

MODELING OF POLLUTANTS FORMATION IN GAS FIRED FURNACES

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ABSTRACT

Natural gas is more and more proving to be the most efficient fuel in the production of electricity in thermal power plants, from the aspect of minimizing the pollution. Burning gas that usually carries negligible amounts of sulfur containing substances therefore results in low sulfur oxides pollution. Due to the fact that gas has negligible amount of particles, the production of soot will also be limited. Therefore, the main pollutants left are the nitrogen oxides and carbon monoxide. With different methods the concentrations of those pollutants can be minimized. In order to advise those methods, detailed chemistry of methane/ethane combustion in air has to be studied and all significant mechanisms isolated. The most important mechanisms of the production of NO_x while burning natural gas in furnaces, which mainly occurs in oxygen rich (fuel lean - superstoichiometric) mixtures are thermal or Zeldovich mechanism and N_2O mechanism. The carbon monoxide chemistry is a very important part of the combustion of any carbohydrates, since nearly all carbon atoms get first oxidized to carbon monoxide, and only then to carbon dioxide. But what is of utmost importance for the concentration of carbon monoxide that leave the furnace, and therefore cause the pollution, is the amount of free radicals containing oxygen. In order to model natural gas combustion with all the details necessary to calculate the rate of formation of pollutants, the reaction system consisting of 143 elementary chemical reactions was devised. The reaction model was linked to the three dimensional mathematical model of heat and mass transfer previously developed by the authors. The model was tested by comparing the experimental results obtained from the literature with results of calculation. The model was then applied to the utility steam generator furnace, and the results considering pollution formation where discussed with the respect to the national regulation.

1. INTRODUCTION

The purpose of this paper is to show mathematical modeling of pollutants production in gas-fired furnaces, mainly carbon monoxide and nitrogen oxides. The mathematical modeling consists of heat and mass transfer and combustion modeling. That includes the convective, conductive and radiative heat transfer and turbulent mixture flow. The extensive work on the model was previously reported by the authors [1-11].

The combustion of methane in air consists of two main parallel chains, one by gradually reducing the number of hydrogen atoms, and other, by recombining two methyl radicals CH_3 into ethane (C_2H_6) and, again, gradually losing hydrogen atoms. In the same time there are several parallel chains of nitrogen oxides formation. The combustion can be represented by various reaction systems, depending of what has to be calculated. If only the heat flux is interesting, the easiest model to use is one-step global model [12]. The most complicated model can consist of several hundred elementary chemical reactions and a large number of chemical species [13-24].

2. MATHEMATICAL MODEL

Combustion

In order to model pollutants formation in a gas fired steam generator furnace, a combustion has to be represented by a kinetic model that includes not only fuel, air and flue gases, but also many intermediate species, that appear and disappear during the process. Those intermediate species are very unstable and reactive free radicals, like OH group or like O, H and N atoms.

There are four basic mechanisms of NO_x formation [25]: thermal, prompt, N₂O, and fuel mechanisms. Thermal or Zeldovich NO, forms at higher temperatures through the following chemical reactions:



In the combustion of fuels not containing nitrogen compounds oxidation of atmospheric nitrogen by the previous mechanism is a major source of NO_x emissions. In order to avoid the calculation of fuel oxidation some authors assume equilibrium values of temperature and N₂, O₂, O, OH and H concentrations. Meanwhile, that assumption is satisfied only in zones of slow production, and not on the flame front, where the O-atom concentration will be superequilibrium one. Therefore, for the calculation of the combustion in furnace, where different situations are occurring, it is necessary to calculate the fuel oxidation, in order to follow the superequilibrium concentrations.

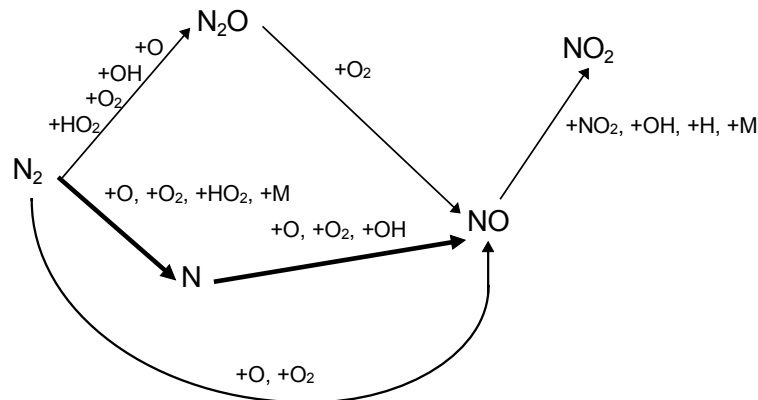


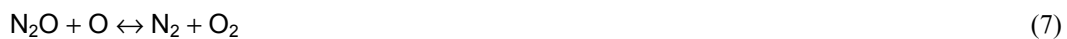
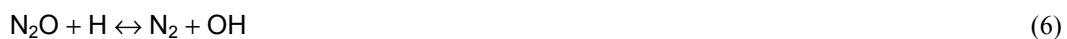
Figure 1. Nitrogen oxides formation mechanisms

Prompt NO forms at the flame front by recombination of CH radical and molecular nitrogen into HCN:



Nitrogen atom will further oxidize into NO. This mechanism is significant at lower temperature (1000 K) than thermal, and becomes very important in understoichiometric flames [26]. Since the combustion in the steam generator furnace is mostly superstoichiometric, this mechanism has been neglected in this paper.

The third, nitrous oxide (N₂O) mechanism is significant at lower temperatures and superstoichiometric flame conditions [26], and can be summarized by the following reactions [19]:



The nitrous oxide concentration will be significant in the low-temperature flame zones, where it is formed, but will be rapidly removed afterwards, in the higher-temperature zone with higher radical concentrations.

Table 1. Nitrogen oxides formation reactions

No	reaction	k_0	E_a	α	lit.
1.	$\text{N} + \text{O}_2 \leftrightarrow \text{NO} + \text{O}$	6.40E+06	2.63E+07	1.00	[24]
2.	$\text{N} + \text{OH} \leftrightarrow \text{NO} + \text{H}$	4.20E+10	0.00E+00	0.00	[21]
3.	$\text{N}_2 + \text{M} \leftrightarrow \text{N} + \text{N} + \text{M}$	9.90E+17	5.54E+08	-1.50	[14]
4.	$\text{N}_2 + \text{O} \leftrightarrow \text{NO} + \text{N}$	6.89E+10	3.14E+08	0.00	[21]
5.	$\text{N}_2\text{O} + \text{M} \leftrightarrow \text{N}_2 + \text{O} + \text{M}$	1.60E+11	2.16E+08	0.00	[13]
6.	$\text{N}_2\text{O} + \text{H} \leftrightarrow \text{N}_2 + \text{OH}$	7.60E+10	6.32E+07	0.00	[21]
7.	$\text{N}_2\text{O} + \text{O} \leftrightarrow \text{N}_2 + \text{O}_2$	1.00E+11	1.18E+09	0.00	[19]
8.	$\text{N}_2\text{O} + \text{O} \leftrightarrow \text{NO} + \text{NO}$	1.00E+11	1.18E+09	0.00	[19]
9.	$\text{N}_2\text{O} + \text{OH} \leftrightarrow \text{N}_2 + \text{HO}_2$	2.00E+09	4.19E+07	0.00	[13]
10.	$\text{NO} + \text{M} \leftrightarrow \text{N} + \text{O} + \text{M}$	5.20E+18	3.16E+08	-1.50	[14]
11.	$\text{NO} + \text{HO}_2 \leftrightarrow \text{NO}_2 + \text{OH}$	3.02E+09	1.00E+07	0.50	[21]
12.	$\text{NO} + \text{NO} + \text{O}_2 \leftrightarrow \text{NO}_2 + \text{NO}_2$	7.95E+03	0.00E+00	0.00	[21]
13.	$\text{NO} + \text{NO} \leftrightarrow \text{N}_2 + \text{O}_2$	4.80E+20	1.80E+08	-2.50	[14]
14.	$\text{NO} + \text{O}_2 \leftrightarrow \text{NO}_2 + \text{O}$	1.90E+09	1.95E+08	0.00	[21]
15.	$\text{NO}_2 + \text{M} \leftrightarrow \text{NO} + \text{O} + \text{M}$	1.10E+13	2.76E+08	0.00	[13]
16.	$\text{NO}_2 + \text{H} \leftrightarrow \text{NO} + \text{OH}$	3.50E+11	6.28E+06	0.00	[13]
17.	$\text{N} + \text{CO}_2 \leftrightarrow \text{NO} + \text{CO}$	1.90E+08	1.42E+07	0.00	[13]

Fuel nitrogen mechanism is significant in nitrogen-rich fuels. The nitrogen bound in the fuel quickly converts into ammonia, NH_3 , or into HCN. Almost two thirds of fuel bound nitrogen will finish up as NO, in case of superstoichiometric combustion, while for understoichiometric flames the ratio decreases, reaching the minimum at $\lambda = 0.7$ [26]. The fuel nitrogen mechanism is particularly important for coal fuels, while for gaseous fuel generally, there is no nitrogen chemically bound in the fuel. Since this paper considers only natural gas as fuel, this mechanism is neglected.

Besides thermal and nitrous oxide mechanisms, nitrogen dioxide (NO_2) and several less significant mechanisms were added, to form the reaction system of nitrogen oxides formation, as shown on Fig. 1. The reactions taken into account are listed in Table 1.

The reaction rate at which an elementary chemical reaction influences the rate of change of participating chemical species concentrations depends on reactant concentrations and the temperature T only, since the pressure is nearly constant in a steam generator furnace. According to the Arrhenius law of reaction rate, the temperature influence can be calculated from:

$$k = k_0 T^\alpha e^{-\frac{E_a}{RT}} \quad (9)$$

where $k_0 T^\alpha$ represents the molecular collision frequency, E_a is activation energy, and R is gas constant.

Boltzmann factor $e^{-\frac{E_a}{RT}}$ represents the ratio of particles that have achieved the necessary energy level to react. The values for activation energy E_a , collision frequency coefficient k_0 and temperature exponent α are mainly determined experimentally for each elementary chemical reaction, and are shown for nitrogen chemistry in the Table 1.

In order to calculate the actual radical concentrations (O, H, OH, etc.) the entire kinetic system of fuel combustion has to be included. The system chosen consists of 143 elementary reactions (including 17 in the Table 1), and 31 chemical species, as shown in [11]. The reaction system consisting of n_r opposed reaction pairs with n_α chemical species can be represented by:



where i and j are not tensor indices. The change of concentration C_i of chemical species M_i as a consequence of participating in n_r reactions with n_α chemical species (10) is given by:

$$\frac{dC_i}{dt} = \sum_{j=1}^{n_r} (v_{ij}'' - v_{ij}') k_{f,j} \prod_{k=1}^{n_a} C_k^{v_{kj}'} \left(1 - \frac{1}{K_{C,j}} \prod_{k=1}^{n_a} C_k^{(v_{kj}'' - v_{kj}')} \right) \quad (11)$$

The reaction rate coefficient $k_{f,j}$ is calculated according to the Arrhenius law (9):

$$k_{f,j} = k_{0,j} T^{\alpha_j} e^{-\frac{E_{a,j}}{RT}} \quad (12)$$

Transport Phenomena

The heat and mass transport in steam generator furnace is calculated in a usual way [9], with a general transport partial differential equation for steady state given as:

$$\frac{\partial}{\partial x_j} (\rho v_j \varphi) = S_\varphi + \frac{\partial J_{\varphi,j}}{\partial x_j} \quad (13)$$

Particular transport equations, mass, momentum, one for each chemical species and enthalpy, are given in Table 2. Chemical species equation, given in the third row, one for each species considered by the reaction model, is used for the calculation of the mass fraction Y_α transport, for each chemical species α . The source term, the production ω_α of chemical species, is a change of mass fraction due to combustion and can be calculated from the change of concentration (11):

$$\omega_\alpha = W_\alpha \frac{\partial C_\alpha}{\partial t} \quad (14)$$

Table 2. Transport equations

name	φ	S_φ	$J_{\varphi,j}$	equation
Continuity equation	1	0	0	$\frac{\partial}{\partial x_j} (\rho v_j) = 0$
Momentum equation	v_i	ρf_i	σ_{ij}	$\frac{\partial}{\partial x_j} (\rho v_j v_i) = \rho f_i + \frac{\partial \sigma_{ij}}{\partial x_j}$
Chemical species equation	Y_α	ω_α	$-j_{\alpha,j}$	$\frac{\partial}{\partial x_j} (\rho v_j Y_\alpha) = \omega_\alpha - \frac{\partial j_{\alpha,j}}{\partial x_j}$
Enthalpy equation	h	$q_C'' + q_R''$	$-q_j$	$\frac{\partial}{\partial x_j} (\rho v_j h) = q_C'' + q_R'' - \frac{\partial q_j}{\partial x_j}$

3. NUMERICAL PROCEDURE

The partial differential equations governing transport processes are integrated over control volume, as in [1]. That will result in a system of algebraic equations, one system for each differential equation:

$$a_{\varphi,P} \varphi_P = \sum_{np} a_{\varphi,np} \varphi_{np} + b_\varphi \quad (15)$$

The chemical reactor model represented by eqs. (11, 14), is then applied to each control volume. The initial conditions are calculated from the values in the upwind control volumes, as explained in [11]. The system of ordinary differential equations (11), one equation for each chemical species, is solved for each control volume by Gear method [27], for the time the flow resides in a particular control volume. The residual time is calculated from the average velocity in the control volume. The resulting concentration differences are used for the calculation of mass fraction production rates (14). The transport problem, represented by the system of algebraic equations (15), is solved by the modified PISO algorithm, as explained in [1].

4. RESULTS

Combustion of hydrogen in air

The chemical reactor model was tested on an example available in the literature [28], the combustion of hydrogen in air, with initial temperature of 1500 K and pressure of 2 bar. The result of comparison for temperature and some chemical species is shown on Fig. 2. The results of simulation are shown by line, and the data from [28] are shown by markers, in corresponding colors. The equilibrium state has been achieved after $5 \cdot 10^{-4}$ s, meaning that afterwards there is no change in chemical species molar fractions and temperature. The data shows very good agreement.

Table 3. Equilibrium molar fractions of nitrogen oxides

Chemical species	This model [kmol/kmol]	lit. [28] [kmol/kmol]
N ₂	$6.12 \cdot 10^{-1}$	$6.08 \cdot 10^{-1}$
NO	$6.91 \cdot 10^{-3}$	$9.75 \cdot 10^{-3}$
N ₂ O	$3.88 \cdot 10^{-7}$	$6.35 \cdot 10^{-7}$
NO ₂	$1.98 \cdot 10^{-6}$	$1.97 \cdot 10^{-6}$

For nitrogen species only the equilibrium values are available from [28]. They are compared in Table 3. The agreement for nitrogen dioxide is excellent, while for nitric oxide and nitrous oxide the values obtained by this model are somehow lower than in [28]. It is probably due to the different reaction model used in two papers, the newer one used in this paper.

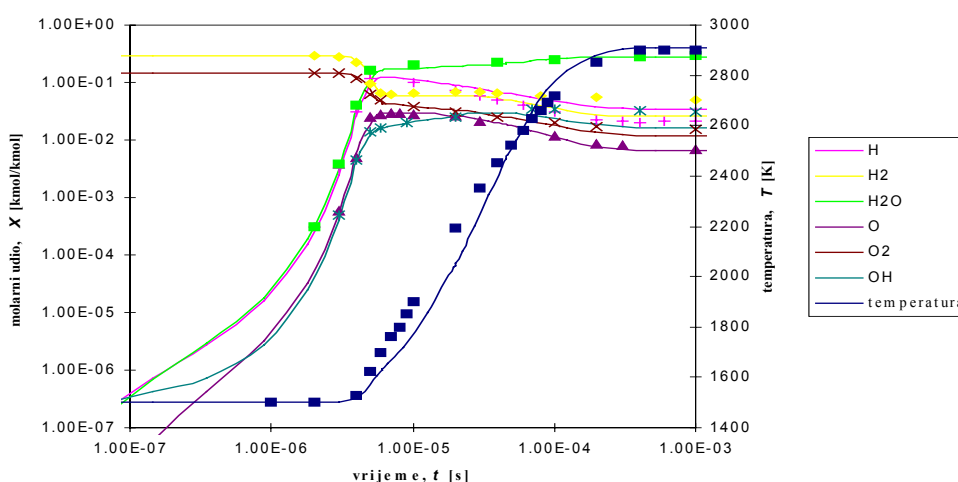


Figure 2. Comparison of results obtained by the model and data from [28]

Nitrogen oxides in utility steam generator

The previously explained model was applied to utility steam boiler RAMZIN P-56, steam capacity $D = 330$ t/h, superheated steam temperature $t_{pr} = 540$ °C and pressure $p = 140$ bar at full load, natural gas fired ($H_d = 34460$ kJ/m_n³). Dimensions of the furnace are area $A = 8.4 \times 8.4$ m² and height $h = 25.1$ m. The flame is superstoichiometric, $\lambda = 1.05$.

Table 4. Comparison of flue gas temperature at the furnace outlet

Calculation	temperature [°C]
Normative [29]	1048
Producer [30]	1015
DVODIM [2]	1044
ZONAL 4.40	1002

The developed mathematical model was applied using grid of $16 \times 32 \times 16 = 8096$ control volumes. Table 4 shows comparison for average flue gas temperature at the furnace outlet. Result of this model, calculated by ZONAL, shows better agreement with the datum given by the producer, than both the classical normative calculation and two-dimensional calculation, obtained by DVODIM, which was expected. Table 5 shows average outlet values for pollutant species.

Table 5. Average values at the furnace outlet

Chem. Species	\dot{m}_α [kg/s]	Y_α [kg/kg]	X_α [kmol/kmol]	X_α [ppm]
NO	1.51E-01	1.45E-03	1.34E-03	1340
N ₂ O	6.00E-07	1.00E-08	3.54E-09	0.00354
NO ₂	2.81E-05	2.70E-07	1.62E-07	0.162
CO	1.52E-02	1.45E-04	1.44E-04	144

Fig. 3 shows carbon monoxide distribution at two side cross-sections, one close to the burner side wall (Fig. 3a), and the other at the central plane (Fig. 3b). Further on, one front view cross-section in the burner plane (Fig. 3c), and one top view burner plane (Fig. 3d) are shown. It is clear from Figs. 3c and 3d that CO does not generate in the cold flame zone, but mainly at the flame front. Soon, the carbon monoxide mass fraction reaches the maximum further on being oxidized into carbon dioxide. Meanwhile, at Figs. 3b and 3c a secondary zone of CO generation can be seen in the region where the flame flows upwards, what is a consequence of CO₂ dissociation.

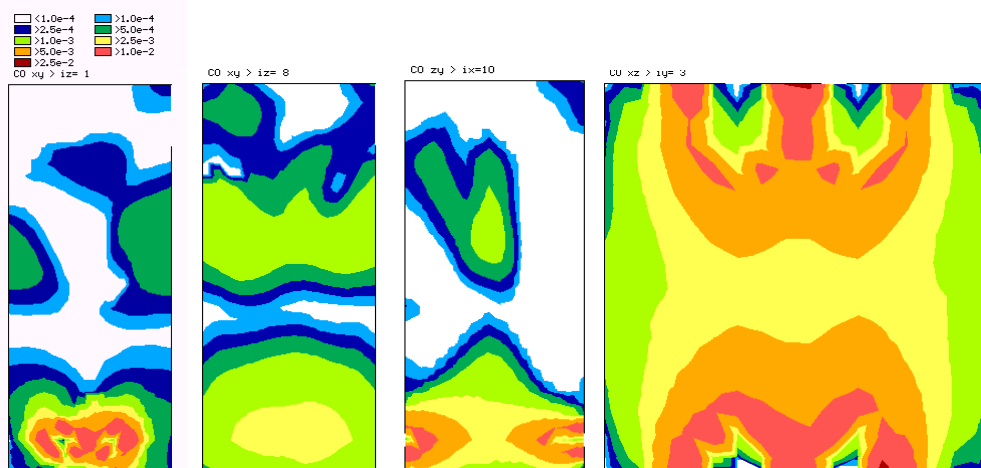


Figure 3. Carbon monoxide distribution at various furnace cross sections

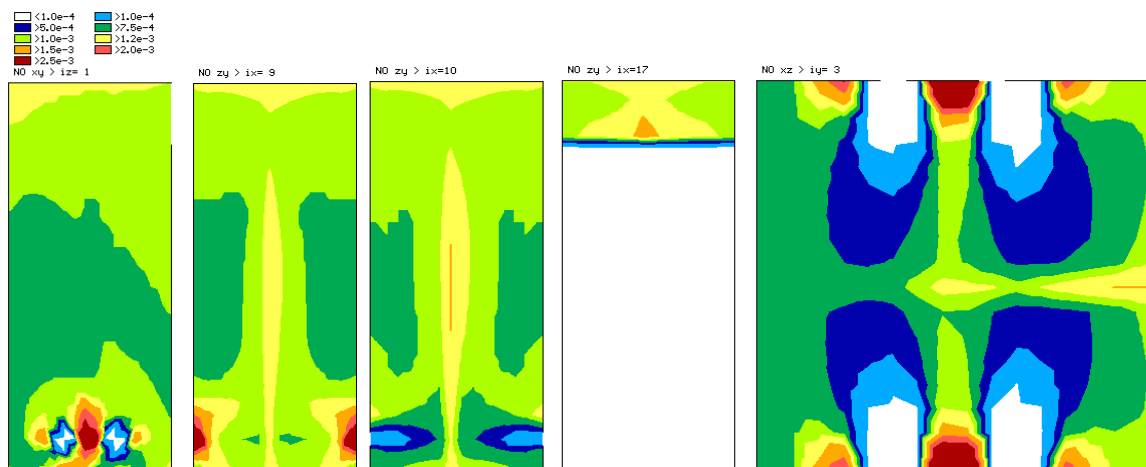


Figure 4. Nitric oxide distribution

Nitric oxide distribution is shown on Fig. 4. Clear mass fraction maximum can be found between the two burners (Figs. 4a, 4b, 4e), and somehow lower, at the outer edge of burners (Fig. 4e). In the cold zone, and in the region

of high velocities, the nitric oxide mass fraction is low, meaning that it takes longer time to generate. That explains maximums close to the wall, where the temperature is high but velocity is low. Figures 4b-c show another zone of NO production, a thin zone along the longest furnace axis. Meanwhile, nitrogen dioxide is generated already in the cold zone, and reaches the maximum concentration quickly (Figs. 5a-b and 5d), to decrease later on. In the upper part of furnace, close to the side walls, NO₂ is again generated, outside of the flame region, what increases the mass fraction (Fig. 5a). In the flame center the NO₂ mass fraction is low (Fig. 5b).

Nitrous oxide distribution is shown on Figure 6. It mainly gets generated in zones of high temperature gradients around burners (Figs. 6b and 6d). With temperature decrease most of N₂O will further oxidize into nitric oxide (Figs. 6a-b).

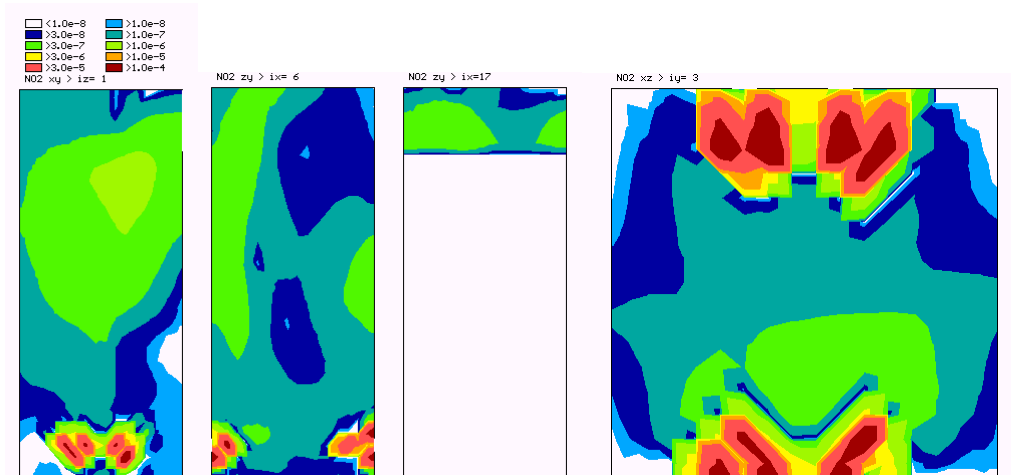


Figure 5. Nitrogen dioxide distribution

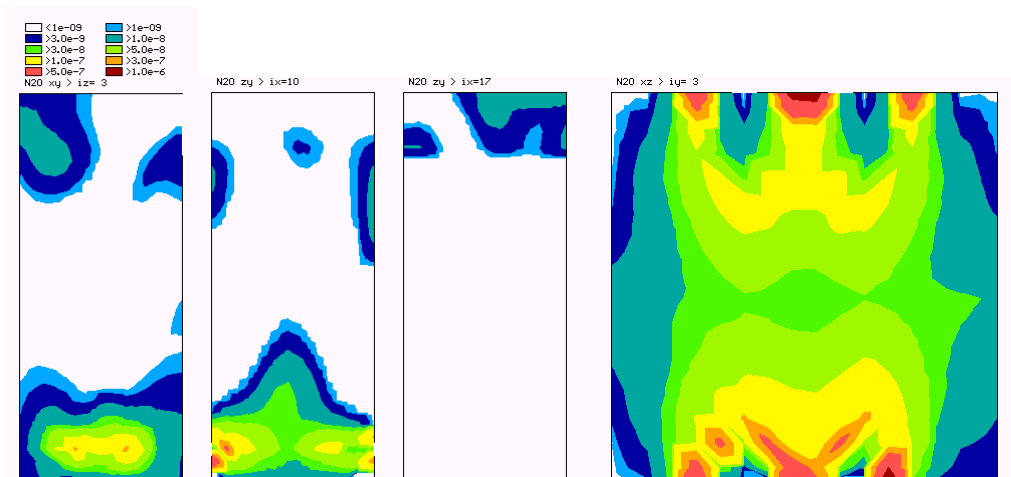


Figure 6. Nitrous oxide distribution

5. CONCLUSION

The obtained results of NO_x and CO pollution for the given steam generator are within the acceptable limits of EU standards which are accepted in Croatia either. The value of 1340 ppm NO_x (Tab. 5) yields approximately 1300 mg/m³ based on dry flue gases with 6 % O₂. The current EU limit is 650 mg/m³ for the new plants. But for the existing plants different countries prescribe different values. For example in France upper limits are 650-1300 mg/m³, in Germany 200-1500 mg/m³, Poland 95-1340 mg/m³, Turkey 940-1875 mg/m³. The range of upper limits depends upon the capacity and type of the plant and economic strength of the country. The CO value of 144 ppm is quite low and well below permissible limit of 250 mg/m³. It depends on the amount of access air and the fuel-air mixing process. In the particular case the fuel burners are equipped with the steam atomizers which enable good mixing of fuel and air.

List of Symbols

symbol	description	unit
a	coefficient	kg/s
A	area	m ²
b	coefficient	*kg/s
C	concentration	kmol/m ³
D	steam flow	t/h
E_a	activation energy	J/kmol
f	specific mass force	m/s ²
h	specific enthalpy	J/kg
	height	m
H_d	lower calorific value	J/m ³
j	chemical species flux	kg/m ² s
k	reaction rate coefficient	(kmol/m ³) ^{1-m} /s
k_0	collision frequency coefficient	(kmol/m ³) ^{1-m} /sK ^{α}
K_C	equilibrium constant	(kmol/m ³) ^{Δn}
M	one of chemical species	–
\dot{m}	mass flux	kg/s
Δn	difference of sums of stoichiometric coeff. of products and reactants	kmol
n_r	number of pairs of opposed reactions	–
n_α	number of chemical species	–
p	pressure	Pa, bar
q	heat flux density	W/m ²
q'''	volumetric heat flux density	W/m ³
R	gas constant	J/kmol K
S	source of physical property	*kg/m ³ s
t	time	s
T	temperature	K
v	velocity	m/s
W	molecular mass	kg/kmol
x	coordinate	m
X	molar fraction	kmol/kmol
Y	mass fraction	kg/kg
α	temperature exponent u Arrhenius law	–
λ	excess-air factor	–
ν	stoichiometric coefficient	kmol
ρ	density	kg/m ³
σ_{ij}	Stress tensor	N/m ²
φ	physical property	*
ω	rate of production of chemical species	kg/m ³ s

unit explanations

m	number of reactants in a reaction
–	non-dimensional
*	unit depends on unit of physical property φ

index	description
b	backward reaction
C	combustion, concentration
f	forward reaction
i	i^{th} chemical species
j	index, j^{th} reaction
k	k^{th} chemical species
np	neighboring control volume
P	central control volume
pr	superheated
R	radiation
α	chemical species
φ	physical property
'	reactant
"	product

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