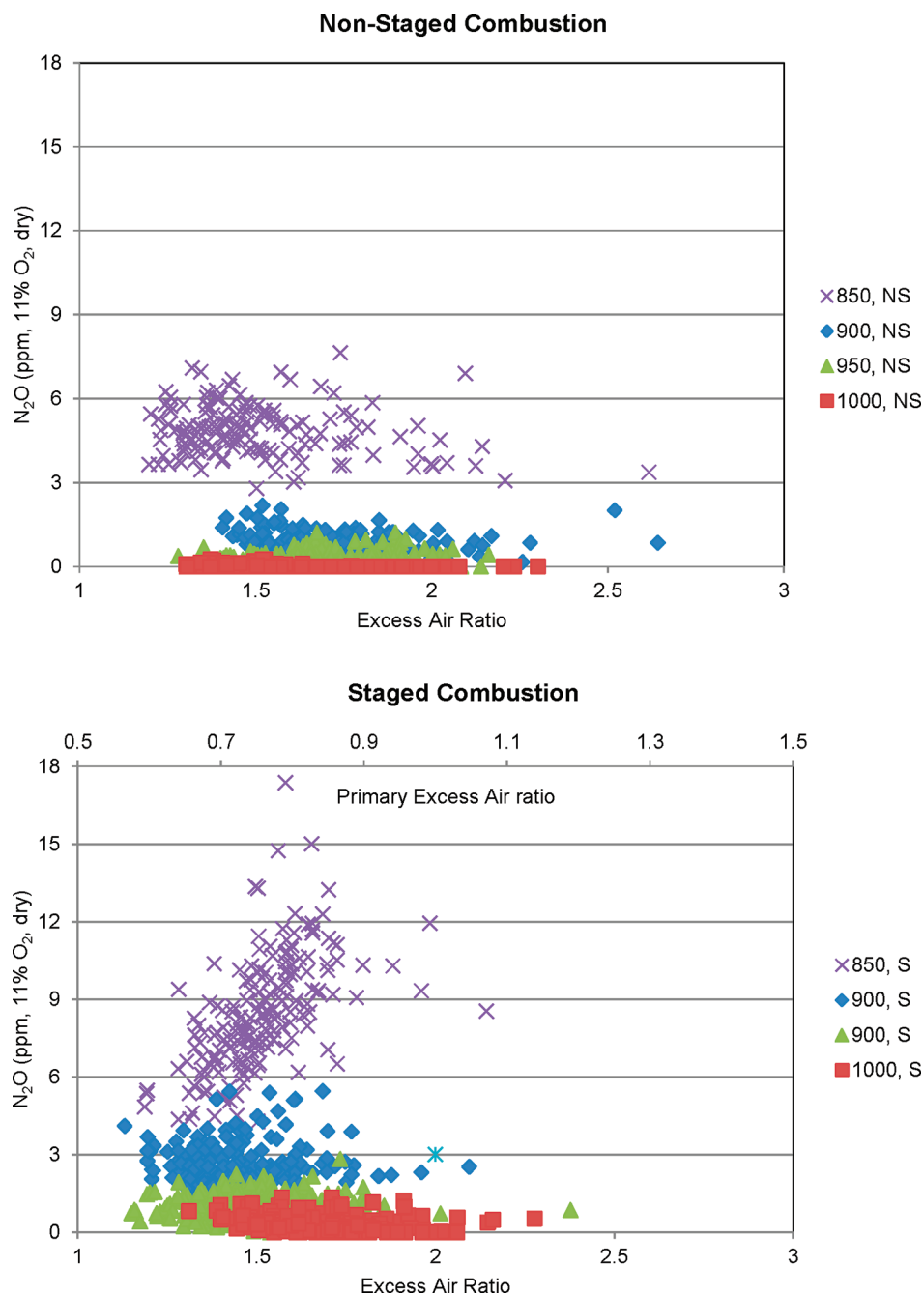


Figure 8. N_2O as ppm corrected to 11% O_2 in the flue gas for staged air combustion and nonstaged combustion as a function of excess air ratio at the different temperature levels.

temperature, some important trends can be established. Figure 9 shows mean, maximum, and minimum NO_x emission level (ppm at 11% O_2 in dry flue gas) for nonstaged (NS) and staged air (S) combustion of DW in the mentioned temperature range. The average (mean value for different excess air ratios over a period of time) is remaining constant at the different temperatures, at almost 150 ppm for nonstaged combustion. The maximum, mean, and minimum values are at the highest level for 900 °C in the nonstaged experiments. NO_x at staged air combustion seems not to be a function of temperature either. The average value is on the order of 80 ppm at 11% O_2 in dry flue gas and the highest mean value is seen at 900 and 950 °C. It is interesting to

note that in both cases the spread between the highest and lowest values are higher in the lower end of the temperature range. The lower amount of NO_x at 1000 °C needs more clarification. Considering the optimum primary excess air ratio as the key point for minimizing the NO_x emission, we can see that at 1000 °C most of the time the reactor was running in the optimum excess air ratio range (see Figure 7). This resulted in a lower average NO_x value (for a complete run). At 900 and 950 °C, in spite of our efforts to operate at optimum conditions, the primary excess air ratio has also been slightly lower than the optimum range (0.8–0.95) for the experiment. Hence, the higher average value at 900 and 950 °C corresponds to a lower average primary excess air ratio.

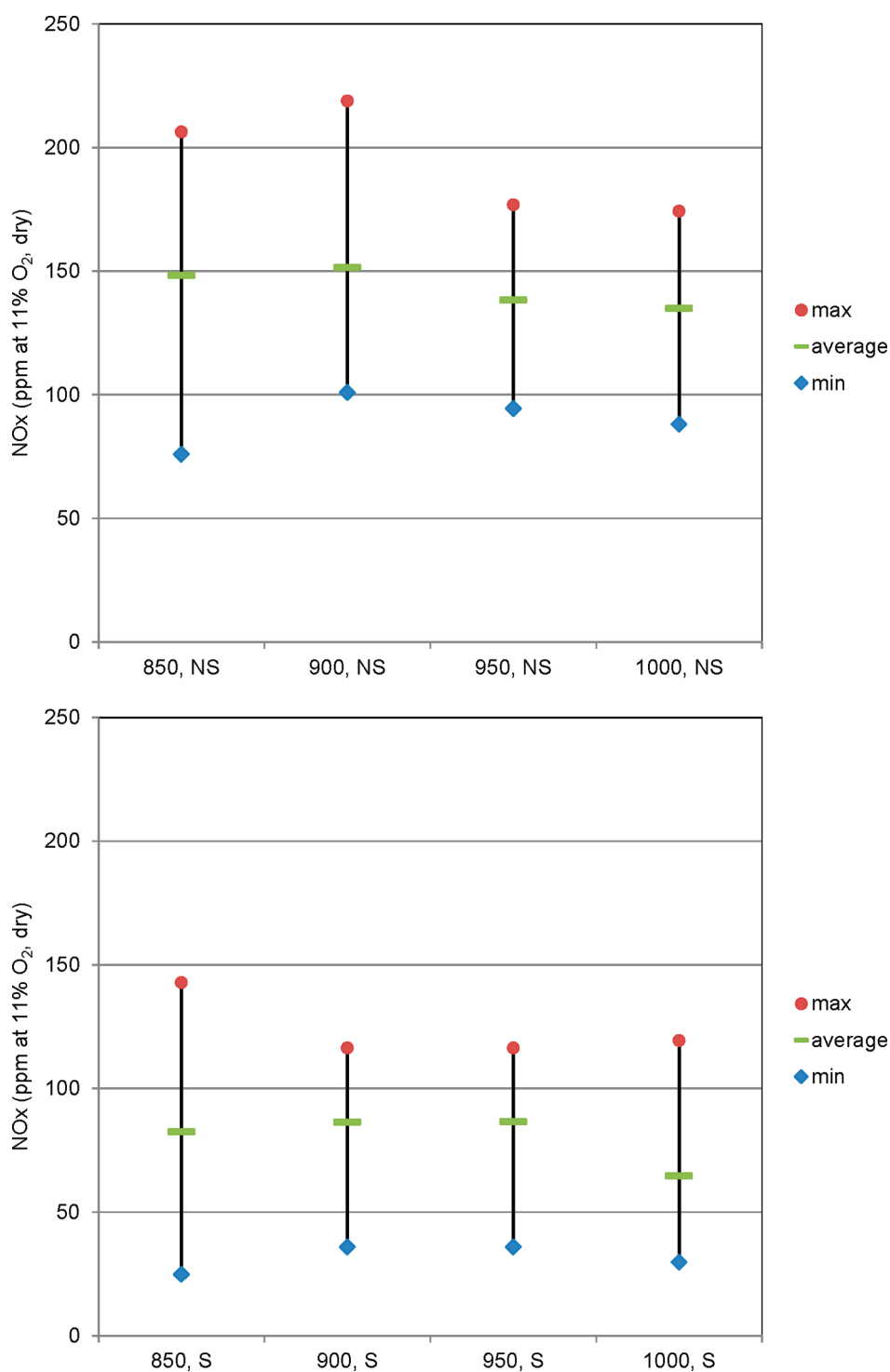


Figure 9. Average, minimum, and maximum value of NOx as ppm corrected to 11% O₂ in dry flue gas for staged air combustion (S) and nonstaged combustion (NS) at the different temperature levels.

N₂O has a different behavior in relation to temperature, showing similar values for staged air combustion and nonstaged combustion. However, temperature has a great influence on mean, maximum, and minimum N₂O levels, shown in Figure 10. They decrease significantly with increasing temperature, and for high temperatures the N₂O level is almost negligible. Yet again the spread between maximum and minimum values are higher in the lower end of the temperature range. Previous studies shows that

the main precursor for N₂O formation is HCN oxidation,^{12,44} while N₂O destruction is mainly by the reaction of N₂O + H = N₂ + OH.⁴⁵ At the high temperatures, the destruction reaction with H radicals is faster and also the HCN oxidation reaction tends to NO formation instead of N₂O. This mechanism causes the N₂O level to be very low at higher temperatures.

3.4. Effect of Stoichiometry, Temperature, and Residence Time on TFN/Fuel-N. In addition to the reactor temperature,

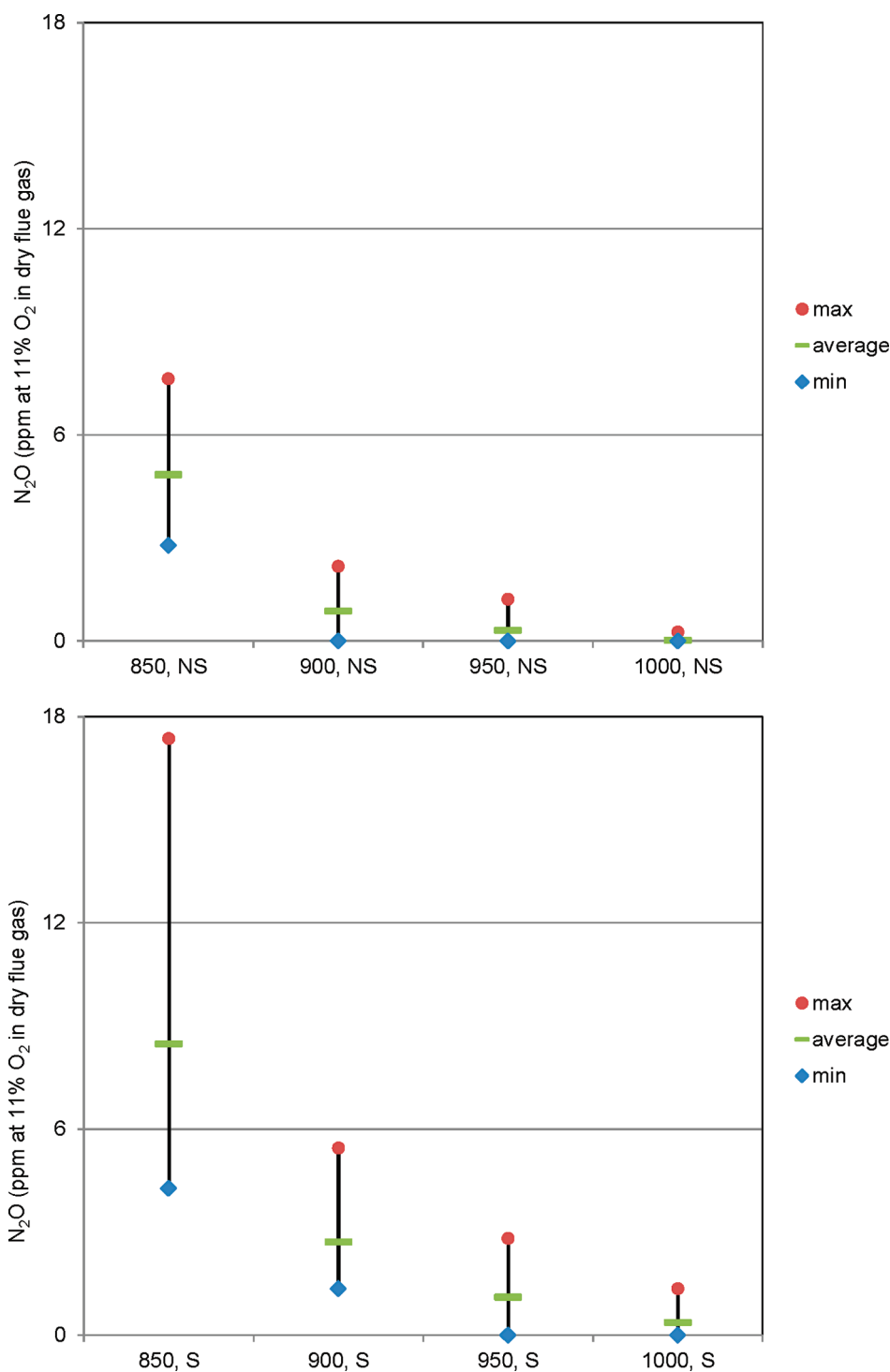


Figure 10. Average, minimum, and maximum value of N_2O as ppm corrected to 11% O_2 in dry flue gas for staged air combustion (S) and nonstaged combustion (NS) at the different temperature levels.

primary excess air ratio, fuel-N content, differences in residence times, and mixing conditions may also have an effect on the fuel nitrogen conversion. The optimum condition for maximum conversion of fuel-N to molecular nitrogen, N_2 , is highly affected by the mentioned parameters. However, since only one fuel was tested, the primary excess air ratio is the only important variable in addition to the potential temperature effect regarding the NO_x emission level. The $\text{NO}_x/\text{Fuel-N}$ and $\text{TFN}/\text{Fuel-N}$ ratios, as

shown in Figure 11, were checked and showed good consistency, always well below unity,⁸ since the values for TFN ($\text{TFN} = \text{NO} + \text{NO}_2 + 2\text{N}_2\text{O} + \text{HCN} + \text{NH}_3$) is calculated after complete burnout where the residence time has been several seconds. Residence time in the reduction zone is of importance for nitrogen conversion, however up to a certain value, beyond which it does not significantly influence $\text{TFN}/\text{Fuel-N}$.^{18,46} In this work, the residence time is well above the expected needed time

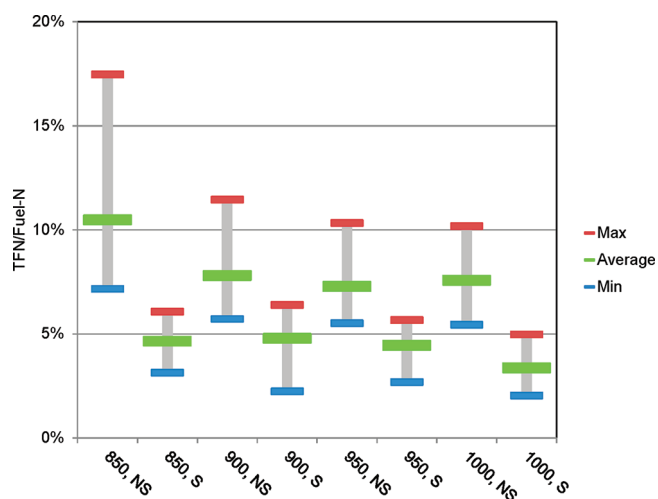


Figure 11. Average, minimum and maximum values of TFN/Fuel-N for staged air combustion (S) and nonstaged combustion (NS) at the different temperature levels.

(below 2 s) and, hence, the results are not influenced by the residence time. After the reduction zone, the combustion is completed by injection of the secondary air. Of course, good mixing of secondary air and the combustible gases are important. In average, a large amount of the TFN is composed of NO_x (96%) and is varying between 81 and 100% depending on the temperature and the combustion scenario. Therefore the mentioned values in the last figure, Figure 11, can be regarded as approximate values for NO_x/Fuel-N.

4. CONCLUSIONS

From the experimental studies carried out in the present work, using demolition wood pellets as fuel, the following conclusions can be drawn: The primary excess air ratio is the most important parameter which can be optimized for maximum conversion of fuel-N to N₂, hence reducing the NO_x level. The effect of two-stage combustion of biomass is significant for reduction of NO_x emission levels. Staged air combustion, in this case, can reduce the emission level by 50–75% and even up to 85% at the optimum conditions. The maximum NO_x reduction happens when the primary air is injected at a primary excess air ratio of 0.8–0.95 and the total excess air ratio is 1.6–1.9 for staged air combustion. The experiments show that also the total excess air ratio has an important effect on the NO_x emission.

The N₂O level has an inverse effect for staged air combustion, i.e., increasing for staged air combustion. The average N₂O level increase for the different temperatures is 75–1660% but with a low amount of N₂O at high temperatures.

However, the experimental results show that the NO_x emission level is not affected significantly by temperature neither in nonstaged or staged air combustion when temperatures are kept below 1000 °C in the reactor. Yet, the effect of temperature on the N₂O level is considerable. This study shows, as expected, that at high temperatures, N₂O emissions almost disappear. The results point to that the effects of temperature and staged air combustion are counteractive for N₂O, meaning a negative influence on N₂O for staged air combustion and a desirable N₂O reduction effect with increasing temperature. Therefore to

minimize N₂O emissions and reduce emissions of unburnt, temperatures of above 900 °C are beneficial.

Hydrocarbons emission, C_xH_y, is almost independent of the combustion condition whether it is staged air combustion or nonstaged combustion. In both cases, the emission levels of hydrocarbons are below 4 ppm at 11% O₂ in dry flue gas. CO emissions increase for staged air combustion compared to nonstaged combustion by a factor of about 1.5 at a given temperature. Increasing temperature decreases the CO emission level in the whole temperature range of 850–1000 °C.

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