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Analysis of Oscillating Combustion for NO_x—Reduction in Pulverized Fuel Boilers

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Abstract: Thermal power plants in different fields are regularly adapted to the state-of-the-art emissions standards, applying "The Best Available Techniques Reference". Since 2016 in the power plant area new, more stringent limits for power plant units with a thermal output of more than 300 MW operated with black coal are valid. Usually, in order to reach the new limits e.g., for NO_X emissions, downstream reduction processes (Selective Non-Catalytic Reduction, SNCR or Selective Catalytic Reduction) are applied, which use of operating resources (essentially ammonia water) thereby increase. By the means of an experimentally validated process, by which pulverized fuel is fed by oscillation through a swirl burner into a pilot combustion chamber with a thermal output of 2.5 MW, nitrogen oxides can be reduced without further activities, for instance from 450 $mg/m_N{}^3$ in non-oscillation operation mode (0 Hz) to 280 $mg/m_N{}^3$ in oscillation operation mode (3.5 Hz), normalized to an O_2–content of 6% each. These findings were patented in EP3084300. Particularly promising are the experiments which utilize oscillation of a large portion of the burn out air instead of the fuel in order to minimize the fatigue of the pulverized fuel oscillator, amongst others. Thereby, the nitrogen conversion rate, which describes the ratio of NO_X to fuel nitrogen, including thermal NO_X can be reduced from 26% for non-oscillation operation mode down to 16%. The present findings show that fuel oscillation alone is not sufficient to achieve nitrogen oxides concentrations below the legislative values. Therefore, a combination of different primary (and secondary) measures is required. This paper presents the experimental results for oscillating coal-dust firing. Furthermore, an expert model based on a multivariate regression is developed to evaluate the experimental results.

Keywords: oscillating combustion; NOx-reduction; pulverized fuel; boiler

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Citation: Jolibois, N.; Aleksandrov, K.; Hauser, M.; Stapf, D.; Seifert, H.; Matthes, J.; Waibel, P.; Vogelbacher, M.; Keller, H.B.; Gehrmann, H.-J. Analysis of Oscillating Combustion for NO_x– Reduction in Pulverized Fuel Boilers. *Inventions* **2021**, *6*, 9. https://doi.org/10.3390/inventions6010009

Received: 19 December 2020
Accepted: 15 January 2021
Published: 19 January 2021

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1. Introduction

Today, despite numerous alternative and renewable energy sources, 80% of the world's primary energy demand is accounted for by fossil fuels. In Germany, coal accounted for 45% of electricity generation in 2017 [1]. Energy conversion from chemical to electrical energy takes place almost entirely via combustion processes of fossil or alternative energy sources, such as waste or biomass.

Combustion processes produce a large number of air pollutants that are harmful to the environment. Against the background of climate change and the public debate on environmental pollution, the reduction of pollutants is of great importance. For this reason, the framework conditions for air emissions from industrial plants are defined by the Bundes Imissionsschutz Gesetz (BImSchG). The nitrogen oxides NO and NO_2 are combined as NO_X and play a special role in the assessment of pollutant emissions.

In the troposphere, nitrogen oxides contribute significantly to the formation of ground-level undesirable ozone and photochemical smog, also known as Los-Angeles smog. Furthermore, nitrogen oxides can react with humidity to form HNO_3 and thus lead to acid rain. In addition to the formation of ground-level ozone, NO_X also contributes to the chain reactions of ozone depletion in the stratosphere. This desired ozone protects the earth from increased ultraviolet radiation from the sun. In addition, NO_X acts as a pure substance or in high concentrations as a strong respiratory poison and leads to damage to the respiratory organs.

In order to comply with the limit values for nitrogen oxide emissions, the NO_X concentration in the flue gas of industrial plants is reduced by two different measures. The so-called primary measures are based on the combustion fundamentals of nitrogen oxide formation. These formation mechanisms can be inhibited by clever air guidance in the combustion chamber. Subsequent denitrification of the flue gas is one of the secondary measures and includes processes such as Selective Catalytic Reduction (SCR) and Selective Non Catalytic Reduction (SNCR). Here, the nitrogen oxides are reduced to ammonia by means of a catalyst or a reducing agent such as urea. The subsequent treatment of the flue gases is usually associated with higher investments and a decrease in the efficiency of the plant, which is why primary denitrification is more economical and ecological [2].

Oscillating combustion is one of the primary measures of nitrogen oxide reduction and is part of current research [3]. The basic principle of oscillating combustion is the interruption of either the fuel or part of the combustion air to change the local stoichiometry. The stoichiometry as the ratio of supplied air to fuel is one of the main influencing parameters to minimize the NO_x formation. The rate of reduction could reach 50% and more compared to non-oscillating combustion, depending also on temperature, residence time and mixing conditions.

Up to now, the oscillating addition of fuel has led to significant reductions in NO_X emissions. The reason for this is the changing stoichiometry that occurs in the flame range, as a result of which the reaction mechanisms of nitrogen oxide formation lead to less NO_X. The investigations carried out up to date have largely been limited to the oscillating addition of natural gas [4], which is easier to oscillate in the process. Due to the high abrasive properties and the associated process engineering challenges in the oscillating combustion of coal dust, there are hardly any investigations in this area. For the investigations presented in this paper the burner was equipped with a valve to initiate the oscillating combustion.

Due to the procedural challenges of oscillating coal dust, the focus of this work was on the reduction of nitrogen oxide emissions under oscillating addition of the entering fuel-free air volume flows.

The goal of this paper is to evaluate the reduction potential for nitrogen oxide emissions based on experimental results from a pilot-scale combustion plant with a coal dust burner and different oscillation setups. For the evaluation, an expert model based on a multivariate regression model is applied for.

2. Basic Principles

2.1. Fuel-NO Formation

Since in these experiments the NO formed results mainly from the fuel NO mechanism, this is described in more detail. A distinction has to be made between homogeneous and heterogeneous NO formation. However, according to [5] the heterogeneous kinetic processes of the nitrogen bound in the residual coke are of secondary importance compared to the homogeneous kinetic processes of the N-containing components of the fuel. According to [6], the typical nitrogen content for water- and ash-free coals is 0.5–1.6%. Nitrogen is present in fuels either in inorganic compounds or in organic compounds [7].

Since the findings of Fenimore it has been the subject of numerous investigations to understand the complex interrelationships of the fuel-N mechanism, ergo the formation and degradation of HCN, NH_3, NO and N_2 [8]. References [7–10] give detailed literature overviews on the fuel-N mechanism, whose simplified scheme in Figure 1 is shown in green with the other two formation mechanisms, thermal in orange and prompt in blue.

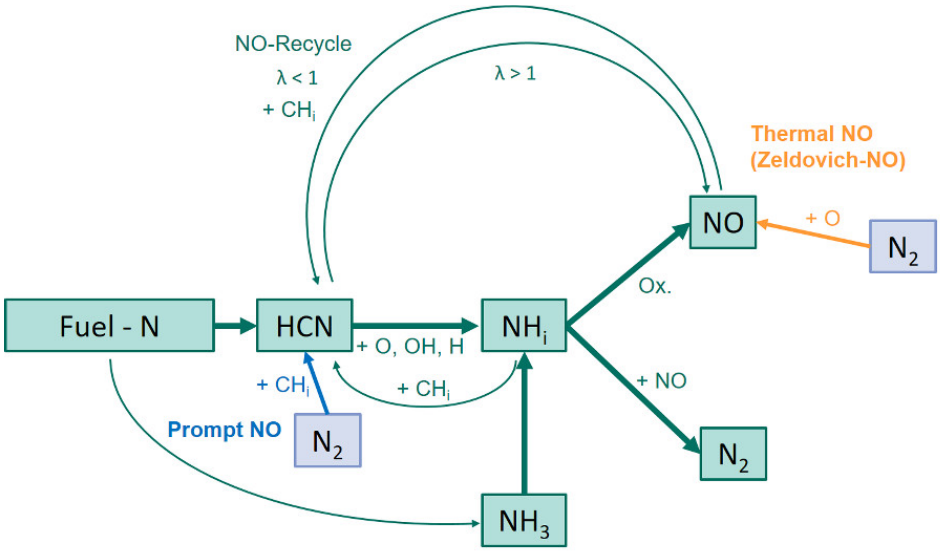

Figure 1. NO formation mechanisms [8].

The fuel-bound nitrogen reacts via thermal decomposition and radical reactions by attacks of H, O, and OH in a rapid reaction to HCN and a subsequent rate determining reaction to NHi. The NH_i molecule then reacts either by oxidation to nitric oxide or under reducing conditions with NO to molecular nitrogen. The NO-recycling reaction of nitric oxide to HCN takes place under CH_i radicals and air deficiency effects and allows the formed nitrogen monoxide to be returned to the fuel N reaction path. In this way, NO that has already been formed can be returned to molecular nitrogen depending on the air ratio λ.

The phenomenon described above of the dependence of the nitrogen oxides formed on the stoichiometric ratio forms the basis for staged combustion as a primary measure for nitrogen oxide reduction. A more detailed description of the NO-Recycle mechanism can be found. The most important reaction partners are the radicals H, O and OH, whose influnce changes on the reaction path depending on temperature, air ratio and other boundary conditions [5,7,8,10].

The investigations of [11] show in Figure 2 the sum of the N-containing species NO, NH_3 and HCN in the flue gas as Total Fixed Nitrogen (TFN) and the shift of their ratios as a function of the air ratio.

The components are summarized as TFN because HCN and NH_3 are also converted to NO in the atmosphere and are therefore classified as harmful emission components [7]. Starting from a low air ratio of $\lambda = 0.6$, the proportion of the NO precursor species HCN and NH_3 decreases significantly and the NO component increases steadily. For low air ratios of $\lambda < 0.7$, this can be explained by the sub-stoichiometric ratios and low concentration of oxidizing components. With an increasing air ratio for $\lambda > 0.7$, the proportion of the NO_X component increases steadily due to the available oxidizing radicals, whereas the concentrations of the two precursor species are close to zero. The minimum TFN is reached at an air ratio of $\lambda \approx 0.7$ and forms the basis for staged combustion for NO_X reduction, since the conditions for NO recycling are met here Figure 1, a increased conversion to molecular nitrogen [6].

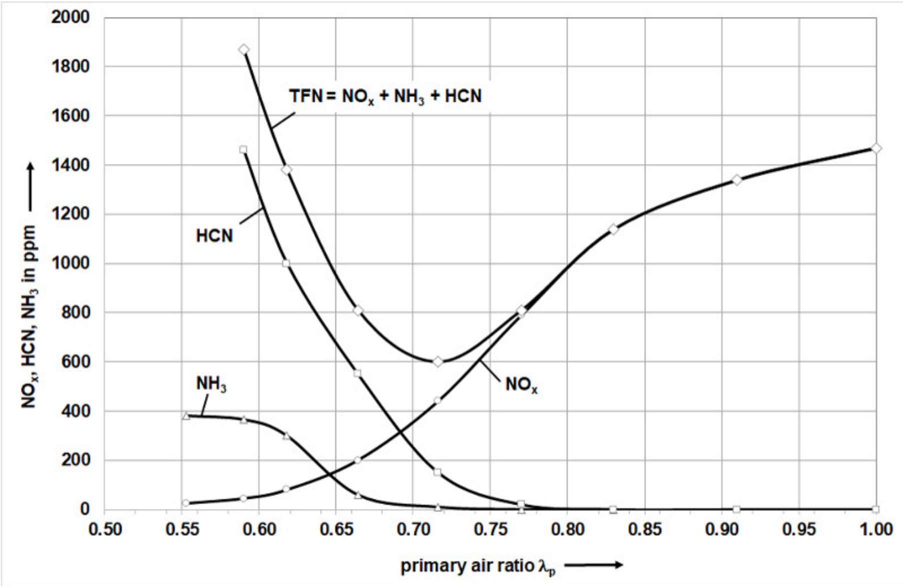

Figure 2. TFN in dependency of the air ratio λ [11].

2.2. Oscillating Combustion as Primary Measure

Oscillating combustion is the state of the art in research and is investigated in numerous studies [3,4,12–14] on the primary reduction of NO_X emissions from furnaces for gaseous fuels.

To the advantages of the oscillatory combustion belong lower operating expense, or in the best-case full omission of secondary measures for NO_X-reduction (SCR, SNCR), due to less formation of NO_X. The disadvantages include higher burner requirements (development of special valves, bigger pipe diameters, abrasion, resonance), as well as lower average burner output at the same burner size.

The result of a field study from 1996 to 2003 on oscillating combustion with gas flames [14] shows a significant reduction in NO_X emissions of 31–67% and an improved heat transfer of the flame of up to 13%. The results were confirmed by [3,4,12,13] for other combustion systems.

The basis is the oscillating addition of fuel or air into the reaction zone of the flame, which causes a temporal change of the local stoichiometry [3]. The different reaction zones occur immediately after the fuel leaves the burner. As the distance to the burner increases, the axial cross mixing of the combustion air progresses. Figure 3 shows this behavior schematically for solid fuels.

The oscillating addition of fuel/air results in oscillating reaction conditions in the reaction zone of the flame. Low-fuel ($\lambda > 1$) and fuel-rich reaction zones ($\lambda < 1$) follow directly one after the other. Figure 3 shows an analogy to the air staging combustion mode. However, the adjustment of the reaction conditions is not carried out by locally targeted dosing of the secondary air, but by oscillating addition of the air. Accordingly, oscillating combustion can be regarded as a time-resolved variant of staged combustion [13]. In the fuel-rich periods of time Δt_1 CO, NH_3 and NO_X are increasingly produced while in the fuel-poor periods of time Δt_2 CO reacts to CO_2 and NO_X with NH_3 to N_2 and H_2O [3].

In the previous work of [4,13,14] the NO_X reduction potential is investigated by a oscillating addition of a gaseous energy carrier such as natural gas. Although [3] shows in his work that a oscillating addition of secondary air during the combustion of chipboard cubes in a fixed-bed reactor also leads to a significant reduction in NO_X emissions, none of the previous investigations shows the effect of the oscillating combustion of a solid on NO_X emissions. The challenge lies in the high abrasion of the valve during the oscillating conveyance of coal dust. In this paper the results of the oscillating combustion of pulverized coal with a coal burner are investigated and complement the previous work of [3].

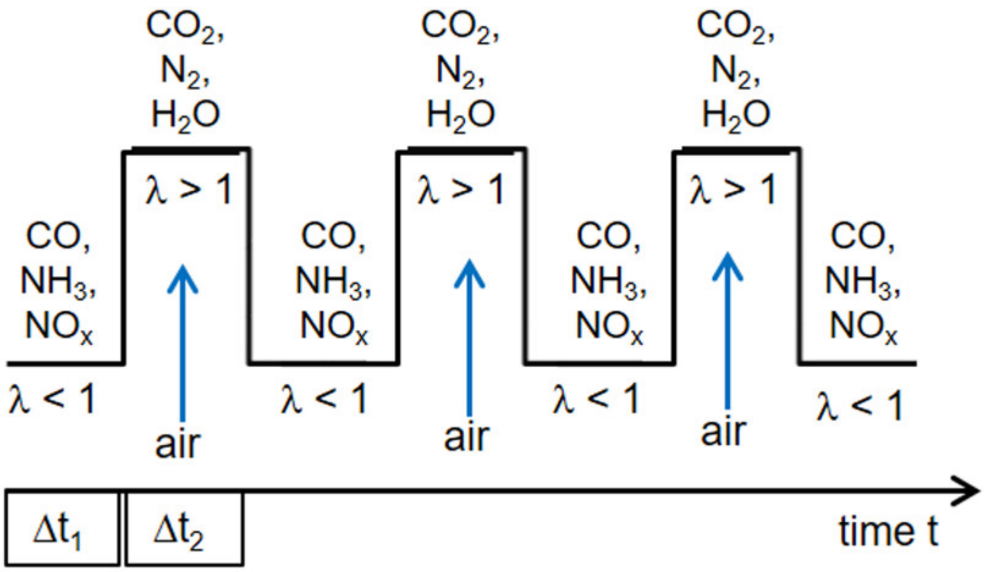

Figure 3. Behavior of stoichiometry in an oscillating combustion [4].

3. Methods

3.1. Pilot-Scale Power Plant BRENDA

The BRENDA power plant pilot plant is used to investigate the combustion and emission behavior of conventional, as well as alternative dusty gaseous or liquid fuels. The focus is on the combination of the fuels and their flexible usage. Against the background of the energy turnaround, both points are among the challenges facing existing power plants. Figure 4 shows the area of the plant section which is part of the investigations carried out in this paper.

The thermal power of the rotary kiln is needed to reach the minimum temperatures according the legal requirements (850 °C above the level 2). An operation without the thermal load of the rotary kiln is not possible.

The rotary kiln burner is required to provide sufficient thermal load for the entire plant and has a thermal output of 1.5 MW. The hot combustion gases pass from the rotary kiln into the afterburner chamber and pass through the measuring point on level 0. Between level 0 and level 2 lies the section in which the pulverized coal furnace is installed. The swirl burner for pulverized coal firing has a thermal output of 1 MW and is manufactured by SAACKE. A gas burner is installed in the opposite furnace wall at the same height as the pulverized coal burner. This ensures that the temperature in the afterburner chamber does not fall below 850 °C in accordance with the 17th BImSchV in the event of failure of the pulverized coal furnace and thus ensures safe operation of the plant. Since the gas burner is not the subject of the investigations, it is not shown in Figure 4. The gas burner was not in operation during the investigations with coal with exception of the ignition burner with 1 m^3/h of natural gas which is required for safety reasons of operation.

The flue gases enter the steam boiler which operates with saturated steam at 220 °C (at 40 bar), so its efficiency is quite low compared to industrial systems. The steam is partially used for air and flue gas pre-heat in the flue gas treatment after the steam boiler but mainly cooled down by ambient air.

In order to clearly determine the NO_X emissions of the pulverized coal burner, the NO_X concentration is measured at level 0 and level 2. Level 0 characterizes the flue gas from the rotary kiln burner which enters the coal burner section. The NO_X emission resulting from the pulverized coal firing is determined by the difference between the NO_X load of the dry flue gas from level 2 and level 0. Since the air ratio at the coal burner was always greater or equal to 1 the NO- recycling mechanism (see Figure 1) is inhibited. Thus, it is assumed that the nitrogen oxides entering level 0 are inert and don't react anymore with other gas compounds. For the discussion of the impact of oscillation on the NOx, in level

"0" and "2" NO_x, CO, O_2 and CO_2 are measured. The emissions of SO_2, HCl etc. are not relevant for this purpose, of course they are analyzed after the boiler and the stack with regard to the emission levels.

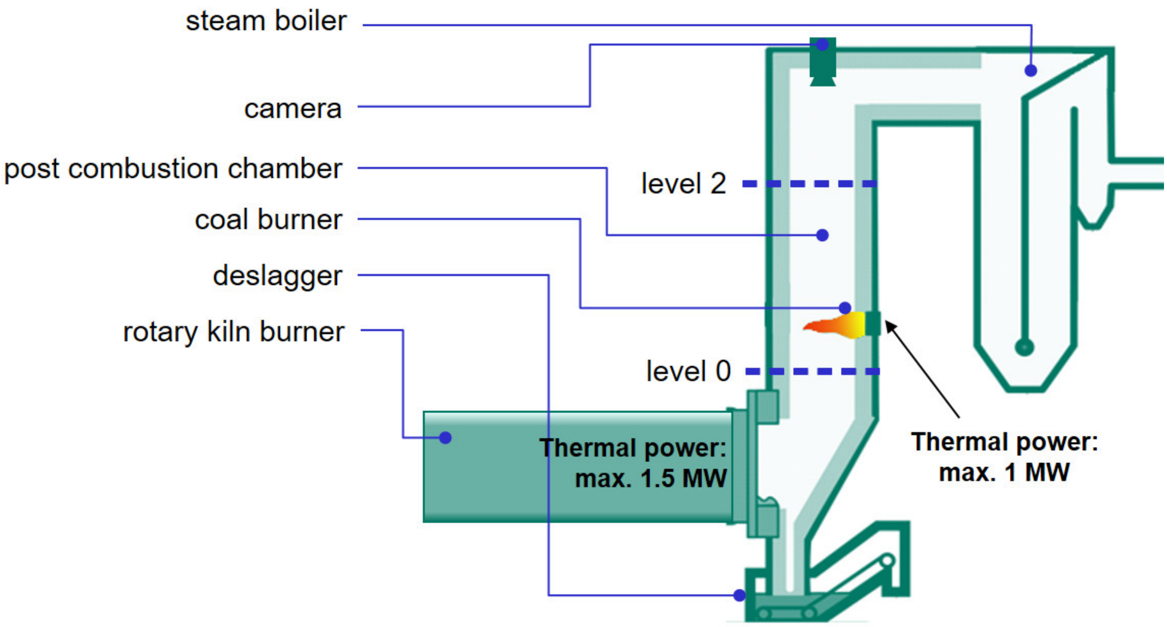

Figure 4. Pilot-scale combustion plant BRENDA.

A high dynamic ranged µEye UI-5240SE-M-GL camera of the manufacturer Imaging Development Systems GmbH (IDS) was aimed vertically at the flame from the coal burner above the post burner chamber in order to analyse the flame image during the experiments. The analysis of the image material was carried out by the Institute for Automation and Applied Informatics [15].

Figure 5 shows a closer look at the geometry of the pulverized coal burner. In the plan view of the burner mouth, the fuel-bearing central tube, annular gap 1, annular gap 2, the burner brick and the combustion air inlet are arranged concentrically. The cone serves to stabilize the flame and is set at a constant distance of 15 mm during the entire test campaign (the maximum diameter of the cone is about 15 mm).

The combustion air passes through the outermost gap into the furnace and is fed tangentially into the burner, whereby it enters the combustion chamber with a swirling flow. during operation, the burner block can be moved along the marked *x*-axis into the furnace wall, whereby the gap width of the combustion air inlet changes. By shifting and/or increasing the combustion air flow, the outlet velocity of the combustion air flow can be influenced.

Equation (1) shows the simplified calculation formula for the swirl number S resulting from this geometry as a function of the position of the traverse path Δx of the burner block in x-direction and the ratio Q of swirled combustion air to the total incoming air flow according to [16]. The distance $\Delta x = 0$ mm corresponds to a swirl number S of 0, the maximum of Δx is 110 mm. The constants c_0, c_1 and c_2 consider the burner geometry whose profile can be seen in Figure 5.

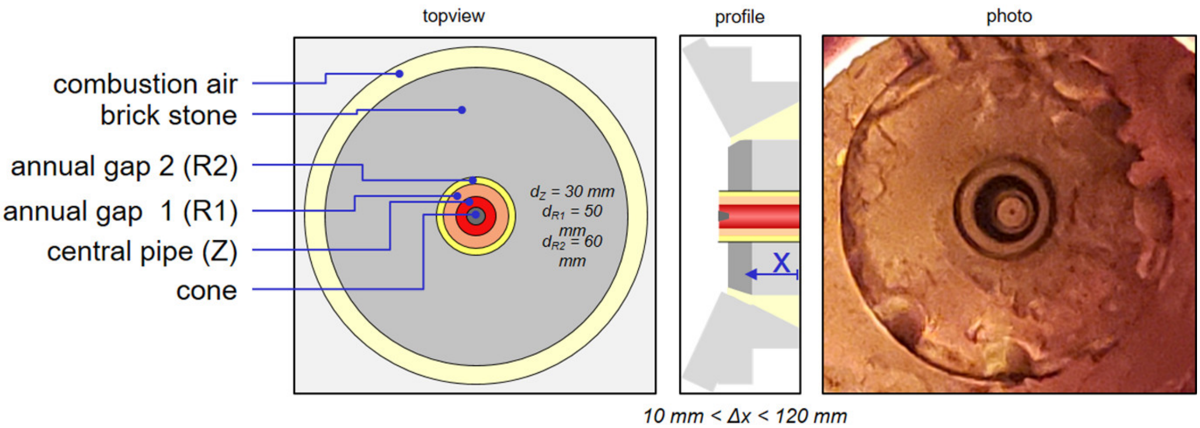

Figure 5. Scheme of coal burner.

$$S = f(\Delta x, Q) = c_0 \cdot \frac{c_1 + \Delta x \cdot c_2^2}{c_1 + \Delta x \cdot c_2} \cdot \Delta x \cdot Q^2 [\frac{1}{mm}], \text{ with } \Delta x \text{ in mm} \tag{1}$$

where Q describes the ratio of the swirled combustion air $\dot{V}_{VL}$ to the total incoming air volume flow from the burner according to Equation (2).

$$Q = \frac{\dot{V}_{VL}}{\sum_i^n \dot{V}_{i,\,L}} \tag{2}$$

For the description of the stoichiometry at the pulverized coal burner, the local air ratio λ_{loc} was calculated as a function of the coal mass flow, the minimum required air quantity l_{min} and the volume flows $\dot{V}_Z$, $\dot{V}_{R1}$, $\dot{V}_{R2}$ according to Equation (3).

$$\lambda_{loc} = \frac{\dot{V}_Z + \dot{V}_{R1} + \dot{V}_{R2}}{\dot{m}_{coal} \cdot l_{min}} \tag{3}$$

A further stoichiometric parameter is the burner air ratio λ_{burner} taking into account the combustion air flow entering the pulverized coal burner $\dot{V}_{VL}$ according to Equation (4).

$$\lambda_{burner} = \frac{\dot{V}_Z + \dot{V}_{R1} + \dot{V}_{R2} + \dot{V}_{VL}}{\dot{m}_{coal} \cdot l_{min}} \tag{4}$$

Before starting the experiments the plant has to be heated up to 900 °C in the post combustion chamber with natural gas and light oil in the rotary kiln burner. After reaching this temperature the supply of solid fuels is possible.

The coal which was used in the experiments is a hard coal with a net calorific value of 32 MJ/kg. The nitrogen content was analyzed to 1.6 wt.-% dry base, the medium particle diameter d_{p50} was about 50 µm.

3.2. Screening Campaign A

In the first screning campaign A, 48 model tests were carried out to derive the main influencing parameters on the oscillating combustion of solid pulverized fuels and classify them regarding their impact on the NO_x−concentration. With the model tests, the previously determined influences of the oscillation frequency and the height of the volume flows in R1 and R2, the volume flow of the combustion air from the rotary kiln, the quantity of coal supplied and the volume flow conveying coal were examined. The data from the screening campaign was used to create a multivariate regression model considering expert knowledge further called Expert model.

Table 1 lists the varied parameters for campaign A and their levels for the pulverized coal burner and the primary burner from the rotary kiln. The oscillation frequencies of the volume flows from annular gap R1 and R2 were examined in 5 levels, as a non-linear behavior of these variables was observed in the previous campaigns. Due to the technical limitation that the pinch valve does not close properly at frequencies higher than 3 Hz, the parameter space for the oscillation frequency moved between 0 and 3 Hz. From previous experiments it was known that oscillation has the greatest effect at high frequencies, which is why an additional level was defined at 2.67 Hz. It was also known, that the oscillation of the fuel flow leads to lower NO_X emissions. That's why in this campaign the effect of oscillating volume flows in R1 and R2 without fuel were investigated in 4 levels. Due to safety limitations at certain test settings, it was not possible during operation to set a minimum volume flow of the air in R2 to 50 $m_N{}^3/h$ ein. In these cases, the volume flow in R2 could only be reduced to 65 $m_N{}^3/h$ and led to a subsequent adaptation of the test plan.

Table 1. Varied parameters and their levels in campaign A.

System	Parameter	Level					Unit
		1	2	3	4	5	
Rotary kiln burner	Combustion air rotary kiln burner	1200	1400	-	-	-	$m_N{}^3/h$
Coal burner	Mass flow coal Z	70	90	-	-	-	kg/h
	Air flow Z	70	90	-	-	-	$m_N{}^3/h$
	Air flow R1	80	90	100	110	-	$m_N{}^3/h$
	Oscillating frequency R1	0	1	2	2.67	3	Hz
	Air flow R2	50 (65)	70	90	110	-	$m_N{}^3/h$
	Oscillating frequency R2	0	1	2	2.67	3	Hz

3.3. Expert Model

The multivariate regression model, which is based on expert knowledge, was used for the campaign to derive an equation for the NO_X concentration as a function of the main influencing parameters. As additional interactions, the two interactions between the oscillation frequency and the level of the oscillated air flow are included in the model. In addition to the varied parameters, the swirl number S according to Equation (1) and λ_{burner} according to Equation (4). were added to the model. Table 2 shows the influencing variables selected for the model and the corresponding abbreviation.

Table 2. Abbreviation of the influencing parameters.

Abbr.	Parameter	Unit
a	Oscillating frequency R1	Hz
b	Oscillating frequency R2	Hz
c	Mass flow coal Z	kg/h
d	Air flow Z	$m_N{}^3/h$
e	Air flow R1	$m_N{}^3/h$
f	Air flow R2	$m_N{}^3/h$
g	Combustion air rotary kiln burner	$m_N{}^3/h$
h	Swirl number S	-
i	λ_{burner}	-
ae	Interaction in R1	Hz $m_N{}^3/h$
bf	Interaction in R2	Hz $m_N{}^3/h$

To determine the significance of the respective explaining variables the F-value and the *p*-value were calculated for the created model [17,18]. Within the model, a significance level α of 5% was chosen, which is usual for engineering questions, found in literature [17]. Moreover, this did not result in a too sharp separation in the reduction of the model parameters in the expert model. Thus it could be ensured that a parameter remains tendentially in the model and in the follow-up the expert could evaluate its plausibility.

Following the described procedure the expert model was used to determine statistically relevant parameters which were further investigated in following campaign B.

3.4. Test Campaign B

The test campaign B was carried out to investigate the findings from the screening campaign A, where the oscillation of a high volume flow leaded to a NO_X reduction in the flue gas. In the first part the influence of the height of the oscillated air flow by keeping the air ratio λ_{burner}, the incoming air and coal flow in Z constant was investigated. The second part focussed on proving the reproducibility of the test settings from the past, where a high air flow was oscillated. The test settings of both investigations are described in the following paragraphs.

Table 3 shows the test settings for the investigation of the influence of the height of the oscillated volume flow in R2. No air from R1 was introduced into the system. With the parameter variation, the combustion air was shifted to the volume flow from R2 with simultaneous change of the swirl setting. Thus λ_{loc} changed, but λ_{burner} and the swirl number S remained constant. During tests 5 and 6, the combustion air could not be reduced below 313 $m_N{}^3/h$ due to the minimum air volume technically required for burner cooling. This resulted in a slight deviation from λ_{burner} of 1.12 instead of 1.11.

In order to determine a statement about the flame image and its influence on the nitrogen oxide concentration in this test series, the optical analysis of these tests was carried out with a software from IAI. The program determined the relative area of the flame relative to the total combustion chamber cross-section in percent. The relative stable flame fraction Area stable results from an averaging of image sequences consisting of 50 images. If the flame shapes of the images coincide, a relative flame stability of 100% is calculated, if the flame shapes differ strongly from each other, a relative flame stability of 0% is obtained. The exact calculation bases for the optical analysis as well as the made assumptions for the calculations are to be taken from [15].

The test settings for proving the reproducibility of the oscillation of a high air volume flow in R2 is shown in Table 4. The results are compared with two trials from 2018. In all trials the settings were kept constant for at least 60 min.

Table 3. Trial plan Campaign B investigation of the.

System	Parameter	Trial						Unit
		1	**2**	**3**	**4**	**5**	**6**	
Coal burner	Mass flow coal Z	70 (constant)						kg/h
	Air flow Z	80 (constant)						$m_N{}^3/h$
	Swirl number S	0.3 (constant)						
	λ_{loc}	0.28	0.28	0.48	0.48	0.51	0.51	-
	λ_{burner}	1.11	1.11	1.11	1.11	1.12	1.12	-
	Combustion air	420	420	321	321	313	313	$m_N{}^3/h$
	Air flow R2	65	65	164	164	180	180	$m_N{}^3/h$
	Oscillating frequency R2	0	2.67	0	2.67	0	2.67	Hz

Table 4. Trial plan Campaign B investigation of the reproducibility of previous test settings.

System	Parameter	Trial 1	Trial 2	Unit
Coal burner	Mass flow coal Z	57 (constant)		kg/h
	Air flow Z	70 (constant)		m_N^3/h
	Swirl number S	0.3		-
	λ_{loc}	0.6 (constant)		-
	λ_{burner}	1.22 (constant)		-
	Combustion air coal burner	258 (constant)		m_N^3/h
	Air flow R1	0 (constant)		m_N^3/h
	Oscillating frequency R2	0	2.67	Hz
	Air flow R2	180 (constant)		m_N^3/h

4. Results and Discussion

In the steady state mode of the plant (operating the burner in the rotary kiln with oil with a thermal power of 1 MW), the coal burner with a supporting flame with natural gas (1 m^3/h) and coal, the measurements start according the parameters shown in Tables 1, 3 and 5.

Table 5. Regression coefficients of the expert model.

Abbr.	Parameter	Unit	β_j/Unit		$\beta_{j,\,norm}$/mg/m_N^3
-	Interception y axis	-	8485.27	mg/m_N^3	598.66
a	Oscillating frequency R1	Hz	55.62	$mg/(Hz\ m_N^3)$	65.94
b	Oscillating frequency R2	Hz	26.08	$mg/(Hz\ m_N^3)$	64.06
c	Mass flow coal Z	kg/h	−43.43	$(h\ mg)/(kg\ m_N^3)$	−421.64
d	Air flow Z	m_N^3/h	2.5	$(h\ mg)/m_N^6$	12.39
e	Air flow R1	m_N^3/h	1.78	$(h\ mg)/m_N^6$	20.71
f	Air flow R2	m_N^3/h	1.92	$(h\ mg)/m_N^6$	44.10
g	Combustion air rotary kiln burner	m_N^3/h	0.04	$(h\ mg)/m_N^6$	3.98
h	Swirl number S	-	−2901.28	mg/m_N^3	−118.61
i	λ_{burner}	-	−3035.93	mg/m_N^3	−444.21
ae	Interaction in R1	$Hz\ m_N^3/h$	−0.61	$(h\ mg)/(Hz\ m_N^6)$	−66.35
bf	Interaction in R2	$Hz\ m_N^3/h$	−0.79	$(h\ mg)/(Hz\ m_N^6)$	−83.19

The temperature distribution in the post combustion chamber does not change much in between the different tests due to the thermal power of the rotary kiln, which is always higher than the thermal power of the coal burner (500 kW for 57 kg/h; 620 kW for 70 kg/h and 800 kW for 90 kg/h). The information given here refer to the test presented in Figure 13 with a coal mass flow about 57 kg/h. The values in brackets ("number") are the data for the case with oscillation. The gaseous species and the temperature in level 0 were measured only during the non-oscillating case. The CO and NO_x values are not normalized to 6 Vol.-% of oxygen.

Level 0: CO: 11.3 mg/m^3; NO_x: 79.7 mg/m^3; O_2 11.9 Vol.-%; CO_2: 6.7 Vol.-%; gas temperature: 842 °C.

Level 2: CO: 6.6 (7.8) mg/m^3; NO_x: 315 (182) mg/m^3; O_2 10.0 (9.8) Vol.-%; CO_2: 8.3 (8.4) Vol.-%; gas temperature: 949 (946) °C; total carbon (TC) in the fly ashes: 1.44 (2.37) wt.-%.

Only a slight decrease of oxygen in the oscillating mode could be recognized while the solid (TC) and gaseous burnout (CO) is quite good for both conditions in level 2.

4.1. Screening Campaign A

Figure 6 shows the dependence of the nitrogen oxide concentration on the oscillation frequency and the volume flow in R1 and R2 from the statistical test planning. The corresponding parameter levels are listed in Table 1. The high disperion of the measured data points is a result from the trial plan, since several parameters were changed simultaniously when the one on the x-axis was kept constant.

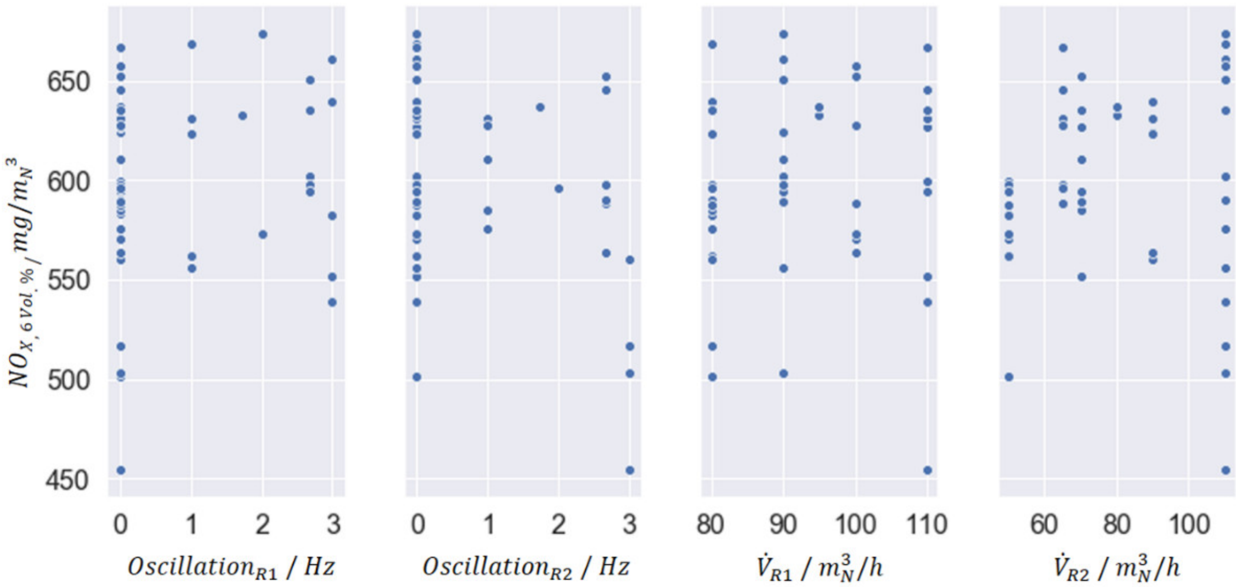

Figure 6. Results from oscillation and air flow in R1 and R2.

The results from the statistical test planning based on campaign A show no obvious dependencies towards the NO_x concentrations, so additional calculations with the EDI-Hive-model as well as the expert model are required to get more specific answers for the question: which are the main influencing parameters for the oscillation combustion?

For the analysis of the interaction between the oscillation frequencies and the oscillated air flow in Figure 7 a statistical calculation tool provided by EDI GmbH (See https://edi.gmbh/de/, last access date: 18 December 2020) which is based on a multivariate regression was used. Since the shown graphs are calculated by a model, there are no data points plotted in these graphs. The shape of the graphs is based on the linear or quadratic influence on the target variable. For further information about the tool see [12].

The dependency of nitrogen oxide from the oscillation is shown in the two upper diagrams and the dependency from the volume flow in the two lower diagrams. Now two scenarios are considered, the two diagrams on the left show the progression of the approximated nitrogen oxide concentration for a oscillation frequency of 0 Hz set by the cursor through the orange line. The two diagrams on the right show the NO_X concentration at a set oscillation frequency of 3 Hz.

For the diagram of the volume flow dependence in the left part of the figure, the nitrogen oxide concentration is increasing with the volume flow, whereas the dependence of the NO_X concentration at a oscillation of 3 Hz in the right part of the figure shows a clear reduction of the NO_X concentration with increasing volume flow. The point at which the effect of the volume flow dependence reverses in R2 could be determined with the model generator and is at a oscillation frequency of 0.8 Hz.

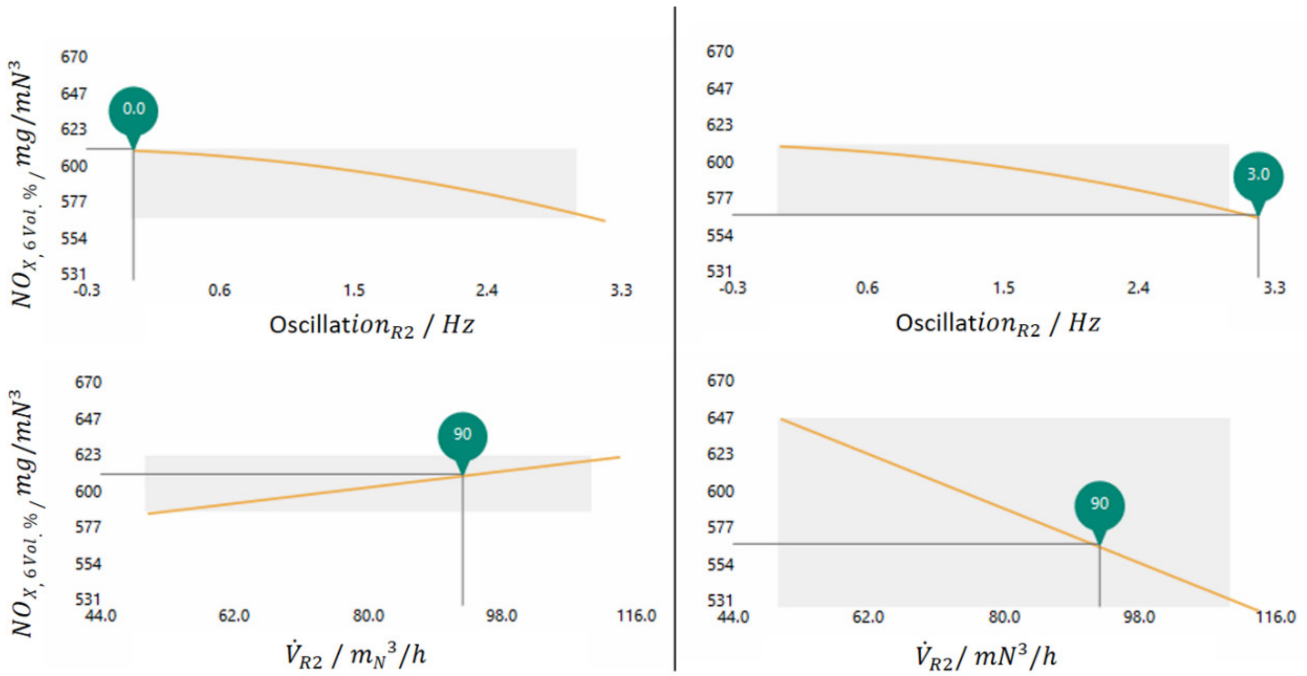

Figure 7. Results of the Interaction in R2.

The evaluation shows that without oscillation the increase in the volume flow leads to an increase in the NO_X concentration. At a oscillation of 3 Hz the increase of the volume flow leads to a significant reduction of the nitrogen oxide concentration.

In the case of a volume flow increase without oscillation, the provision of more oxygen leads to higher emission values of the nitrogen oxides. The NH_i species from Figure 1 react directly to NO under these conditions and the reaction to molecular nitrogen is inhibited here. In the case of an increase in volume flow at a oscillation of 3 Hz, a similar result should also be obtained. But apparently a more concise oscillating profile of the local stoichiometry can be generated by oscillation of higher volume flows. This allows the reaction of the formed NO to molecular nitrogen in the time gaps of the interruption of the volume flow. It should be mentioned that a higher volumetric flow has a larger impulse than a lower volumetric flow due to the equation $\dot{I} = \dot{m} * v$. The higher the impulse of air (by an increase of the volumetric flow and corresponding to a higher velocity) the better the interruption of the coal mass flow is possible. Not to the extent that the oscillation of the fuel flow would cause it, but to the extent that favourable reaction conditions prevail for the reduction of nitrogen oxide emissions.

4.2. Expert Model

A second tool was applied based on the data from campaign A including the knowledge of experts. The expert model includes all influencing parameters that were specifically modified in the statistical design of the experiment, as well as the air ratio λ_{burner}, the swirl number S and the interaction of volume flow and oscillation in annular gap R1 and annular gap R2. The explanatory variables and their abbreviations for the following considerations are shown in Table 2. First, the multivariate regression model with eleven descriptive variables is considered. The approximated NO_X emission data, the calculated predictive accuracy of the model for training and test data and the F-value of the model are shown in Figure 8. For the tabular F-value with the degrees of freedom $f_1 = 11$ and $f_2 = 36$, an F-value of $F_{11,36,0.95} = 2.07$ is obtained. The model has a higher F-value and therefore contains variables describing the problem more significantly.

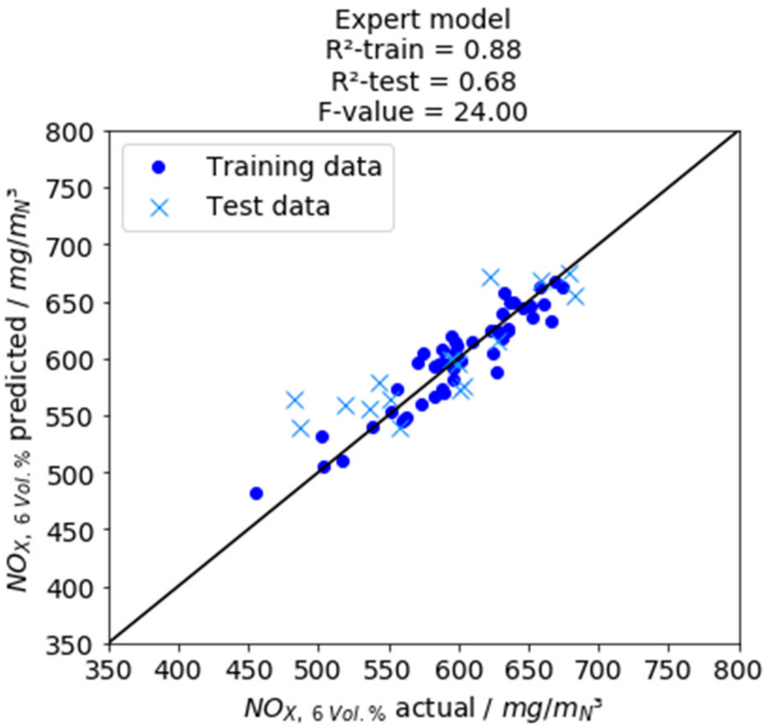

Figure 8. Results of the Expert model.

Table 5 shows the regression coefficients of the individual influencing variables for the regular and the standardized training data. With the non-standardized coefficients, the set parameter variations can be used directly to calculate the nitrogen oxide concentration and the units of the coefficients reduce to the unit of the target value to be approximated, nitrogen oxide concentration in the flue gas in $mg/m_N{}^3$. The unit and the value of the coefficients are given in the table below. For the non-standardized regression coefficients, no comparison is possible regarding the weighting due to the different orders of magnitude of the parameters. For this reason, the standardized, dimensionless beta coefficients are also shown. With them the coefficients can be compared regarding their weighting because of the previously carried out standardization of the data.

The corresponding model based on the standardized coefficients of the linear terms a_{norm} to g_{norm} and the interaction terms ae_{norm} and bf_{norm} is given by the following pattern.

$$\hat{y} = [\beta_{0,norm} + \beta_{1,norm} \cdot a_{norm} + \beta_{2,norm} \cdot ae_{norm} + \beta_{3,norm} \cdot b_{norm} + \beta_{4,norm} \cdot bf_{norm} + \beta_{5,norm} \cdot c_{norm} + \beta_{6,norm} \cdot d_{norm} + \beta_{7,norm} \cdot e_{norm} + \beta_{8,norm} \cdot f_{norm} + \beta_{9,norm} \cdot g_{norm} + \beta_{10,norm} \cdot h_{norm} + \beta_{11,norm} \cdot i_{norm}] \left[\frac{mg}{m_N 3}\right] \quad (5)$$

Inserting the standardized regression coefficients from Table 5 which are calculated with the least square method and fitted by the training data set for the multivariate regression model the following equation concludes. This equation is applied to the test data for showing the prediction accuracy of the model (see Figure 8).

$$\hat{y} = [598.66 + 65.94 \cdot a_{norm} - 66.35 \cdot ae_{norm} + 64.06 \cdot b_{norm} - 83.19 \cdot bf_{norm} - 421.64 \cdot c_{norm} + 12.39 \cdot d_{norm} + 20.71 \cdot e_{norm} + 44.1 \cdot f_{norm} + 3.98 \cdot g_{norm} - 118.61 \cdot h_{norm} - 444.21 \cdot i_{norm}] \left[\frac{mg}{m_N 3}\right] \quad (6)$$

The largest absolute values for the standardized coefficients βi are the parameters c and i, which both result from the coal mass flow and are thus directly involved in increasing the fuel nitrogen content. The third largest value results for h, the swirl number. All three quantities mentioned have a negative sign, i.e., the approximated target quantity decreases with an increase of the parameter. This is of interest for the coal mass flow, as its increase will introduce more nitrogen into the system and NO_X emissions should increase accordingly. The effect of the stoichiometric air ratio can be explained by the anti-proportionality to the coal mass flow (see Equation (3)). Similarly, the linear influences of the oscillation frequency a and b have a positive sign and the interaction terms of the two variables ae and bf have a negative sign. Thus, according to Equation (5), the effects are contrary related to the target quantity. Such dependencies can be used to map the interactions for the oscillation frequency and the volume flow in R2 from Figure 7. with a statistical model. Furthermore, the model assumes that the parameters are varied within the investigated limits of the design space.

Since Equation (5) is a mathematically determined model equation for the investigated parameter space from Table 1, it is only valid for this particular parameter space. Thus, the relations behind the examined regression coefficients follow the adjusted parameter combinations, whereby by using the parameters the target quantity can be predicted with an accuracy of 88%.

The results for the examined parameters of the expert model are shown in Figure 9. Here, the red line represents the significance level $\alpha = 0.05$, which determines if a parameter is significant or not. p-values greater than α are non-significant and p-values less than α point to significant model parameters, p-values less than $\alpha = 0.001$ are strongly significant.

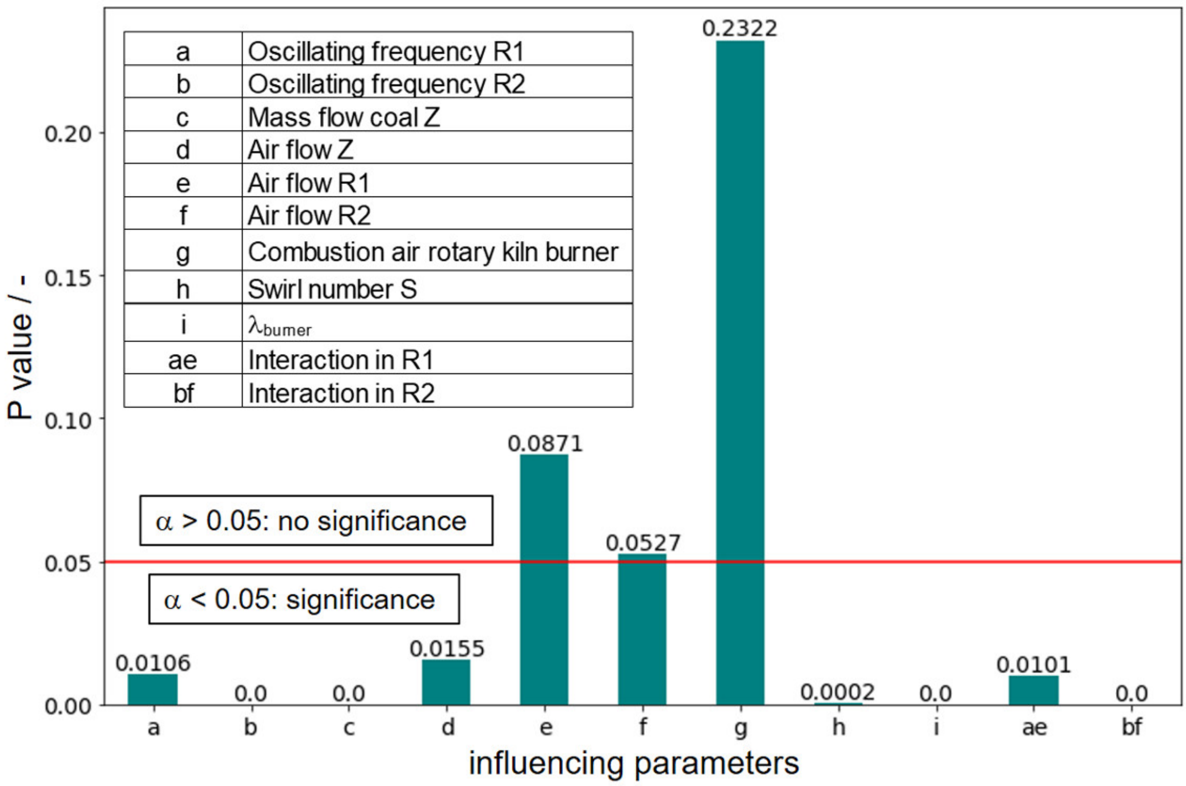

Figure 9. p-Value of influencing variables.

The evaluation clearly shows that the influencing parameters e volume flow in R1, f volume flow in R2 and g combustion air from the rotary kiln burner have a higher p value than the selected significance level α of 0.05. According to the evaluation carried out, the parameters mentioned have no explanatory content for the model formed. The parameters b oscillation frequency in R2, *c* coal mass flow in Z, *h* swirl number, *i* air ratio λ_{burner} and bf interaction of oscillation frequency and volume flow in R2 are considered strongly significant for the model formed. Furthermore, it is shown that the interactions ae and bf of the oscillation frequency with the respective volume flow from R1 or R2 are to be classified as significant parameters, whereas the linear influences of the volume flows in R1 and R2 have no significant effect on the model and these only contribute to the model in the interaction with the respective oscillation frequency. As obvious significant effects with a *p*-value of 0, *b* oscillation frequency in R2, *c* carbon mass flow in Z, *i* air ratio λ_{burner} and bf interaction of oscillation and volume flow in R2 can be identified. The high significance of the two parameters coal mass flow and λ_{burner} regarding the NO_X emission is due to the influence of the increased fuel input and corresponds to the findings from the analysis of the regression coefficients from Equation (5). The increase in the fuel input is directly reflected in the coal mass flow; the air ratio λ takes into account the incoming air flows and is anti-proportional to the coal mass flow according to Equation (4). The oscillation frequency in R2 and the interaction of the oscillation frequency with the volume flow in R2 reflect the investigated effect of air oscillation. Obviously, it is possible to exert a significant effect on NO_X emissions by oscillating the air volume flow. The influence of the swirl number also has a significant effect in the evaluated data. This is consistent with the observation in Equation (5), where the swirl number has the third largest standardized regression coefficient.

4.3. Test Campaign B

Based on the findings of the statistical analysis by the expert model, that the oscillation of a high-volume flow in R2 leads to a lower nitrogen oxide concentration, the oscillating addition of a high-volume flow of 180 m_N^3/h from the annular gap R2 was investigated.

In Figure 10 the dependence of NO_X emissions on the level of the oscillated volume flow in R2 and the oscillation frequency at a constant swirl and a constant air ratio at the burner λ_{burner} is shown. The reference value is the NO_X concentration at the respective non-oscillated state. The further parameter settings of the constant incoming flows can be seen on the side of the figure and in Table 3. The nitrogen oxide emissions at a volume flow of 65 m_N^3/h increase slightly due to the oscillation of 2.67 Hz. By increasing the volume flow entering the annular gap R2 to 164 m_N^3/h, the nitrogen oxide emissions in the non-oscillated state are significantly increased. On the other hand, the oscillation of this volume flow shows a significant reduction in nitrogen oxide emissions. For a volume flow of 180 m_N^3/h, the nitrogen oxide emissions in the non-oscillated state are somewhat higher than for a volume flow of 164 m_N^3/h, but a similar behavior in NO_X reduction can be observed here regarding the oscillated state.

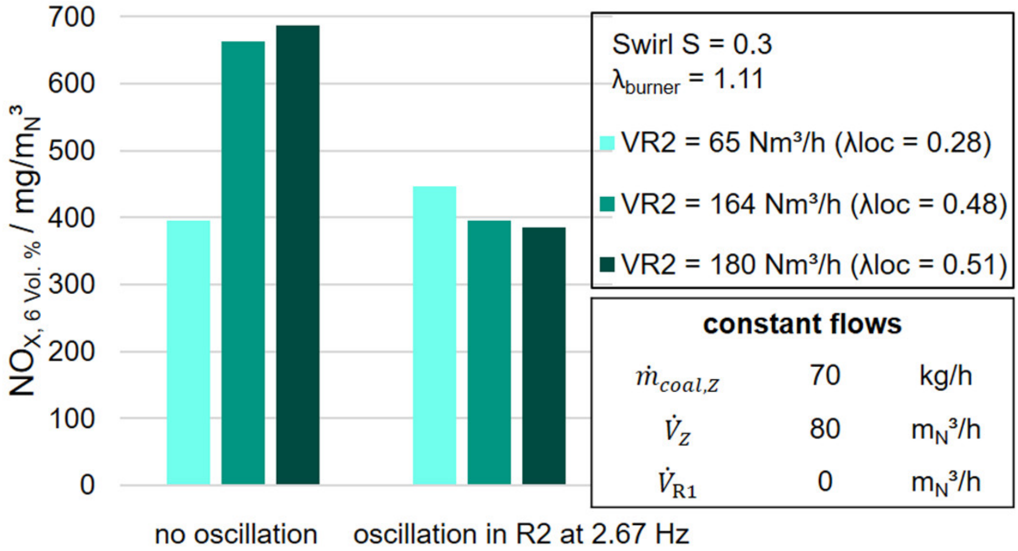

Figure 10. Investigation of the oscillated air flow.

As a result of the increased oxygen supply in the flame associated with the increase in the volume flow in R2, the nitrogen oxide emissions in the tests increase to 164 and 180 m_N^3/h in the oscillated state. Reference [19] emphasizes the careful coordination of the different flow fields of the fuel-related primary air streams and the fuel-remote secondary air streams in a pulverized coal burner. Accordingly, the increase in the nitrogen oxide concentration in the flue gas during the shift of the combustion air into the annular gap R2 is explained by a worse preheating of the coal mass flow. This leads to an unfavorable combustion behavior of the coal dust. At the same time, a swirl number of 0.3 is not enough to create a recirculation area which would guarantee better preheating of the fuel flow.

According to the literature of [2,10,20], the backflow area for Type I and II flames only develops at a critical swirl number of $S_{theo} \approx 0.5$. However, at the prevailing temperatures the formation mechanisms are limited to the fuel N mechanism. According to Figure 1, an increase in oxygen concentration leads to the preferred reaction path from HCN via NH_i to NO. Since there are no reducing conditions at an increased oxygen supply in the flame, the reaction of NH_i with already formed NO to N_2 is inhibited. Although $\lambda_{loc} < 1$ applies to the local stoichiometries of all investigated test settings, λ_{loc} is significantly larger at the higher volume flows in R2. If one now compares the local stoichiometry with the TFN curve shown in Figure 2, the NO_X concentration increases with increasing air ratio. In addition, the exhaust gas mixture contains more HCN and NH_3 at the local air ratios of 0.28 to 0.51. The concentration of HCN and NH_3 in the exhaust gas mixture increases with the local air ratios of 0.28 to 0.51. For both N-species, the residence time in the post-combustion chamber is enough to react to NO_X at a total air ratio in the post-combustion chamber of 1.1 to 1.4 up to the measuring point of the nitrogen oxide concentration. From consideration of the oscillation effect of the two high oscillated volume flows of 164 and 180 ${m_N}^3$/h at 2.67 Hz, the nitrogen oxide emission can be reduced despite the high local air ratios. The interruption of the volume flow thus leads to alternating reducing and oxidizing conditions in the flame, resulting in a NO_X reduction. Considering Figure 1, more NO is produced under oxidizing conditions. Under the reducing conditions the NO is reduced to N_2. In parallel, NO molecules can be returned to the reaction path of the fuel-N mechanism via the NO-Recycle mechanism. These two mechanisms explain the lower nitrogen oxide emissions. In further considerations, it would be useful to measure the local concentrations of the N-species in order to obtain more conclusions about the formation mechanism under oscillating conditions. Related to the reference value of the nitrogen oxide concentration at 65 ${m_N}^3$/h and without oscillating addition of air, no reduction of the nitrogen oxide emissions can be observed at the selected test settings, but the results show that the NO_X formation mechanisms can be substantially reduced by oscillation.

A more detailed analysis of the flame pattern for the phenomenon of increasing the nitrogen oxide concentration in the flue gas by shifting the air volume flow from the combustion air into the annular gap R2 with oscillation-free process control is explained in more detail below. The optical observation of the flames of the non-oscillated test settings is shown the photo series of 0. 5, 10, 15 and 30 s of the test time in Figure 11. With a volume flow from R2 of 65 ${m^3}_N$/h, a longer flame and a more uniform distribution of the bright flame range over the entire length of the flame result. The higher volume flow in the tangentially introduced combustion air leads to a more stable, elongated flame pattern.

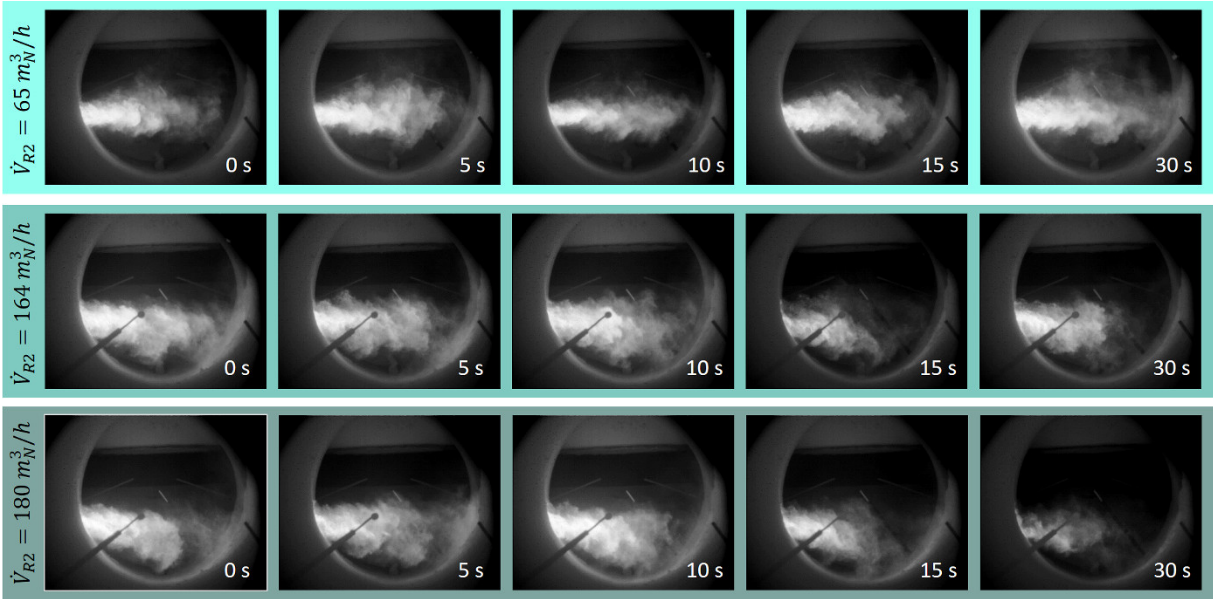

Figure 11. Photo series of a non-oscillating flame at different air flows in R2.

The two lower photo series at a volume flow in R2 of 164 and 180 m_N^3/h show a higher flame brightness in the area near the flame root than in the burnout area of the flame. Here the volume flow of combustion air is significantly reduced (cf. Table 3), which leads to a shorter and bushier flame.

The evaluations with the IAI program from Figure 12 confirm this fact for the shorter and bushier flame shapes observed in Figure 11 for the high-volume flows in R2 of 164 and 180 $m^3{}_N$/h, since both the relative flame fraction area and the relative stable flame fraction area stable are significantly lower for the two flames.

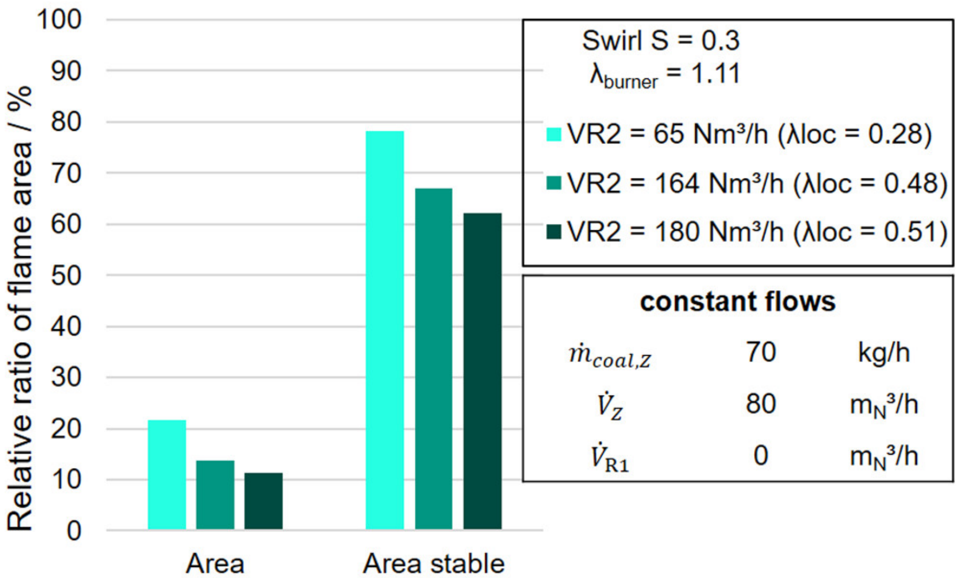

Figure 12. Image analysis of relative flame area.

Consequently, for the longer flame at a non-oscillated volume flow in R2 of 65 $m^3{}_N$/h, a more uniform burnout of the combustion gases along the flame length is possible, which is associated with a lower nitrogen oxide concentration in the flue gas (cf. Figure 3). At Definition there are no Type I/II flames due to the too low swirl number, which means that the results cannot be compared with the previous studies of [10]. However, the influence of the flame shape on the nitrogen oxide concentration in the flue gas can be clarified from the optical evaluations discussed above.

The second part of the test campaign B focused on the reproducibility of specific test settings when a high-volume flow in R2 was oscillated. The measured concentrations of nitrogen oxide, oxygen and CO in the experiments carried out are shown in Figure 13 over the test period of 60 min per setting. The constant currents are shown sideways in the figure.

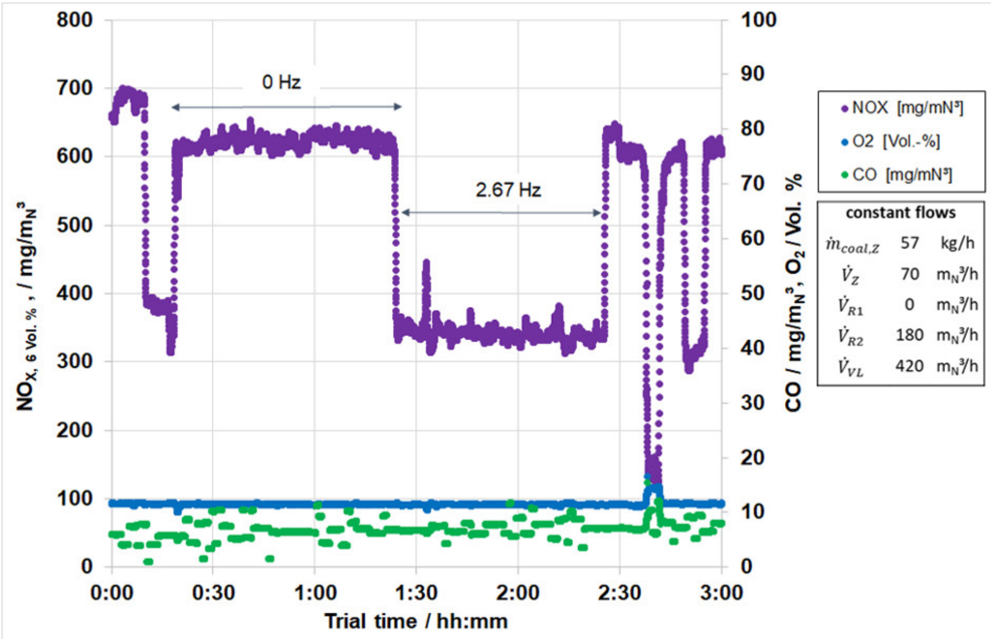

Figure 13. Oscillation of a high-volume air flow in R2 at 2.67 Hz.

The reduction in NO_X emissions due to oscillation of the volume flow in R2 at 2.67 Hz is clearly visible. For the two experiments a constant nitrogen oxide concentration of 620 $mg/m_N{}^3$ for the oscillation-free and 350 $mg/m_N{}^3$ N for the oscillating process control is achieved over the 60-min test period. Accordingly, the nitrogen oxides in the flue gas are reduced by 45%.

This represents the best measurement result of the tests carried out in this paper regarding nitrogen oxide reduction by oscillation of the air volume flows. The oxygen and carbon monoxide concentrations remain constant over the entire period shown. This indicates a stable combustion process during the experiments.

In order to prove the reproducibility of the results from Figure 13, the measured NO_X concentrations of the trials from 2018 are compared to the results from 2019. The settings of the parameters in Figure 14 are according to Table 4.

The results from Figure 14 underline the previously discussed findings and show that the reproducibility of the trial is high, leading to an average reduction of the NO_X concentration by −43%.

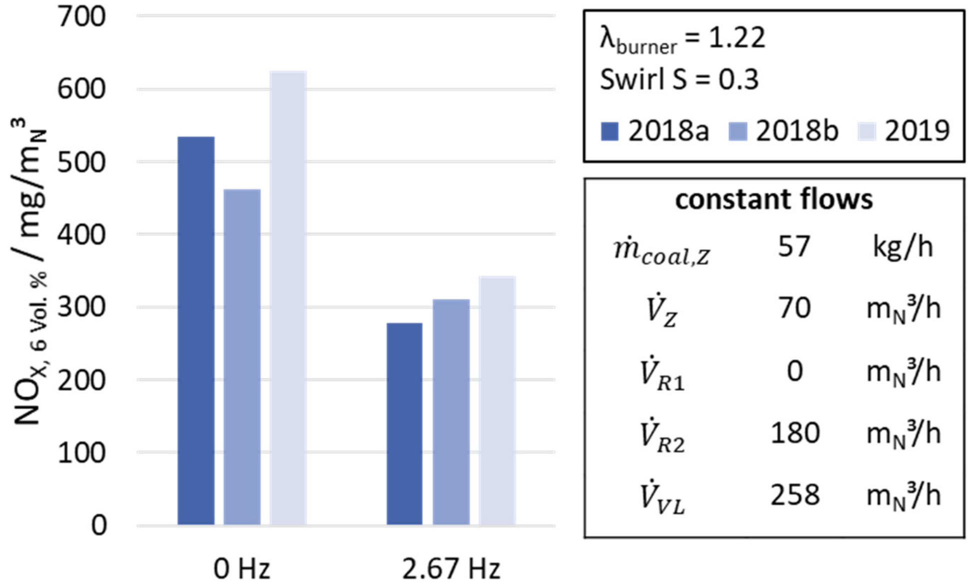

Figure 14. Campaign B reproducibility of test settings.

5. Conclusions

The focus in this paper was to identify influencing paramters on an oscillating combustion process of coal. The test setup contained a rotary kiln burner to ensure the minimum heat load for the process which was kept constant over the trial campaigns. In the after burning chamber the coal burner was used to examine the reduction potential of oscillating the incoming air flows in the inner and outer annular gaps in order to reduce the NO_X emissions. The campaign consisted of a screening trial where the data was used to identify relevant influencing parameters using a statistical model. Based on these results a second trial was initiated to further investigate the optimal test settings and the reproducability of the test setup.

The preparation of a statistical test plan was used to determine influencing parameters of an oscillating coal dust combustion without interrupting the fuel mass flow. Due to the oscillating addition of the air volume flows, the secondary optima and obvious influences on the nitrogen oxide concentration in the flue gas could be identified. Especially for oscillation at frequencies of 3 Hz, the NO_X concentration of the investigated test area could be reduced by up to 25%.

However, the exact distribution of the air volume flows and the distance between the addition location and the fuel mass flow played an important role. The coal was added at the burner mouth via the central inlet pipe and the oscillation of the air volume flows took place in the concentrically arranged annular gaps. The results showed a high influence for the oscillation of the outer annular gap (R2) and hardly an effect for the oscillation of the inner annular gap (R1). This finding can be explained by the dependence on the residence time of the primary combustion flue gas before the secondary air is mixed into the flue gas. Similar findings were made in a coal flame described by [21]. The oscillation of the locally closer oscillated volume flow from the inner annular gap (R1) resulted in a maximum nitrogen oxide reduction of 11%, whereas the oscillation of the air volume flow from the outer annular gap (R2) further away from the coal mass flow resulted in a maximum nitrogen oxide reduction of 25%. In addition, the results of the different test settings could be used to determine a relationship between the height of the oscillated volume flow and the oscillation frequency. Accordingly, the increase of the oscillated volume flow at a high oscillation frequency of 3 Hz led to a reduction of the nitrogen oxide concentration in the flue gas, whereas the increase of the oscillated air volume flow at a low oscillation frequency of less than 0.8 Hz led to an increase of the nitrogen oxide concentration in the flue gas.

Based on the findings from the screening trials further experiments focussed on oscillating a high volume flow in the outer annular gap (R2). Simultaniously, the reproducibility of the test settings was investigated. During the reproduction experiments, 180 $m_N{}^3$/h of air was added from the outer annular gap R2 at a oscillating frequency of 2.67 Hz. Here, the nitrogen oxide concentration in the flue gas could be reduced by 45% from 624 mg/$m^3{}_N$ at 0 Hz to 342 mg/$m^3{}_N$ at 2.67 Hz in relation to 6% by volume oxygen. This result was in line with the above-mentioned finding that the level of the oscillated air volume flow is essential for nitrogen oxide reduction in the flue gas. The results could be reproduced and lead to an average reduction potential of 43%.

For modelling, the data was divided into training and test data. The training data included all attempts of statistical test planning in order to be able to reproduce the investigated influencing parameters in the model. The test data consisted of a random variation of the investigated parameters and only served to validate the trained model.

With the statistical planning of the trial and the application of statistical analysis algorithms on the results, it was possible to identify significant influences such as -the oscillation frequency in the inner and outer annular gaps (R1 and R2), as well as its interaction with the respective volume flows, the coal mass flow, the air ratio λ, and the swirl number S.

It could be summarized that applying statistical algorithms and expert knowledge lead to a quick and reliable prediction result, avoiding a costly CFD simulation. But for fully understanding the thermo chemical phenomenons more valuable measuring instruments and a simulation based on these measurings are needed.

Author Contributions: Conceptualization, N.J., J.M. and H.-J.G.; experimental and data curation, M.H., J.M. and H.-J.G.; software, J.M.; expert model development: N.J. within the frame of his master thesis at KIT; validation, J.M. and H.-J.G.; writing—original draft, N.J. and H.-J.G.; writing—review and editing, K.A., D.S., H.S., J.M., P.W., M.V., H.B.K. and H.-J.G. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Helmholtz Association (HGF) within the German federal research program "Energy efficiency, materials and resources" (internal grant number 34.11.01).

Conflicts of Interest: The authors declare no conflict of interest. Nicklas Jolibois did his Masterthesis at KIT until October 2019 prior to his employment at BASF, starting in January 2020. The company had no role in the design, collection, analyses, or interpretation of data, the writing of the manuscript, or the decision to publish the results.

References

1. Inshakov, O.V.; Bogachkova, L.Y.; Popkova, E.G. *Statistics Global Energy Data at Your Fingertips*; Springer: Cham, Switzerland, 2019; pp. 135–148.
2. Joos, F. *Technische Verbrennung: Verbrennungstechnik, Verbrennungsmodellierung, Emissionen*; Springer: Berlin/Heidelberg, Germany, 2006.
3. Gehrmann, H.-J.; Baris, D.; Stapf, D.; Seifert, H.; Zorbach, I. Stickoxidminderung durch oszillatorische Verbrennung bei der Rostfeuerung. In *Proceedings of the Energie aus Abfall*; Institut für Technische Chemie (ITC): Berlin, Germany, 2018; pp. 243–253.
4. Cho, H.; Cho, K.W.; Kim, H.J. NOx Emission Characteristics in Radiant Tube Burner with Oscillating Combustion Technology. *Trans. Korean Soc. Mech. Eng. B* **2008**, *32*, 100–106. [CrossRef]
5. Ricken, V. Untersuchungen zur Stickoxidemission von Gasturbinenbrennkammern mit Luftgestufter Verbrennungsführung. Doctoral Dissertation, Universität Karlsruhe, Karlsruhe, Germany, 2006.
6. Marro, R. Einfluss der Verbrennungsbedingungen auf NOx-Emissionen und Belagsbildung bei der Staubfeuerung aufbereiteter biogener Brennstoffe. Ph.D. Thesis, Technische Universität München, Munich, Germany, 2018.
7. Koger, S. *Reaktionskinetische Untersuchungen zur Umwandlung stickstoffhaltiger Gaskomponenten unter Bedingungen der Abfallverbrennung*; KIT Scientific Publishing: Karslruhe, Germany, 2010.
8. Stapf, D. *Experimentell Basierte Weiterentwicklung von Berechnungsmodellen der NOx-Emission Technischer Verbrennungssysteme*; VDI-Verl.: Düsseldorf, Germany, 1998.
9. Miller, J.A.; Bowman, C.T. Mechanism and modeling of nitrogen chemistry in combustion. *Prog. Energy Combust. Sci.* **1989**, *15*, 287–338. [CrossRef]
10. Jansohn, P. Bildung und Abbau N-haltiger Verbindungen, Insbesondere von HCN, NH\Textsubscript 3 und NO, in Turbulenten Diffusionsflammen. Ph.D. Thesis, Universität Karlsruhe (TH), Karlsruhe, Germany, 1991.

11. Eberius, H.; Just, T.; Kelm, S. *NOx-Schadstoffbildung aus Gebundenem Stickstoff in Propan/LuftFlammen, Vergleich mit Kinetischen Modellen*; VDI-Berichte: Düsseldorf, Germany, 1983; Volume 498, pp. 183–192.
12. Freudenmann, T.; Gehrmann, H.J.; El-Haji, M.; Stapf, D.; Gmbh, F.E. *Hybridmodelle zur Effizienten Regelung, Optimierung und Überwachung von Thermo-Chemischen Prozessen und Anlagen am Beispiel der Oszillierenden Verbrennung*; 29. Deutscher Flammentag: Bochum, Germany, 2019.
13. Scherello, A.; Konold, U.; Flamme, M.; Kremer, H. *Experimentelle Untersuchung zum Einfluss der Oszillierenden Verbrennung auf die Wärmeübertragung und die Stickoxidemissionen vom Gasflammen*; Gaswärme-Institut Essen: Essen, Germany, 2002.
14. Wagner, J.C. *NOx Emission Reduction by Oscillating Combustion*; U.S. Department of Energy, Ed.; Gas Technology Institute: Des Plaines, IL, USA, 2004. [CrossRef]
15. Matthes, J.; Waibel, P.; Vogelbacher, M.; Gehrmann, H.J.; Keller, H.B. A new camera-based method for measuring the flame stability of non-oscillating and oscillating combustions. *Exp. Therm. Fluid Sci.* **2019**, *105*, 27–34. [CrossRef]
16. Schmid, C. Drallbrenner-Simulation durch Starrkörperwirbel-Strömungen unter Einbeziehung von drallfreier Primärluft und Verbrennung. Ph.D. Thesis, Universität Karlsruhe (TH), Karlsruhe, Germany, 1991.
17. Siebertz, K. *Statistische Versuchsplanung; Design of Experiments (DoE)*; Springer: Berlin/Heidelberg, Germany, 2017.
18. Kleppmann, W. *Taschenbuch Versuchsplanung; Produkte und Prozesse Optimieren*; Hanser Verlag: Munich, Germany, 2011.
19. Günther, R. *Verbrennung und Feuerungen*; Springer: Berlin/Heidelberg, Germany; New York, NY, USA, 1974.
20. Leuckel, W. Schadstoffe aus Verbrennungsprozessen. *Gas Wärme Int.* **1971**, 18–24.
21. Malek, C. *Zur Bildung von Stickstoffoxid bei einer Staubfeuerung unter gleichzeitiger Berücksichtigung des Ausbrandes*; Cuvillier Verlag: Göttingen, Germany, 1993.