

# Experimental investigation of N<sub>2</sub>O formation in selective non-catalytic NO<sub>x</sub> reduction processes performed in stoker boiler

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Stoker fired boiler plants are common throughout Eastern Europe. Increasingly strict emission standards will require application of secondary NO<sub>x</sub> abatement systems on such boilers. Yet operation of such systems, in addition to reducing NO<sub>x</sub> emissions, may also lead to emission of undesirable substances, for example N<sub>2</sub>O. This paper presents results of experimental tests concerning N<sub>2</sub>O formation in the selective non-catalytic NO<sub>x</sub> emission reduction process (SNCR) in a stoker boiler (WR 25 type). Obtained results lead to an unambiguous conclusion that there is a dependency between the NO<sub>x</sub> and N<sub>2</sub>O concentrations in the exhaust gas when SNCR process is carried out in a coal-fired stoker boiler. Fulfilling new emission standards in the analysed equipment will require 40–50% reduction of NO<sub>x</sub> concentration. It should be expected that in such a case the N<sub>2</sub>O emission will be approximately 55–60 mg/m<sup>3</sup>, with the NO<sub>x</sub> to N<sub>2</sub>O conversion factor of about 40%.

**Keywords:** selective non-catalytic reduction process, nitrous oxide emissions, stoker boiler.

## INTRODUCTION

Emission standards for the fuel combustion plants in Poland are gradually tightened. The first legislation regulating those matters directly were adopted in 1990. Start of negotiations of the European Union Accession Treaty marked the start of process of closing the gap between national regulations and EU emission standards. In case of hard coal combustion, direct limits concerned emission of sulphur oxides, nitrogen oxides (NO<sub>x</sub>, aggregated emission of NO and NO<sub>2</sub> converted to NO<sub>2</sub> equivalent) and particulate matter 1. From 2016 on, the emission standards within the EU are mandatorily based on the Directive 2010/75/EU – the Industrial Emissions Directive (IED) 2. Transposition of the IED into Polish regulations has been effected by a regulation of the Minister of Environment issued in 2014<sup>3</sup>.

Municipal heating plants equipped with stoker boilers proved to be a group of businesses particularly strongly affected by the new tighter standards, which in this case are much stricter than before<sup>4</sup>. For example, for a WR 25 boiler, the current NO<sub>x</sub> emission limit of 400 mg/m<sup>3</sup> in new regulations may get reduced to even 200 mg/m<sup>3</sup>. Technical capabilities of primary reduction of NO<sub>x</sub> emissions from stoker boilers fired with hard coal are exhausted somewhat above the threshold of 300 mg/m<sup>3</sup>. Due to this fact it is becoming necessary to employ secondary techniques. The most common secondary NO<sub>x</sub> abatement methods are based on reactions between the nitrogen oxides and ammonia radicals NH<sub>2</sub> supplied by reducing agents, mainly ammonia and urea (SNCR<sup>5</sup> and SCR<sup>6</sup> technologies).

Actual reaction mechanism for both technologies is quite complex – as many as 31 compounds and 173 relevant reactions have been identified as occurring during the NO<sub>x</sub> reduction process<sup>7</sup>. Starting points for reactions of different reducing agents are different for obvious reasons, yet further course of the chemical process is common.

In case of combustion equipment of smaller outputs (stoker boiler), thanks to less stringent applicable emission standards, NO<sub>x</sub> reduction of about 50% is typically

sufficient. For this type of boilers SNCR is considered to be the Best Available Technique (BAT)<sup>8</sup>.

Ammonia is a toxic flammable gas, which creates an explosive mixture with the air. All operations involving gaseous ammonia or ammonia water requires special safety measures. On the other hand, urea is not considered to be hazardous. It is not flammable, explosive or harmful for health. Urea transport and storage is much easier and safer than in case of ammonia. Due to operational reasons, using urea as the reagent is therefore more favourable.

However, a distinctive feature of the SNCR process based on urea is formation of reaction by-products. These include nitrous oxide (N<sub>2</sub>O). This gas, along with carbon dioxide and methane, is one of the three main gases which affect the greenhouse effect. The GWP100 (100-year global warming potential) for the nitrous oxide is 296. Moreover, in the upper atmosphere layers N<sub>2</sub>O may be transformed into other nitrogen oxides reacting with ozone. Its excessive emission may be therefore a considerable environmental concern.

Natural presence of nitrous oxide in the atmosphere results from Earth's natural nitrogen circulation. It is estimated that 40% of total emission is attributable to human activities in the area of agriculture (use of nitrogen-based fertilisers), energy industry (fuel combustion), industrial processes (chemistry), transport and waste disposal<sup>9</sup>. In Polish conditions, information on greenhouse gas emissions may be sourced from the national inventory report by the National Centre for Emissions Management<sup>10</sup>. The report includes data on following greenhouse gases: CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub>, and HFC. In May 2014 KOBiZE updated data on emission and absorption of greenhouse gases for 2012. Total greenhouse gas emission for 2012 was 399.267.97 Gg of CO<sub>2</sub> equivalent (excluding the sector of land utilisation, change of land utilisation and forestry). Emission of main greenhouse gases from specific sectors is presented in the Table 1<sup>10</sup>.

**Table 1.** Emission of main greenhouse gases in Poland in 2012 (data by KOBiZE)

???????????????	CO <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub> O
	[Gg]	[Gg]	[Gg]
National gross emission (excluding sector 5)	320.861.67	1.953.93	95.45
National net emission (including sector 5)	286.189.28	2.062.15	97.21
1. Energy	302.127.65	734.85	6.77
A. Fuel combustion	298.403.80	149.46	6.77
B. Volatile emission from fuels	3.723.85	585.38	0.00
2. Industrial processes	17.819.61	14.47	3.39
3. Utilisation of solvents and other products	635.67	–	0.40
4. Agriculture	–	545.79	81.27
5. Land utilisation, changes of land utilisation, forestry	–34.672.39	108.21	1.76
6. Waste	278.74	658.83	3.63

The energy industry was a dominating source of carbon dioxide emissions. The N<sub>2</sub>O emissions were to the largest extent attributable to agriculture.

Results of emission stocktaking expressed in CO<sub>2</sub> equivalent units using global warming potential indicators reveal that carbon dioxide is responsible for about 80% of total greenhouse gas emissions in Poland, share of methane is six times lower (about 13%), and nitrous oxide is responsible for about 7%<sup>11</sup>.

Due to the low share of nitrous oxide emitted by the industry in total greenhouse gas emissions, European strategies concerning emission problems (EU ETS Directive, IED) do not take N<sub>2</sub>O emissions into account. New sources of emission of this gas, including NO<sub>x</sub> abatement technologies, may change those proportions.

The N<sub>2</sub>O emission attributable to the SNCR process has been investigated by other authors in<sup>12, 13</sup>. One of the earliest publications on N<sub>2</sub>O emission from the NO<sub>x</sub> abatement process was<sup>14</sup>. It presented results of tests involving decomposition and oxidation of urea used as a reagent in a flow reactor. Then authors of the publication<sup>15</sup> have carried out some simulation calculations aimed at investigating influence of SNCR process parameters on related N<sub>2</sub>O emission. Basing on the simulations it was concluded that carrying out the process at temperatures around 930°C increases N<sub>2</sub>O formation, while increase of CO concentration in the exhaust gas reduces it. Presented results indicate that amount of N<sub>2</sub>O formed in the SNCR process may even reach 40% of removed NO<sub>x</sub>. The publication<sup>16</sup> presented results of research of N<sub>2</sub>O formation during SNCR system operation in a pulverised bed boiler (semi-industrial scale). No information concerning production of nitrous oxide in the process of NO<sub>x</sub> selective non-catalytic reduction carried out in coal-fired stoker boilers has been found in scientific literature. Combustion process in coal-fired stoker boiler has a decidedly different character from the process carried out in the pulverized coal fired boiler or fluidized bed boiler. Distributions of the exhaust gas temperature and velocity inside the furnace are quite different<sup>17, 18</sup>. The combustion process in coal-fired stoker boiler is carried out with much higher excess air ratio in comparison to other types of furnaces. The maximum flue gas temperature as well as the initial value of NO<sub>x</sub> concentration in the exhaust gas is also different. That is why experiences from pulverized coal fired boiler or fluidized bed boiler should not be transferred to a stoker-fired boilers.

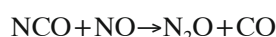
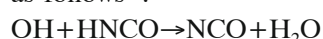
The purpose of presented research was to determine the dependency between the NO<sub>x</sub> and N<sub>2</sub>O concentrations in the exhaust gas when SNCR process is carried out in a coal-fired stoker boiler. This paper presents results of experimental investigation of N<sub>2</sub>O formation during the urea based selective non-catalytic NO<sub>x</sub> reduction process in a stoker boiler. Presented results have been obtained during tests of an SNCR system installed on a WR 25 boiler.

## N<sub>2</sub>O FORMATION MECHANISMS

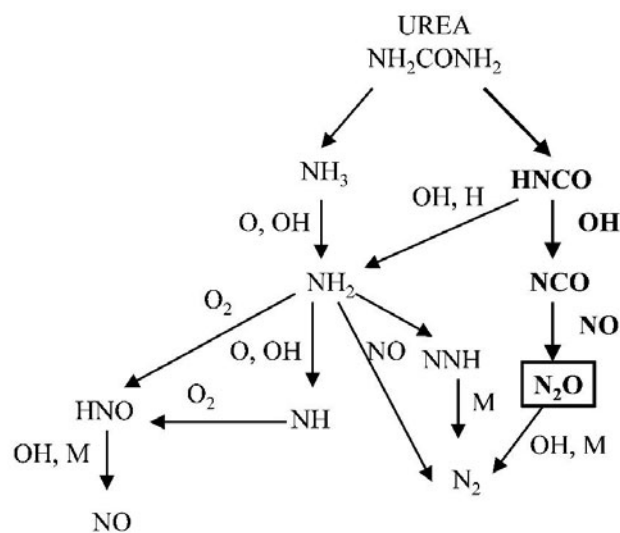
The mechanism of formation of N<sub>2</sub>O as a by-product of urea utilisation in SNCR systems has been comprehensively discussed in<sup>19, 20</sup>.

Urea (in form of aqueous solution) produces amine radicals in a gradual manner. Decomposition of urea molecules to reactive NH<sub>2</sub> radicals occurs only after water evaporates from a solution droplet. It is known that at the first stage the urea molecules decompose into ammonia and cyanic acid. Simplified scheme of reactions occurring during the SNCR NO<sub>x</sub> abatement process based on urea is shown in diagram (Fig. 1)<sup>15</sup>.

The N<sub>2</sub>O formation path involves the formation of NCO which subsequently reacts with NO to form N<sub>2</sub>O as follows<sup>15</sup>:



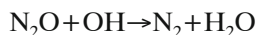
Decomposition of NH<sub>2</sub>CONH<sub>2</sub> into ammonia NH<sub>3</sub>, cyanic acid HNCO and isocyanate NCO is the fullest



**Figure 1.** Schematic diagram of urea-based SNCR process kinetics<sup>15</sup>

explanation of formation of  $N_2O$  as a by-product of using urea as SNCR reagent<sup>21</sup>.

At relatively higher temperatures (greater than  $1000^\circ C$ ), any  $N_2O$  formed will tend to be rapidly removed by the reaction:



Reagent injection temperature is significantly lower for coal stoker boiler than for a pulverized boiler. Moreover, results of research<sup>13</sup> suggest that mixing of flue gas with reagent has an effect on the  $N_2O$  formation.

## EXPERIMENTAL TESTS

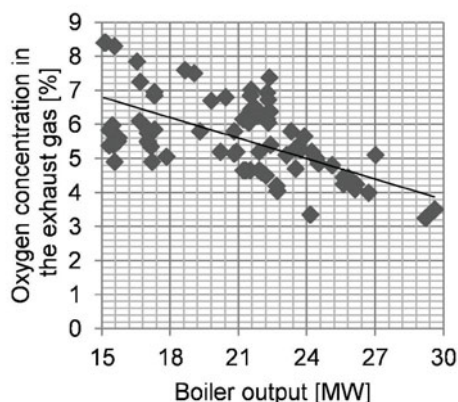
Results presented in this paper have been obtained through research of a stoker hot water boiler with a nominal rating of about 30 MW (Table 2).

**Table 2.** Basic technical data of tested boiler WR 25

Parameter	Value
Rated power	29 MW
Efficiency	85%
Fuel type	Coal
The calorific value of the fuel	21 MJ/kg
The temperature of exhaust gas for maximum power	$155 \pm 10^\circ C$
Minimum temperature of feed water	$70^\circ C$
The water flow for maximum power	90.0 kg/s

Measurements of  $N_2O$  concentration were performed during tests of the SNCR system dedicated for this boiler design.

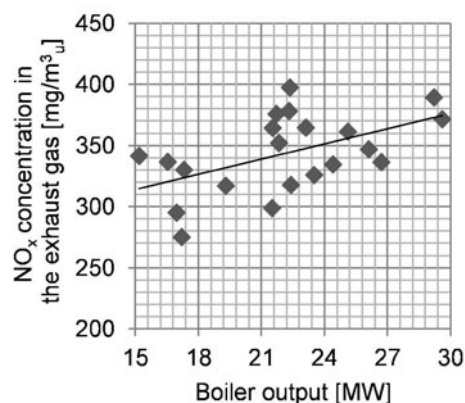
The measurements were taken while the boiler operated according to the consumer heat demand. During that time the boiler output considerably varied, from ca. 15 MW to nearly 30 MW. Output variations were accompanied by variations of oxygen concentration in the flue gas (Fig. 2).



**Figure 2.** Oxygen concentration in the exhaust gas in a function of boiler output. The measurement results

Also the base concentration of  $NO_x$  in the flue gas varied during the measurements – from ca.  $300 \text{ mg/m}^3$  for the output of 15 MW to ca.  $400 \text{ mg/m}^3$  for the nearly-nominal output. The chart (Fig. 3) presents  $NO_x$  concentration in the flue gas as a function of output (with SNCR not in operation).

In order to meet the new emission standards valid for the analysed boiler, it will be necessary to reduce the



**Figure 3.**  $NO_x$  concentration in the exhaust gas in a function of boiler output. The measurement results

$NO_x$  concentration by  $100\text{--}200 \text{ mg/m}^3$  (average reduction of  $150 \text{ mg/m}^3$ ).

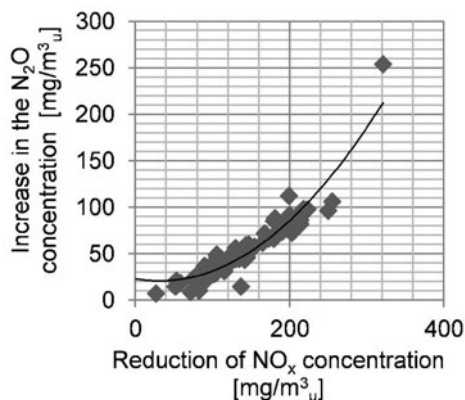
During the tests, the SNCR system was operated with a variable operating parameters – the reagent normalised stoichiometric ratio (NSR) – in range of  $0.5\text{--}3$ . The normalised stoichiometric ratio (NSR) of the reagent is defined as the ratio of the reagent supplied to the process to the amount of reagent in stoichiometric conditions.

During the tests, the gas composition was analysed with a Gasetm DX 4000 analyser. The sampling was performed by introducing a probe into the flue section downstream from all the heat transfer surfaces in the boiler, but upstream from the de-dusting system. Parameters monitored during the tests included concentrations of  $NO_x$ ,  $N_2O$  and CO in the exhaust gas.

## RESULTS AND DISCUSSION

Obtained measurement results conclusively demonstrate that there is a dependency between the  $NO_x$  and  $N_2O$  concentrations in the exhaust gas when the SNCR process is active in a stoker-fired boiler. As the  $NO_x$  reduction intensifies, the  $N_2O$  concentration grows (Fig. 4). Presented measured data is well correlated.

According to the presented data, reduction of  $NO_x$  concentration by  $150 \text{ mg/m}^3$  results with the increase of  $N_2O$  concentration in the exhaust gas by about  $55 \text{ mg/m}^3$ . Increased  $NO_x$  reduction leads to increased  $N_2O$  emission. Achieved values of  $N_2O$  concentration in the exhaust gas are high compared to emissions occurring when SNCR is not operating (about  $5\text{--}7 \text{ mg/m}^3$ ).

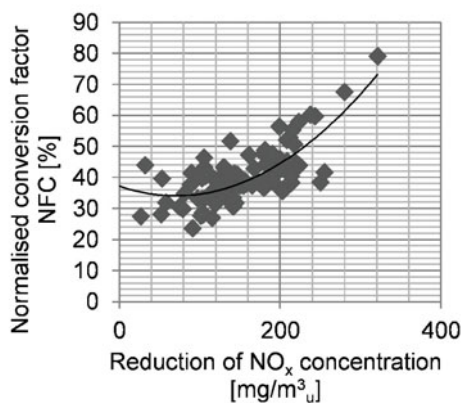


**Figure 4.** Increase in the  $N_2O$  concentration in a function of reduction of  $NO_x$ . The measurement results

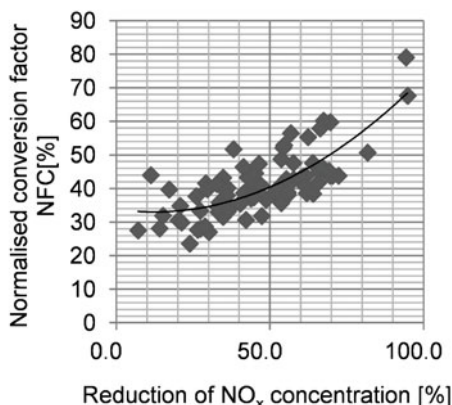
Moreover, the measurement results reveal that the  $\text{NO}_x$  to  $\text{N}_2\text{O}$  normalised conversion factor NFC (defined by the equation 1) is a variable and may be expressed as a function of  $\text{NO}_x$  emission reduction.

$$NCF = \frac{\Delta N_2O}{\Delta NO_x} \cdot 100\% \quad (1)$$

For  $\text{NO}_x$  reduction up to 40%, this factor (NFC) stays at steady level of about 30%. If the  $\text{NO}_x$  reduction exceeds 40%, the conversion factor starts to grow, reaching even 70% (Fig. 5, Fig. 6).



**Figure 5.** Normalised conversion factor NFC in a function of reduction of  $\text{NO}_x$  concentration. The measurement results



**Figure 6.** Normalised conversion factor NFC in a function of reduction of  $\text{NO}_x$  concentration. The measurement results

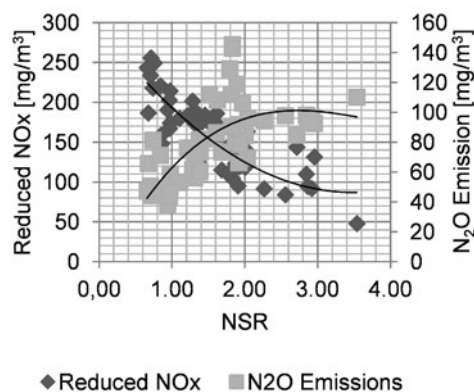
Obtained values of  $\text{NO}_x$  to  $\text{N}_2\text{O}$  conversion factor are higher than those published as valid for other types of boilers<sup>15, 16</sup>. The issue of  $\text{N}_2\text{O}$  emission attributable to SNCR process in stoker boilers may therefore be significant. This may be due to a lower injection temperature of the reagent and the use of higher NSR value during tests – up to 3.

Fulfilling new emission standards in the analysed boiler will require  $\text{NO}_x$  reduction of 40–50%. Thus it needs to be expected that the  $\text{N}_2\text{O}$  emission will be approximately 55–60  $\text{mg}/\text{m}^3$ , and  $\text{NO}_x$  to  $\text{N}_2\text{O}$  conversion factor (NCF) will be around 40%.

#### NSR impact on $\text{N}_2\text{O}$ emission

The diagram (Fig. 7) presents  $\text{NO}_x$  and  $\text{N}_2\text{O}$  concentrations as functions of NSR.

Growth of NSR value leads to reduction of  $\text{NO}_x$  (the reduction progresses), and  $\text{N}_2\text{O}$  concentration in the exhaust gas grows. Presented data indicates, that upon exceeding NSR value of 2, concentration of both  $\text{N}_2\text{O}$  and  $\text{NO}_x$  stabilises.

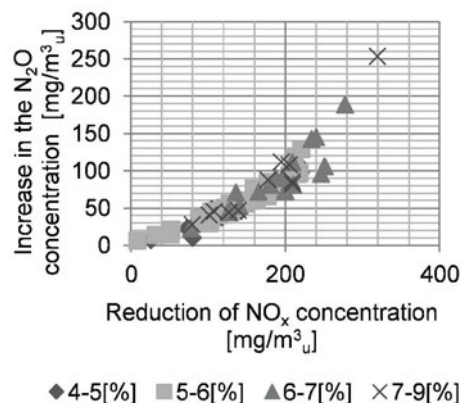


**Figure 7.** Effect of NSR on  $\text{NO}_x$  Reduction and  $\text{N}_2\text{O}$  formation. The measurement results

#### Impact of oxygen content in the flue gas on $\text{N}_2\text{O}$ emission

During the tests the oxygen content in the flue gas varied. The diagram (Fig. 8) shows the growth of  $\text{N}_2\text{O}$  concentration as a function of  $\text{NO}_x$  reduction for different oxygen contents in the flue gas.

Analysis of those data leads to a conclusion that in the conditions of the investigated boiler, oxygen concentration in the flue gas has no impact on the  $\text{N}_2\text{O}$  formation process which accompanies the  $\text{NO}_x$  reduction.



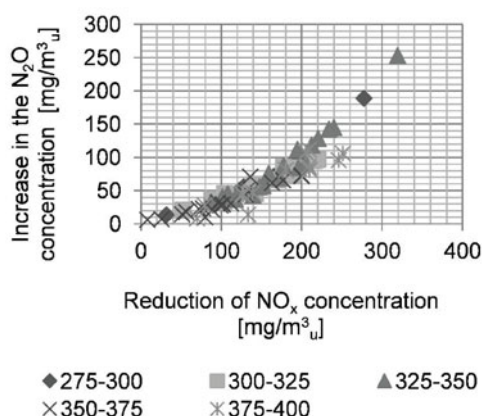
**Figure 8.** Increase in the  $\text{N}_2\text{O}$  concentration in a function of reduction of  $\text{NO}_x$  concentration for a various oxygen concentration. The measurement results

#### Impact of initial $\text{NO}_x$ concentration in the flue gas on $\text{N}_2\text{O}$ emission

According to the published data<sup>15</sup>, the initial  $\text{NO}_x$  concentration in the flue gas should have an impact on  $\text{N}_2\text{O}$  emissions attributable to the  $\text{NO}_x$  reduction process. Of course the higher the  $\text{NO}_x$  concentration is, the more significant reduction is needed to attain required emission levels. And larger  $\text{NO}_x$  reduction leads to higher  $\text{N}_2\text{O}$  emission, as demonstrated on the chart (Fig. 4). The authors of the paper decided to check whether or not the initial concentration of  $\text{NO}_x$  has any impact on the amount of  $\text{N}_2\text{O}$  being formed, if the  $\text{NO}_x$  reduction is comparable. The chart (Fig. 9) shows increase of  $\text{N}_2\text{O}$

concentration as a function of  $\text{NO}_x$  reduction for different initial  $\text{NO}_x$  concentration in the flue gas.

Presented data leads to a conclusion that in the conditions of the tested boiler the initial  $\text{NO}_x$  concentration has no impact on  $\text{N}_2\text{O}$  formation intensity.



**Figure 9.** Increase in the  $\text{N}_2\text{O}$  concentration in a function of reduction of  $\text{NO}_x$  concentration for a various initial  $\text{NO}_x$  contents. The measurement results

## CONCLUSIONS

Nitrous oxide is one of the major atmosphere pollutants. This gas, along with carbon dioxide and methane, is one of the three key gases which affect the greenhouse effect.  $\text{N}_2\text{O}$  emission in high temperature combustion exhaust from stoker coal fired boilers is mostly less than  $7 \text{ mg/m}^3$ , therefore this type of boilers is not a major  $\text{N}_2\text{O}$  source.

Increasingly strict emission standards for this type of equipment will enforce application of measures including secondary  $\text{NO}_x$  abatement methods. Adoption of those methods, except for desirable results of  $\text{NO}_x$  emission reduction, may lead to emission of undesirable substances such as  $\text{N}_2\text{O}$ .

The paper presented results of experimental tests concerning  $\text{N}_2\text{O}$  formation during the non-catalytic  $\text{NO}_x$  reduction process in a stoker boiler rated at around 30 MW. Measurements of  $\text{N}_2\text{O}$  concentration were carried out during tests of the urea based SNCR system dedicated for such a boiler design.

Obtained results conclusively demonstrate that there is a dependency between the  $\text{NO}_x$  and  $\text{N}_2\text{O}$  concentrations in the exhaust gas when the SNCR process is active in a coal-fired stoker boiler. If the  $\text{NO}_x$  reduction grows, so does the  $\text{N}_2\text{O}$  concentration.

Moreover, the measurement results indicate that the value of  $\text{NO}_x$  to  $\text{N}_2\text{O}$  conversion factor is variable (from 30 to 70%) as a function of  $\text{NO}_x$  reduction.

Meeting the new emission standards in the analysed boiler will require  $\text{NO}_x$  reduction of about 40–50%. It should be expected that the  $\text{N}_2\text{O}$  emission in such circumstances will be approximately  $55\text{--}60 \text{ mg/m}^3$ , and the  $\text{NO}_x$  to  $\text{N}_2\text{O}$  conversion factor – around 40%.

Obtained values of  $\text{NO}_x$  to  $\text{N}_2\text{O}$  conversion factor are higher than those published in the literature for other boiler types.

Moreover, it has been found that in the conditions of the tested boiler neither oxygen concentration in the exhaust gas nor the initial  $\text{NO}_x$  concentration influence

the  $\text{N}_2\text{O}$  formation process attributable to  $\text{NO}_x$  reduction process.

Values of the  $\text{NO}_x$  emission reduction and  $\text{N}_2\text{O}$  concentration growth is strongly correlated (correlation coefficient of 0.916).

It seems therefore, that the amount of formed  $\text{N}_2\text{O}$  mainly depends on the level of  $\text{NO}_x$  emission reduction and does not significantly depend on other parameters. In such a situation the  $\text{N}_2\text{O}$  emission may not be optimised in a wider range. It is a kind of a cost of using the SNCR technique based on urea as the reagent for the investigated type of boiler.

Based on the presented results it can be concluded that to maintain the ratio NCF at the low level i.e. 30–40%, the NSR value equal 1.6–1.8 should not be exceeded. This allows to maintain the required  $\text{NO}_x$  emissions in most cases.

## ACKNOWLEDGMENTS

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