Measurements of NOx emissions from biomass combustion in small to medium-scale power plants

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Abstract

Measurements at two different biomass-fuelled combustion power plants are conducted in order to investigate NO_x emissions. The thermal energy input of the industrial-sized combustion facilities ranges from 0.85 MW_{th} for the injection combustion to 40.4 MW_{th} for the bubbling fluidized bed. These two facilities with different combustion technologies also employ different fuels, e.g., wood chips, waste wood and pencil residues. The measurements are conducted simultaneously with a two-measuring point approach entailing flue gas measurements near the particle filter and gas measurements in the combustion chamber. A specially designed high-temperature probing lance is implemented to collect gas from the combustion chamber before the gas is analysed with a Fourier-transformed infrared spectrometer (FTIR). This analysis method enables the measurement of NO_x and its intermediates (NH_3 and HCN) in the combustion zone and a comparison with the emitted NO_x downstream. The results indicate that the emission levels depend on the fuel characteristics and are slightly below but close to the permitted limits.

Introduction

In many parts of the Globe, the generation of electricity and heat heavily relies on combustion processes. However, as the shift away from fossil fuels, particularly coal, gains momentum, the utilisation of biomass and biogenic residues is becoming increasingly important [1]. It should be noted that nitrogen oxide gases (NO_x) are produced during combustion, which pose significant environmental and health risks [2]. Therefore, it is crucial to closely monitor and control the emissions of these gases, mainly when using various types of biomass and biogenic fuels. This is because these fuels may contain a significant amount of nitrogen, which can result in higher NO_x emissions during combustion.

The formation of nitrogen oxides from biomass combustion may come from the oxidation of the nitrogen in the air or in the fuel and it can be categorized into three forms [3, 4]:.

- Thermal NO_x are formed at high temperatures with the oxidation of the N_2 in the air.
- Prompt NO_x are formed through the reaction of nitrogen and oxygen from the air in the presence of radicals of hydrocarbons. Usually in fuel rich conditions.
- Fuel nitrogen oxides are formed from the chemically bonded nitrogen in the fuel, which is released and oxidized. This pathway is the main contributor to the NO_x emissions from biomass.

A myriad of factors influences the release of fuel-bound nitrogen, e.g., temperature, heating rate, excess air ratio, air staging, and the nitrogen compounds in the fuel. Additionally, the volatile content of the fuel also influences the release behaviour of nitrogen, and since biomass usually has a high volatile content, most of the fuel nitrogen is released during volatilisation [5–8]. The released species are mainly NH_3 and HCN with the former being the main component [6, 9]. However, the distribution of these species depends on how the nitrogen is bound in the fuel and on the system temperature [8–10].

In order to reduce the emission of NO_x, different methods can be applied and they are usually categorized into primary and secondary measures. The primary measures, such as air or fuel-staging, usually change the combustion behaviour and, therefore, can directly contribute to minimizing the conversion of fuel nitrogen into oxides or reduce the already formed NO_x into N_2. Whereas secondary measures include the addition of reducing agents into the flue gas, such as ammonia and ammonia derivatives. The secondary measures can be further separated into selective non-catalytic reduction (SNCR) and selective catalytic reduction (SCR). [11, 12]

This study conducts measurements at two different power plants or industrial-sized combustion facilities to investigate the NO_x emissions from biomass combustion. The size of these facilities is 0.85 MW_{th} for the injection furnace and 40.4 MW_{th} for the bubbling fluidized bed power plant. A two-point measurement approach is employed, involving simultaneous flue gas measurements near the particle

filter and gas measurements in the combustion chamber. The gas from the combustion chamber is collected using a specially designed high-temperature steel probing lance and is analysed using Fourier transform infrared spectroscopy (FTIR). The FTIR analysis enables the measurement of both NO_x and its intermediate species (NH_3, HCN), which are highly important for understanding the distribution of the nitrogen species in the gas phase. Furthermore, these intermediates can even reduce NO_x emissions when in favourable conditions [4, 13]. The composition of gases in the combustion chamber and downstream areas is compared, with a particular focus on the nitrogen species.

Experimental set-up and power plants

The measurements at the power plants were gathered using a two-point measurement system, where the flue gas was analysed near the electrostatic precipitator, simultaneous to measurements in the combustion chamber. The gas measurement device utilised for the flue gas downstream was a MGA Prime from *MRU Messgeräte für Rauchgase und Umweltschutz GmbH* with its own probing lance and particle filter, able to measure NO, NO_2, SO_2, CO, CO_2, CH_4, C_3H_8 and O_2. The gas from the combustion chamber was collected using a high-temperature probing lance specially designed for the specific power plants. The probing lance is air-cooled and has a ceramic tube at the tip. The gas passes a heated tube at 350°C and reaches an electrically heated filter also at 350°C. The high-temperature filtering is needed in order to avoid loss of ammonia due to adsorption on the solids or salt formation [14, 15]. After being filtered, the flue gas enters a preconditioning unit, where it is separated into two measuring devices, a multi-component gas analyser VarioLuxx from *MRU Messgeräte für Rauchgase und Umweltschutz GmbH* for O_2 measurements and a portable multi-component FTIR gas analyser *Gasmet DX4000* able to measure H2O, . To obtain a more detailed analysis of the interior of the combustion chamber, the probing lance was positioned in the opening at different depths and gas was sampled from closer to the furnace wall and further in the chamber. The set-up is represented schematically in **Figure 1**.
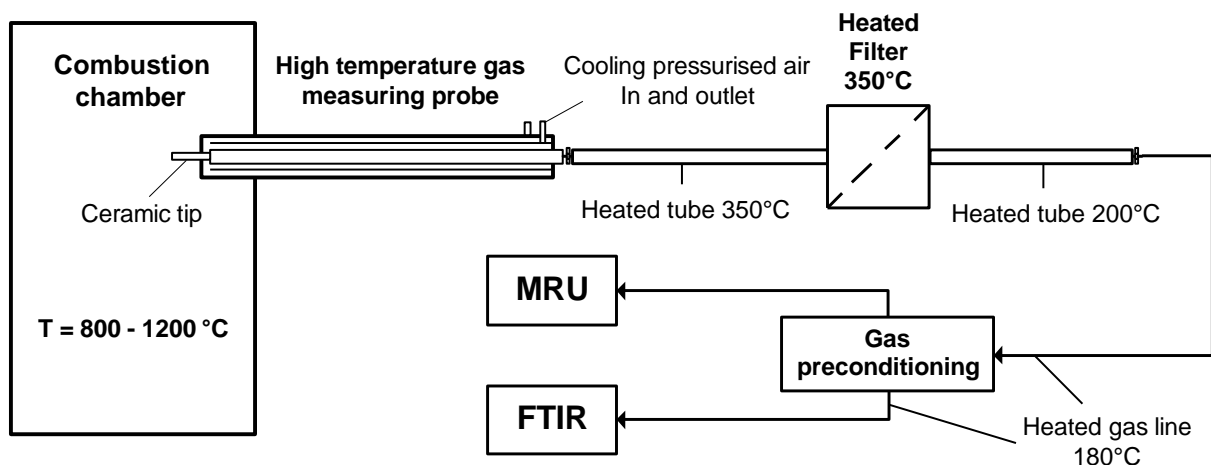

Figure 1. Schematic representation of the employed measurement system.

Experiments were conducted at two different power plants. The first is an injection combustion plant for the in-house heat production of the company Staedtler [16]. In contrast, the second power plant is a combined heat and power cycle using waste wood and other waste products as fuel, e.g. plastics, in a bubbling fluidized bed combustion reactor in Altenstadt, Germany [17]. The main characteristics of the two power plants are summarised in **Table 1**.

Table 1: Summary of the power plants with key characteristics

	Power plant 1	Power plant 2
Company	Staedtler	Heizkraftwerk Altenstadt GmbH & Co.KG
Type of plant	Injection furnace	Bubbling fluidised bed
Output thermal power	0.85 MW	40.4 MW
Output use	In-house heat	Heat and power
Type of fuel	Residue from pencil production	Waste wood
Location	Neumarkt i. d. O., German	Altenstadt, Germany

For power plant 1, the boiler and its main components are represented schematically in **Figure 2**. Furthermore, the measuring points are also represented. The fuel used is waste wood and pencil shavings from the pencil production and it is fed through a screw together with primary air into the combustion chamber. The first measuring point was just above the fuel and primary air injection zone. Moreover, flue gas recirculation and air-staging are performed, and the flue gas goes through the heat exchangers before being filtered and emitted through the chimney. No further NOx mitigation methods are employed. The second measuring point is located just after the electrostatic precipitator.
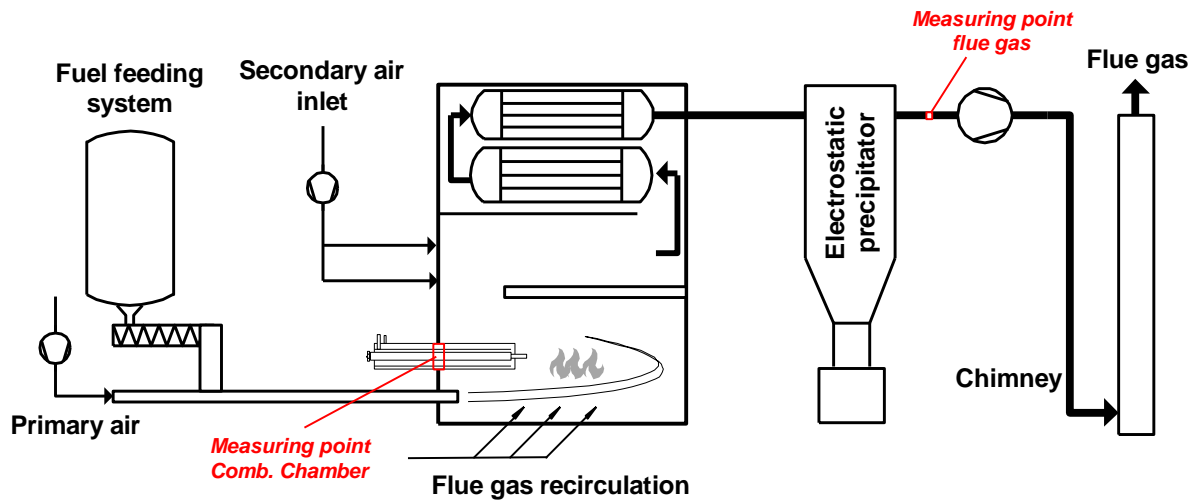

Figure 2. Schematic drawing of the injection combustion plant from Staedtler.

A drawing of the power plant 2 is given in **Figure 3**. As depicted, the primary air is injected from the bottom as the fluidizing medium for the inert material quartz sand, and the secondary air is added 6 meters above. The flue gas then passes through the heat exchangers and is separated in streams for flue gas recirculation and for the chimney. For the measuring point in the combustion chamber, two openings were used 9 meters above the primary air inlet. The flue gas was measured from the recycling stream just before the filter. During the measurements presented here, the implementation of SNCR was being investigated and discussed but not yet built.

The measurements in the flue gas stream were conducted from gas retrieved as close to the centre of the pipe as possible. For the measurements in the combustion chamber, the gas was extracted at different gas probe depths, i.e., the sampling probe was inserted into the opening and moved axially to retrieve gas close to the chamber wall and further in.
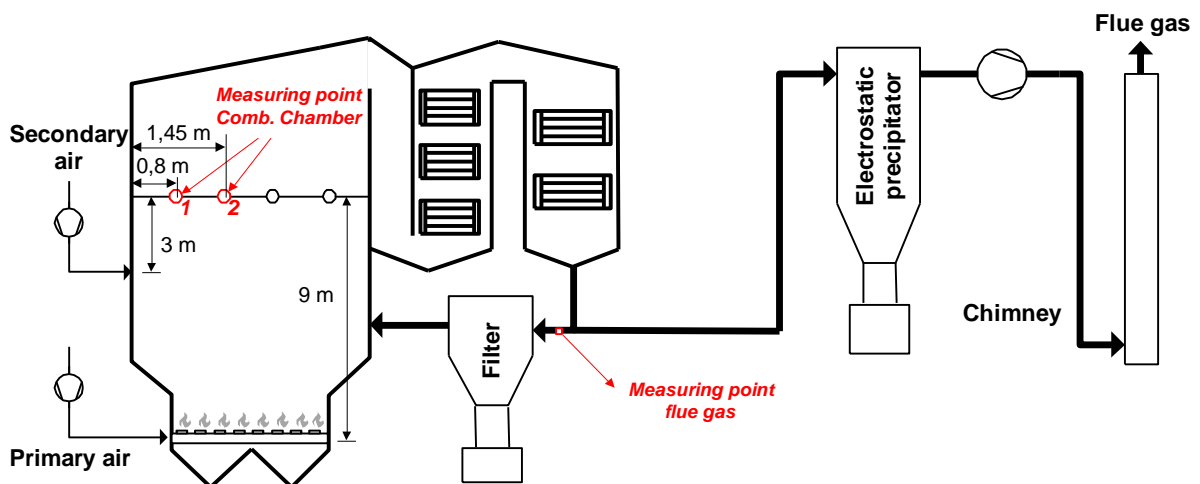

Figure 3. Schematic drawing of the fluidised bed combustion power plant in Altenstadt

Results and discussion

The measurements for each point were conducted for at least 30 minutes at each power plant. An example course of the gas concentration over one hour is depicted in **Figure 4** for the nitrogen species as well as for O_2, CO_2 and CO. The values of the gas species fluctuate and are dependent on the

technology implemented at the power plant. However, the values display a stationarity over the course of the measured period. In order for the values to be comparable between power plants and to avoid any air dilution, the measured concentrations for the flue gas have to be converted using a reference oxygen content $x_{O_2,ref}$ of 6 vol-% according to the German 44th Federal Immission Control Ordinance (BlmSchV) for medium-sized combustion plants as given in **Equation 1** [18]. In this work, the gas concentration values from the flues gas are given with the reference oxygen content.

$$c_{x,ref} = \frac{20.95 - 6}{20.95 - x_{O_2,m}} \cdot c_{x,m}$$ **Equation 1**

Where $c_{x,ref}$ is the converted concentration of the gas component to the reference oxygen content, $c_{x,m}$ is the measured concentration of the gas component and $x_{O_2,m}$ is the measured oxygen content of the gas in vol-%.
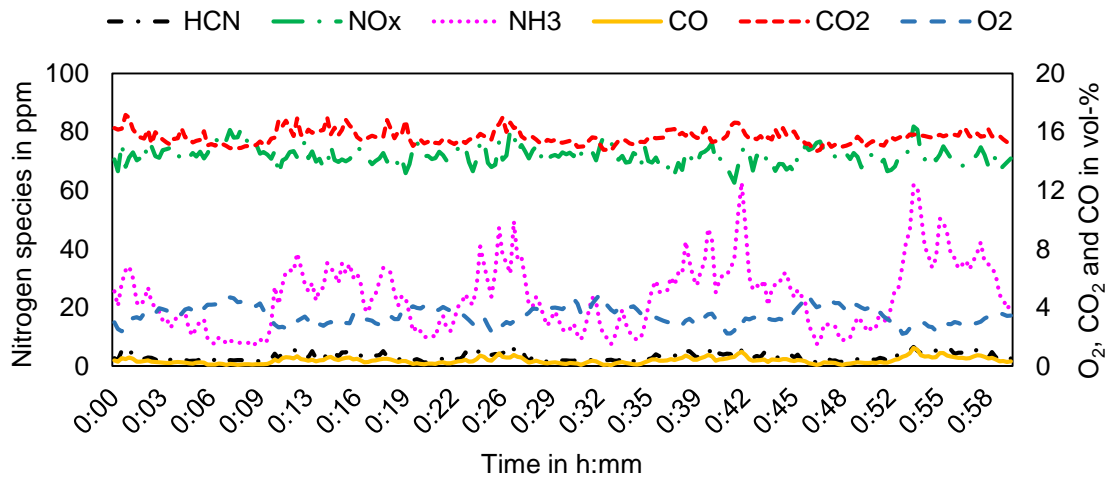

Figure 4. Example course of the gas concentration from the combustion chamber over one hour for the power plant 1

The air-fuel-ratio λ, which determines the stoichiometric conditions of the combustion process, can be approximated for values greater than 1 as in **Equation 2**, due to the surplus of oxygen, as follows:

$$\lambda = \frac{20.95}{20.95 - x_{O_2,m}}$$ **Equation 2**

A proximate analysis of the fuels from Staedtler and Altenstadt was performed using the norms DIN EN ISO 18134-3, DIN EN ISO 18122, DIN EN ISO 18125. Furthermore, the elemental analysis was performed with a Vario Macro Elementar Analyser and the data is given in **Table 2**.

Table 2. Fuel analysis from the power plants

Fuels used in	C	H	N	S	O	a	Volatiles	w	LHV
	wt-% (db)							wt-%(ar)	MJ/kg
Power plant 1	47.48	5.31	0.58	0.06	46.28	0.30	80.46	6.35	19.50
Power plant 2	35.94	4.00	1.77	0.17	35.13	23.00	56.22	7.70	15.23

C: Carbon; H: Hydrogen; N: Nitrogen; S: Sulphur; O: Oxygen; a: Ash; w: Moisture; LHV: Lower Heating Value; db: dry basis; ar: as received

Injection furnace (0,85 MW$_{th}$)

During the measurement period at the power plant 1, the flue gas after the filter had an average NO_x value of 126.59 mg/Nm³. The measurements inside the combustion chamber were performed at different gas sampling depths and a maximum depth of 60 cm was reached due to the wall thickness and available opening of the power plant. The concentration of the nitrogen species at different gas sampling depths in the combustion chamber are displayed in **Figure 5** for NO_x, NH_3, HCN as well as the sum of the nitrogen species (NO_x, NH_3 and HCN). The temperature at this zone averaged 1060 °C. It can first be observed a decrease in λ the deeper the gas is sampled with the gas concentration cloud moving to lower λ. Moreover, for the NO_x content, the gas concentration can be calculated using linear regression, as shown in **Figure 5 a)** for the different sampling depths. This results, for a set λ of 1.2, at

a depth of 60 cm, in a NO_x concentration of 172.39 mg/Nm^3. Additionally, NH_3 was the dominant intermediate component for biomass, as described in the literature [5, 13, 19].

The sum of the nitrogen species is displayed in **Figure 5 c)**, and it can be observed that the total nitrogen species does not drop below 80 ppm. The measured values closer to the inner wall are constant and do not display a dependency on the oxygen availability. On the other hand, however, the deeper measurement displays a rapid decrease in the nitrogen species concentration with increasing air-fuel ratio. These results lead to believe in an effect on the gas velocity and thus on the mixing and concentration profiles in the thickness up to 40 cm from the wall possibly due to a boundary layer [20].

Lastly, comparing the gas in the combustion chamber to the stack NO_x at the electrostatic precipitator, a total NO_x reduction of 25.27% was measured. Although air-staging is performed in between the measured points, the presence of the intermediate species could also lead to a reduction of the NO_x concentration further down-stream.
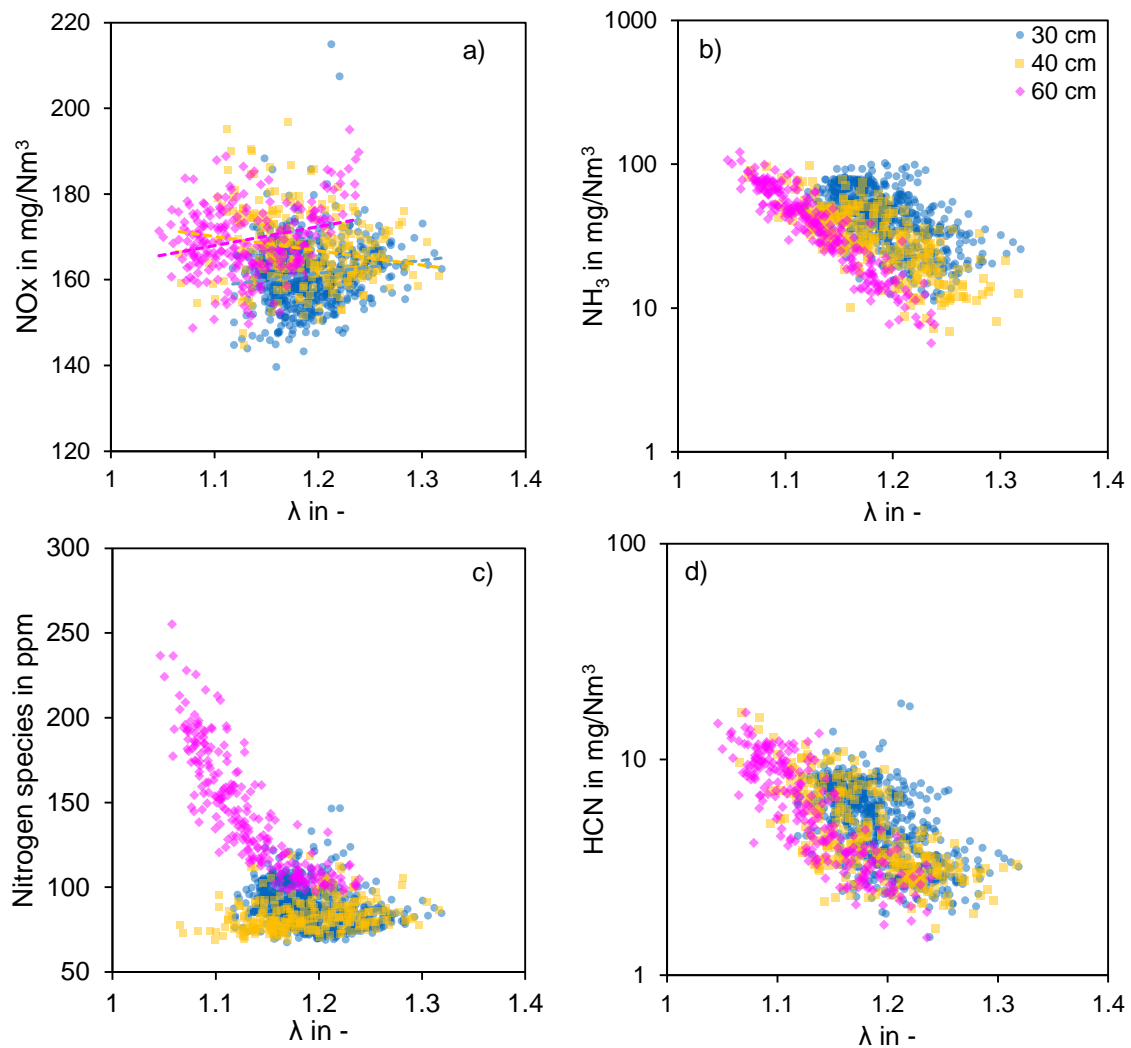

Figure 5. Gas concentration with reference 6% O_2 in the power plant 1 combustion chamber at different gas sampling depths a) NO_x b) NH_3 c) HCN d) Total nitrogen species

Bubbling fluidized bed power plant (40.4 MW_th)

For the time of the measurements at the opening number 2, the NO_x concentration in the flue gas of the fluidized bed power plant was stable and averaged 133.81 mg/Nm^3. This is lower than the emission limit of 200 mg/Nm^3 for solid biomass firing furnaces over 20 MW$_{th}$ [18]. However, at a later time, during the measurements at the opening number 1, the average NO_x concentration was 239.65 mg/Nm^3. This is due to the very inhomogeneous fuel used, waste wood, which can have very different characteristics and fuel nitrogen content leading to higher emissions. Overall the average NO_x emissions during the measurement period was 163.80 mg/Nm^3.

The gas measurements from point number 1 were performed close to the chamber's inner wall because of the geometry of the opening in relation to the combustion chamber and delivered therefore inconsistent values. Hence the gas concentration from opening number 1 are not discussed further. On the other hand, the concentration of the nitrogen species from the measuring point number 2 in the combustion chamber after air-staging are depicted in **Figure 6** for NO_x, NH_3, HCN and total nitrogen species. The temperature in this zone ranged from 890 to 910 °C. The maximum depth reached was 50 cm due to the wall thickness and the available opening of the fluidised bed reactor, which is a flange with a ball valve.

It can be observed again that deeper sampling leads to on average lower excess oxygen values. Furthermore, at a sampling depth of 50 cm, the NOx concentration at λ of 1.2 was 247.88 mg/Nm³. By comparing the measured gas concentration of NO_x directly above the fluidized bed with the emitted flue gas during the measurements in the opening number 2, a reduction of 46.02% can be observed.
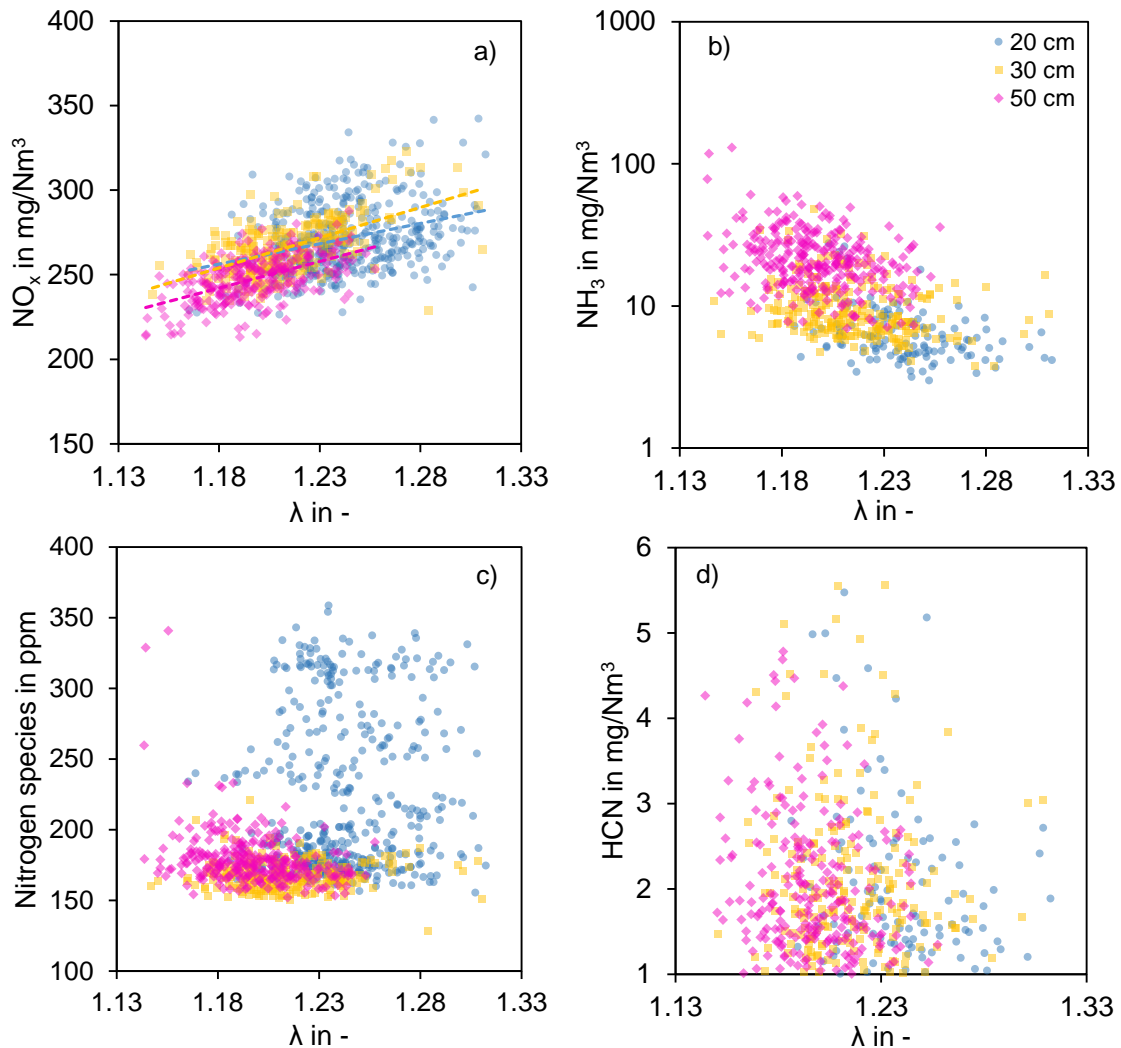

Figure 6. Gas concentration with reference 6% O_2 from power plant 2 combustion chamber at different gas sampling depths a) NO_x b) NH_3 c) HCN d) Total nitrogen species

The NH_3 concentration increases with increasing gas sampling depth, whereas for HCN this effect is not observed and the concentration are very low. Here, the NH_3 concentration is also the dominant species. The total concentration of the nitrogen species, as in **Figure 6 c)**, shows a lower limit of around 150 ppm and is quite stable for the range of the varying λ. The higher content of nitrogen species at the sampling depth of 20 cm can be due to a boundary layer off the inner wall leading to diminished gas mixing. Moreover, the trend of the nitrogen species from power plant 2 are different than from power plant 1. This is due to the difference in gas sampling position and power plant geometry, since the sampling from power plant 2 was done after air-staging whilst for power plant 1 it was done before.

By comparing the fuel nitrogen content to the NO_x emissions as displayed in **Table 3**, it can be observed that the emissions are greater for the fuel with the higher amount of nitrogen. Although no direct comparison of the combustion technologies can be applied, the trend of emission dependency on the initial nitrogen content has been verified [21, 22]. Moreover, both furnaces displayed a minimum of total nitrogen species across the varying oxygen content in the combustion chamber.

By comparing the NO_x emitted to the nitrogen species in the combustion chamber, on the one hand, a small decrease of nitrogen species can be detected for the injection furnace. On the other hand, however, the reduction of the NO_x and its intermediates for the fluidized bed furnace is greater. Although the measurements of the combustion gases occur during different steps of the combustion process, the difference in the reduction can be explained by better mixing of the gases in the fluidized bed power plant.

Table 3. Comparison of the NO_x emissions and nitrogen species concentration in ppm with a reference O_2 content of 6 vol-% at a sampling depth of 60 cm for power plant 1 and 50 cm for power plant 2 with the initial fuel bound nitrogen.

	Fuel-N	Emission limit	Stack NO_x	NO_x in-furnace at $\lambda=1.2$	N-Species in-furnace
	wt.-%	ppm 6% O_2			
Power plant 1	0.58	365.40	61.67	83.99	143.88
Power plant 2	1.77	97.44	72.46	120.77	152.93

Conclusions

Measurements were conducted at two different furnaces, an injection furnace and a fluidized bed power plant. A two-point system was implemented with measurements of the flue gas before being emitted to the atmosphere and of the gas in the combustion chamber. The gases from the combustion were sampled at high temperatures with a specially designed air-cooled steel probing lance with ceramic tip and they were analysed using an FTIR gas measuring device coupled with another portable gas measuring device for the oxygen analysis. The lance was introduced to the combustion chamber and sampled gas at different depths. It was observed that the gas concentration changed depending on the depth of the sampling from the combustion chamber with measurements closer to the wall having higher oxygen concentrations due to a boundary layer effect. Moreover, the measurements at the combustion chamber displayed a dominance of NH_3 over HCN.

The total nitrogen species displayed a minimum level in the combustion chamber for both firing concepts and by comparing the combustion gas to the flue gas, the nitrogen species were further reduced before being emitted, with the fluidized bed reactor having a greater nitrogen reduction than the injection furnace.

The gathered data can be applied in computational fluid dynamic simulations in order to gain further insight into possible reduction potential of the NO_x emissions from solid biomass combustion.

Acknowledgement

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