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NO_X PREDICTION FOR FBC BOILERS USING EMPIRICAL MODELS

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Abstract. Reliable prediction of NO_X emissions can provide useful information for boiler design and fuel selection. Recently used kinetic prediction models for FBC boilers are overly complex and require large computing capacity. Even so, there are many uncertainties in the case of FBC boilers. An empirical modeling approach for NO_X prediction has been used exclusively for PCC boilers. No reference is available for modifying this method for FBC conditions. This paper presents possible advantages of empirical modeling based prediction of NO_X emissions for FBC boilers, together with a discussion of its limitations. Empirical models are reviewed, and are applied to operation data from FBC boilers used for combusting Czech lignite coal or coal-biomass mixtures. Modifications to the model are proposed in accordance with theoretical knowledge and prediction accuracy.

Keywords: NO_X prediction, FBC, empirical models.

1. Introduction

Fluidized bed combustion (FBC) technology provides an efficient and ecological way for low quality fuel combustion. The fuel is combusted in a bed of inert material that is brought into a fluidized state by passing through air, which leads to very intensive mixing of gases and solids inside the bed. A high degree of mixing enhances the heat and mass transfer by orders of magnitude compared to other combustion technologies. Intensive heat transfer is beneficial for keeping the combustion temperature low and uniform throughout the bed. The mass transfer helps to keep high combustion efficiency even for low-quality fuels, and facilitates emission control.

Nitrogen oxides NO and NO_2 (referred to as NO_X) are pollutant gases that cause photochemical smog, respiratory problems and damage to organisms. Emissions of these gases are therefore monitored and must be kept at a minimal level. Although transportation (internal combustion engines) is the major source of NO_X, control of NO_X emissions is efficient only in stationary combustion sources. Under typical combustion conditions of a solid fuel, about 95 % of the total NO_X is in the form of NO, and just 5 % is in the form of NO_2, which is much more noxious. Emissions of nitrogen oxides are influenced by fuel properties, combustion conditions and combustor design. Authors of NO_X prediction models for FBC usually combine a kinetic modelling approach with FBC hydrodynamic models. However, all existing models suffer from inaccuracy, overcomplexity, or both. A much simpler approach can be found for pulverized coal combustors (PCC), where the application of empirical models leads to very simple correlations that can achieve good agreement with experiments. However, these correlations are used exclusively for PCC, and no reference is available for modifying this method for FBC conditions [1, 2].

2. Theory

2.1. Formation of nitrogen oxides

Many authors have already written in detail about the formation of nitrogen oxides, see e.g. [2]. In general, there are three mechanisms of NO formation that are generally accepted: thermal, prompt and fuel. NO_2 is formed through oxidation of NO by HO_2 radicals that are present in low temperature regions of the flame. N_2O is formed from NO by reaction with NCO or ammonia radicals.

In FBC conditions, the vast majority of NO has its origin in fuel. Thermal and prompt NO formation mechanisms are insignificant, due to the low temperature in a fluidized bed. A further reduction compared to PCC can often be achieved, because the most of the NO is reduced to N_2 or N_2O. Homogeneous reduction occurs both in the freeboard and in the bed by reaction with CO and volatiles. Heterogeneous reduction takes place on the surface of devolatilized char particles inside the bed. Ash and bed material can have a catalytic effect on NO reduction. The low combustion temperature enhances the reduction of NO to N_2O (with the exception of biomass combustion, which is for example a case of waste combustion) [2, 3].

2.2. Prediction of NO_X for FBC boilers

The complexity of NO_X chemistry and the large number of influencing parameters make an accurate prediction very difficult. The most common approach for predicting the emissions of nitrogen oxides of FBC boilers is kinetic modelling combined with a detailed

flame	Premixed flame	Diffusion combustion	Staged
k_1	285	340	150
k_2	1280	835	80
k_3	180	20	−30
k_4	−840	−395	100

TABLE 1. Pohl's correlation coefficients for PCC boilers.

FB hydrodynamic model of the bed and freeboard. Recent models taking into account all occurring phenomena contain hundreds of reversible chemical reactions, and divide the bed into control volumes that can respect different flow patterns and hydrodynamics in different parts of the bed. These arrangements increase the complexity beyond acceptable limits.

An undisputed advantage of kinetic models is the prediction of nitrogen oxide concentration profiles through the bed and freeboard, which can be used to identify and validate the detailed chemistry. However, this information is not necessary for predicting stack emissions. [2] [4] [5]

2.3. Empirical modelling approach

The main advantage of empirical models is their simplicity. The data required is usually easy to obtain through proximate and ultimate analysis of the fuel and combustion parameters. By contrast with kinetic models, there is no need to solve an extensive equation system, or to have high for computation capacity available.

The prediction is based on experimentally derived correlations accounting for the dependency of the emissions on the influencing parameters. The parameters that have been identified to have the largest influence, and that are used in empirical models, can be classified within three groups:

- Fuel related (nitrogen content, volatile matter content, etc.)
- Boiler design related (staged/ unstaged combustion, extent of fuel - air mixing, etc.)
- Boiler operation related (excess air, combustion temperature, etc.)

The influence of individual parameters can be observed experimentally by keeping the other parameters constant. However, this approach presumes independent effects of the parameters, and this is not necessarily valid for all fuels and combustion conditions.

The main disadvantage of empirical models is uncertainty originating from lack of input data, e.g. ash composition can promote NO reduction under certain conditions, petrographic composition can significantly influence the devolatilization and char formation process. Another consideration is the extent of mixing of fuel and combustion air. To minimize the uncertainty,

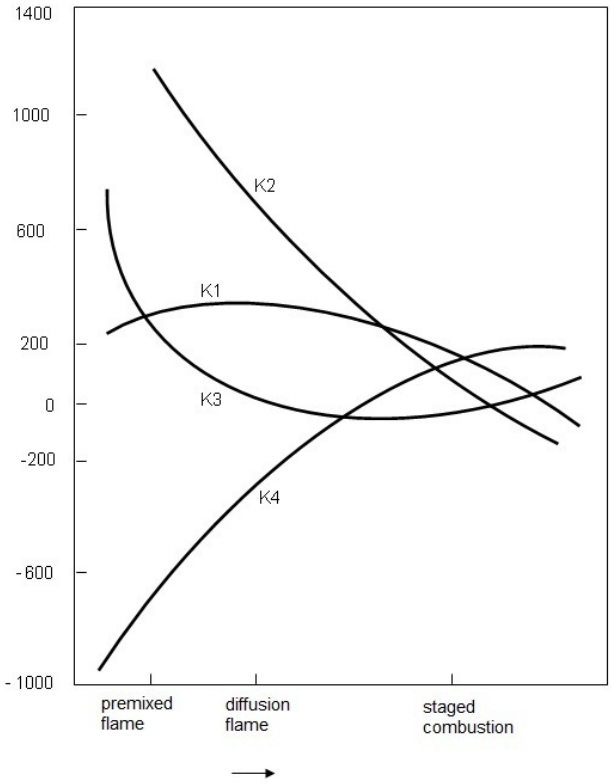

FIGURE 1. Pohl's correlation coefficients for PCC boilers [9].

correct parameters must be used in the model in order to cover all important factors, and at the same time not to increase the complexity.

Influencing parameters not included in the input data are taken into account via constants, and their applicability determines the limitations of the model. Input parameters and selection of constants should be carefully considered. Nevertheless, deviations in NO_X concentration can be measured in the flue gas stream due to inhomogeneity, so prediction reliability of ± 50 ppm can be considered acceptable.

From the models found in the literature, only Pohl's and Ibler's were chosen and applied to boiler data, because they assume general applicability [1, 6].

2.4. Pohl's model

A simple correlation was developed by Pohl et al. [1] to estimate NO emissions for controlled mixing conditions (various types of PCC flames, cf. Table 1):

$$NO[\text{ppm}] = k_1 + k_2 \frac{N^{\text{daf}}}{1.5} + k_3 \frac{VM}{40} NO_{\text{eq}} 3200 + k_4 \frac{FC}{60} \frac{NO_{\text{eq}}}{3200}, \quad (1)$$

where NO_{eq} [ppm] is the maximum emission of NO provided that all fuel nitrogen converts to NO, N^{daf} [%] is the nitrogen content in combustible, VM [%] is combustible volatile matter, and FC [%] is the fixed carbon content. NO_{eq} can be calculated from the nitrogen content in the fuel and dry flue gas volume

Steam output	125 t/hour
Steam temperature	490 °C
Steam pressure	7.3 MPa

Table 2. Steam nominal parameters for Komořany I CHPP.

LHV	13 MJ/kg
W^{r}	28 %
A^{r}	25 %
N^{daf}	1 %

Table 3. Fuel parameters for Komořany I CHPP.

V_{fd} [N m^3/kg], ash content A^{r} [–] and water content W^{r} [–], by

$$NO_{\mathrm{eq}}[\mathrm{ppm}] = \frac{2.1422 N^{\mathrm{daf}}(1 - A^{\mathrm{r}} - W^{\mathrm{r}})}{V_{\mathrm{fd}}} \cdot 10^6. \quad (2)$$

A different set of constants will presumably be needed for FBC conditions. As can be seen from Figure 1, only three combustion regimes are accounted for, and other fuel-air mixing regimes are not defined. Pohl's model was constructed on the basis of a wide range of experimental data from PCC boilers (diffusion flame) [1, 6–8].

2.5. Ibler's model

Ibler et al. [10] proposed the following correlation for predicting fuel nitrogen conversion to NO:

$$\frac{NO}{NO_{\max}}[-] = 7 \cdot 10^{-5} K \, C_{\mathrm{O_2}} \sqrt[3]{T - 1025}, \quad (3)$$

where K [–] is a fuel related constant (Ibler recommended using values of constant K between 4 and 6 for Czech coals), $C_{\mathrm{O_2}}$ [%] is the flue gas oxygen concentration and T [K] is the combustion temperature. The predicted concentration in ppm can be calculated by multiplying the fuel nitrogen conversion by NO_{eq} from Equation (2).

The constant $7 \cdot 10^{-5}$ in Equation (3) represents the PCC conditions, and a different constant will presumably be needed for FBC conditions. As can be seen from Equation (3), Ibler's model is targeted more on combustion conditions than on fuel properties, which are characterized by constant K only.

3. Experimental

The main aim of this paper is to make an evaluation of real measurement NOx emissions data from two large-scale fluidized bed boilers, and to make a comparison with the NOx levels predicted by Pohl's model and by Ibler's model.

Steam output	140 t/hour
Steam temperature	535 °C
Steam pressure	12.5 MPa

Table 4. Steam nominal parameters for Mladá Boleslav CHPP.

	Hard coal	**Lignite coal**	**Biomass pellets**
LHV	24.31	18.77	15.23
W^{r}	13.2	28.18	13.66
A^{r}	11.67	6.37	4.51
$Ndaf$	0.89	1.38	1.9

Table 5. Fuel parameters for Mladá Boleslav CHPP.

3.1. Komořany I CHPP

The K3 FBC boiler with a bubbling bed at the Komořany I combined power plant was used as the first reference. The lower part of the combustion chamber containing the bed is lined and contains an in-bed evaporator. The upper part contains wall and grid parts of the evaporator. The convection part, which follows the combustion chamber, contains the superheaters (primary, secondary and output) and the economizer. A tube-type air heater with a separate part for fluidization and secondary air is the last heat transfer surface of the boiler. The boiler is equipped with a bed material recirculation system as well as bed height control. The combustion process is controlled by the fluidization air flow rate and the fuel input. The steam parameters are adjusted by feed water injection before the last superheater. The steam nominal parameters are shown in Table 2. The lignite coal used was analysed before each combustion test. The results of the analysis were coupled with the NO_X emissions for the model predictions. The average parameters of coal are shown in Table 3.

3.2. Mladá Boleslav CHPP

The K90 FBC boiler with a circulating bed at the Mladá Boleslav combined heat and power plant was used as the second reference. This boiler is designed for hard coal combustion, but the recent fuel is a mixture of hard and lignite coal with the addition of biomass. The combustion chamber with a lined lower part contains the membrane-wall type evaporator. After the combustion chamber there is a cyclone for coarse particle separation. The second duct contains a membrane wall, tube and wall type superheaters and an economizer, followed by a hopper. The third duct contains a tube-type air preheater, which is the last heat transfer surface of the boiler. The steam parameters are controlled by feed water injection before the second and last superheater. The steam nominal parameters are shown (for hard coal) in Table 4. The

	Komořany	**Mladá Boleslav**
Oxygen concentration after economizer [%]	3.8–4.7	4–5
Fluidized bed temperature [°C]	815–866	873
Boiler load [%]	100	75–100
Fuel mixture by mass [%] – lignite coal, biomass, hard coal	100, 0, 0	40–85, 0–25, 0–50
CO concentration [mg/m^3]	79–222	

TABLE 6. Combustion parameters.

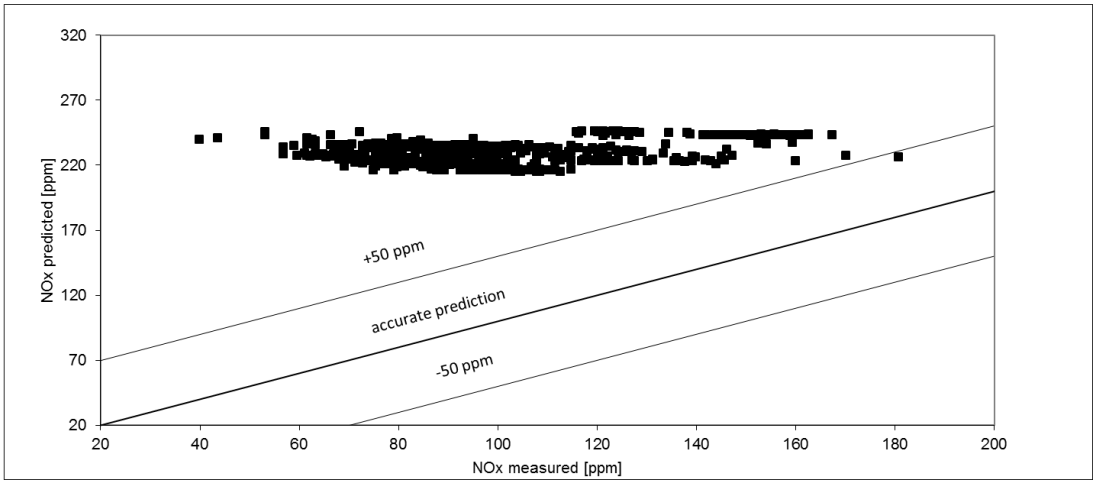

FIGURE 2. NO_x prediction reliability using the original Pohls method.

hard coal, lignite coal and biomass that were used were analysed before each combustion test. The results of the analysis were coupled with the NO_X emissions for the model predictions. The average fuel parameters are shown in Table 5:

4. Results

Experimental data from combustion tests on these boilers was taken from [11] and [12]. Combustion tests were carried out in these boilers covering the combustion conditions described in Table 6.

4.1. Pohl's model results

The coefficients for the staged combustion model were adopted as a basis for NO_X prediction using Pohl's model for FBC boilers. Three options were explored. The first option used Pohl's original model, as it was proposed by the authors in Equation (1). As expected, the reliability was very low, see Figure 2.

In the second option, coefficient k_1 was optimized by the least squares method for a better fit with the $x = y$ line. The best fit with determination index $R^2 = 79.43\,\%$ was found for $k_1 = 19.87$ — see Figure 3.

The third option incorporated the temperature and the excess oxygen dependency proposed by Ibler into coefficient k_1:

$$k_1 = 0.17 C_{O_2}^2 \sqrt[3]{T - 1025}. \qquad (4)$$

The modified constant k_1 was consecutively optimized by the least squares method for the best fit with the $x = y$ line, see Figure 4. The modified version of Pohl's method showed slightly better results with the determination index $R^2 = 79{,}54\,\%$.

Biomass	−2.2
Hard coal	3.1
Lignite coal 1	5
Lignite coal 2	3

TABLE 7. Values of fuel constant K.

4.2. Ibler's model results

The prediction results using Ibler's model were not in good agreement with the measured data, see Figure 5. To increase the reliability, the combustion constant was modified from $7 \cdot 10^{-5}$ to $2.9 \cdot 10^{-4}$ and fuel constants K were optimised to the values presented in Table 7, in both cases using the least squares method fitting the $x = y$ line. See the results in Figure 6 with $R^2 = 81.3\,\%$.

5. Conclusions

This paper has discussed the advantages and limitations of empirical prediction of NO_X emissions from the FBC boilers. With careful choice of input parameters and constants, empirical modelling can be in very good agreement with experimental data while

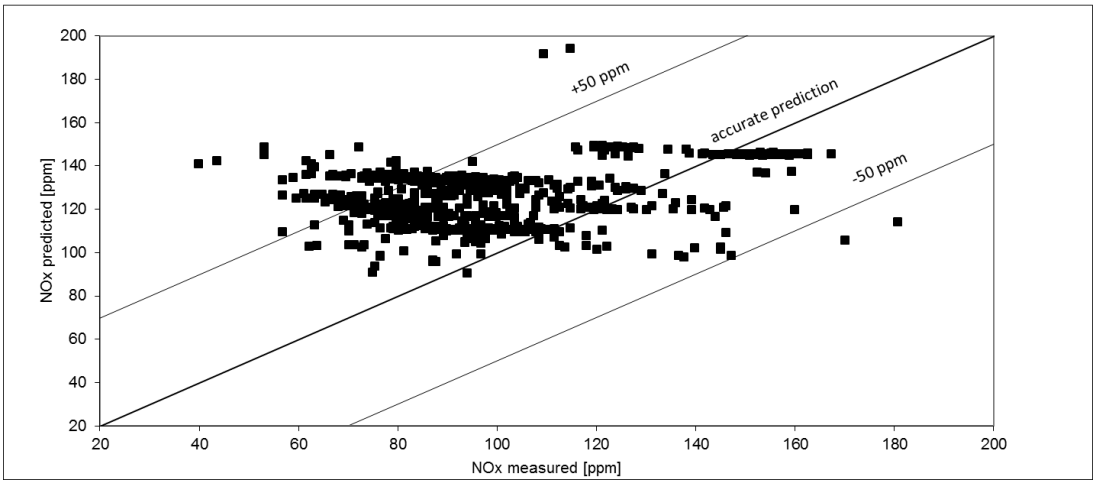

Figure 3. NO_x prediction reliability using Pohl's method with modified constant k_1

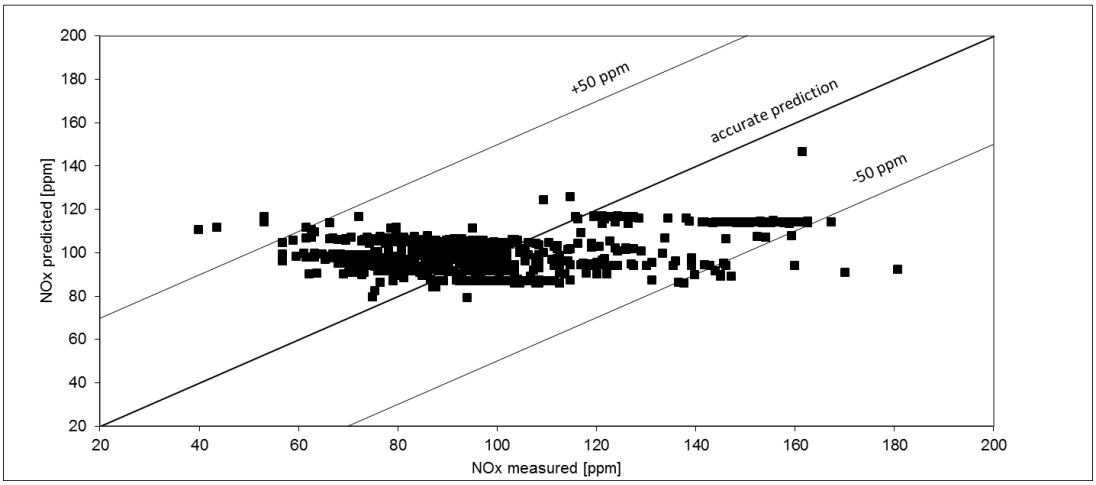

Figure 4. NO_x prediction reliability using Pohl's method with incorporated temperature and excess oxygen dependency.

keeping the model simple and the input data easy to obtain. Given the inhomogeneity of the flue gas stream, prediction accuracy of ±50 ppm can be considered reliable.

For FBC conditions, neither Pohl's model nor Ibler's model for PCC boilers provided satisfactory results without modifications. The models were adapted by least squares methods to fit the experimental data from two FBC boilers, in Komořany (combusting lignite coal) and in Mladá Boleslav (combusting a coal-biomass mixture).

The modified Pohl model for staged combustion with $k_1 = 19.87$, which uses only fuel parameters as input data, shows relatively good agreement with the measured data with $R^2 = 79.43\,\%$. The prediction accuracy increases to $R^2 = 79.54\,\%$ with the adoption of a modification to coefficient k_1 for temperature and excess air dependency taken from Ibler's model. However, most of the predicted value originates from the other constants, so the NO_X prediction is limited to a quite narrow range (cca 80–120 ppm), irrespective of the combustion conditions, and does not follow the measured trend.

Ibler's model, which focuses more on combustion parameters (temperature and oxygen concentration), and accounts for fuel properties by constant K only, shows better agreement, with $R^2 = 81.3\,\%$ for the combustion constant $2.9 \cdot 10^{-4}$ and fuel constants K taken from Table 7. The predicted NO_X emissions are from a much wider range (50–180 ppm), and seem to follow the experimentally observed trend well.

The prediction results from the modified models are in almost all cases within the 50 ppm limit, and can be considered reliable. However, the modified Ibler model has higher prediction accuracy and seems to be more suitable for FBC conditions.

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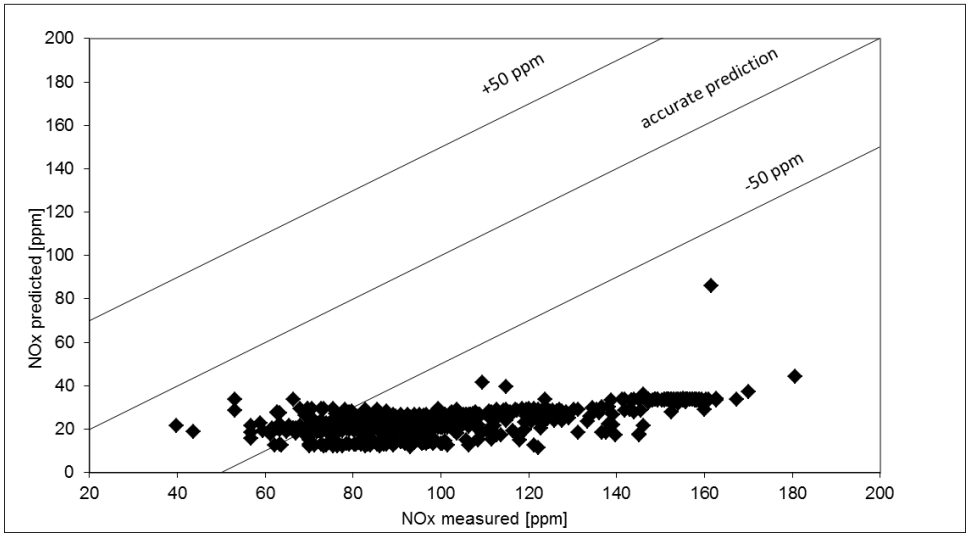

FIGURE 5. NO_X prediction reliability using Ibler's method.

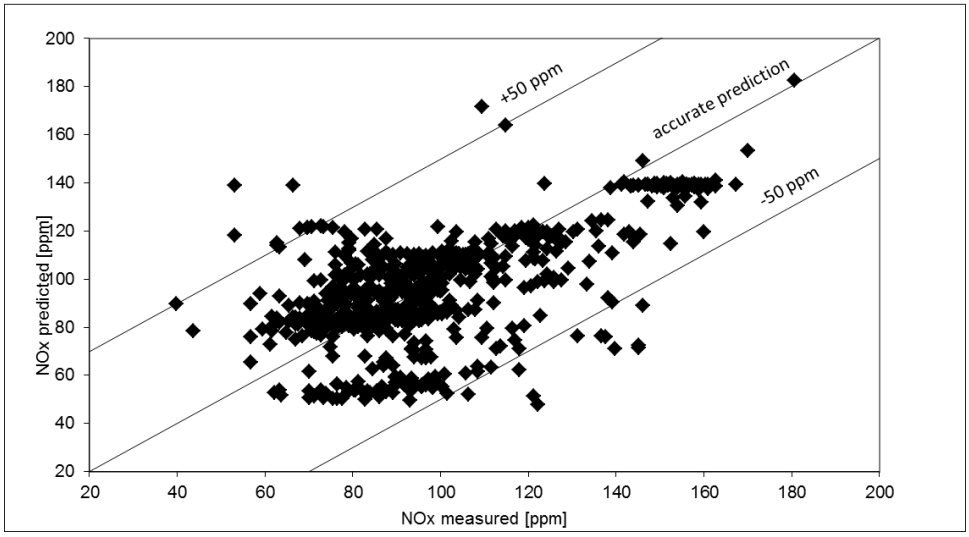

FIGURE 6. NO_X prediction reliability using Ibler's method with optimized constants.

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