

# Dynamics of Harmful Emissions from a Coal-Fired Thermal Power Plant in the Industrial Region of Devnya, Bulgaria

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**Abstract:** The survey was made at Deven Power Plant – an installation for combined generation of heat and electrical power (cogeneration system). The fuels at the plant are imported coal, anthracite, petcoke and diesel oil for firing the boilers. The main pollutants from Deven Power Plant have been tracked: nitrogen oxides  $\text{NO}_x/\text{NO}_2$ , sulphur oxides  $\text{SO}_x/\text{SO}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$  and particulate matter PM, for the period 2007 – 2015. The period was chosen in relation with the coming into force of the IPPC permit of the installation (№ 93/2006) for environment protection. The survey has shown that as a result of the measures implemented and given in the IPPC permit and the introduction of a new steam generator with a circulating boiling layer (CBL-SG) at the end of 2009, a reduction of emissions is observed of nitrogen oxides, sulphur oxides and PM, and the released quantities of PM for 2015 are under the threshold. The concentration of harmful substances from the emitting devices: steam generators  $\text{SG}_1\text{-SG}_6$  within one common chimney and the CBL-SG have decreased considerably after the coming into force of the IPPC permit, and the differences have high statistical significance ( $0.001 \leq P \leq 0.05$ ).

**Keywords:** Thermal power plant, emissions,  $\text{NO}_x/\text{NO}_2$ ,  $\text{SO}_x/\text{SO}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$ , PM

## I. INTRODUCTION

The Law on Environment Protection in Bulgaria requires that all operators of industrial and combustion installations, as in Annex 1 of Regulation (EO) 166/2006, report data about the release and transfer of pollutants from said installations [1]. With Regulation (EO) 166/2006, the European Parliament and Council of the European Union established a European Pollutant Release and Transfer Register (PRTR) as a publicly accessible database, which facilitates the prevention and reduction of pollution of the environment, and enhances the participation of the public into the decision-making concerning environment protection. In accordance with the terms of the Regulation, the EU member-states are obliged to report the release of pollutants from all kinds of activities within the framework of Annex II of the Regulation. It includes 65 activities, grouped in 9 sectors: Energy; Production and Processing of Metals; Mineral Industry; Chemical Industry; Waste and Wastewater Management; Paper and Wood Production and Processing; Intensive Livestock Production and Aquaculture; Animal and vegetable products from the food and beverage sector and Other Activities [2,3]. The emissions as per the European Pollutant Emission Register are reported, when they are above the threshold [4]. Annex II of Regulation 166 includes 91 pollutants in ambient air, water and soil, and each pollutant is given a limit (emission threshold).

One of the requirements of the Law on Air Quality in Bulgaria is limit emissions of harmful substances from different activities with the aim to protect the health of the exposed population and of the environment as a whole [5]. In industrial regions, such containment is achieved through the regulation mechanisms of the IPPC permits in accordance with Directive 2008/1/EO of European Parliament and the Council concerning integrated pollution prevention and control and the Ordinance on the Conditions and Order of Issuing of IPPC permits [6,7]. The IPPC permits guarantee the implementation of the best production methods, suitable technologies for purification of gases and reduction of the amount of emissions into the air; introduction of new facilities for purification of flue gas or modernisation of existing ones.

When producing electricity from thermal plants using fossil fuels, large quantities of specific pollutants are released, such as: sulphur dioxide  $\text{SO}_2$ , nitrogen oxides  $\text{NO}_x$ , non-methane volatile organic compounds NMVOC, methane  $\text{CH}_4$ , the main greenhouse gas-carbon dioxide  $\text{CO}_2$ , as well as other key pollutants in the region of power plants – particulate matter (PM) [8-15]. The content of  $\text{SO}_2$  in flue gas depends mainly on the sulphur content in coal, and this of  $\text{NO}_x$ , on the technological combustion process, and the main measures for reducing  $\text{NO}_x$  emissions are directed at the optimisation of the combustion process [16,17]. Over 50% of the total emissions come from combustion installations and directly relate to  $\text{SO}_2$ ,  $\text{CO}_2$ , heavy metals and polycyclic aromatic hydrocarbons; 10% go to  $\text{NO}_x$ ,  $\text{CO}$ , NMVOC and PM and a bit over 20% from the total amount of emissions – to  $\text{CH}_4$  and nitrous oxide  $\text{N}_2\text{O}$  [18].

When preparing an emission inventory from electricity production, it has been established that the annual emissions of  $\text{CO}_2$  amount to 67 mln tons, of  $\text{NO}_x$  – 0.15 mln tons, of  $\text{SO}_2$  – 0.04 mln tons and PM – 0.005 mln tons [19]. For the period 1990-2010, the use of coal in China increased, as well as the harmful substance emissions released from coal thermal stations – for  $\text{SO}_2$  with 56%;  $\text{NO}_x$  – 335%;  $\text{CO}_2$  – 442%. After 2005, control measures were increased and sulphur purification installations were introduced for reducing  $\text{SO}_2$  emissions [20].

Implementing a suitable dispersion model, the spatial distribution of  $\text{SO}_2$  and  $\text{PM}_{10}$  has been tracked, which survey established a significant impact of pollutants on air quality in the region of coal-fired thermal power plants, which requires integrated control measures [21].

The aim of this survey is to evaluate the amount of harmful substance emissions from coal-fired thermal power plants in an industrial region by comparing the emissions from before and after the introduction of the IPPC permit, and also track and evaluate the implemented measures for emissions reduction and the improvement of ambient air quality in the region.

## II. MATERIAL AND METHODS

The survey was performed at *Deven AD*, an installation for combined production of heat and electricity, used by the plants in the industrial region of Devnya. It covers 121.052 km<sup>2</sup> in Northeast Bulgaria. On its territory are located plants for the production of soda ash, nitrogen and phosphate fertilisers, clinker and cement, *Varna-Zapad /Varna-West/Port* and a phosphogypsum depot.

The main fuels in *Deven Power Plant* are: imported coal, anthracite, petcoke and diesel oil for firing the boilers. Petcoke is delivered from refining processes of petrol and is characterised by high concentrations of carbon (over 90%), high sulphur content (5-6%) and heavy metals (mainly nickel and vanadium) which makes it a pollutant fuel. Up until 30/11/2009, *Deven Power Plant* had 6 fossil fuel burning steam generators with total capacity 640 MW and 8 turbine units with total electrical capacity 125 MW. At the end of 2009, within the steam production scheme a new No 7 steam generator with circulating boiling layer was included, and put into operation with Report and Implementation Permit No CT-05-1459/01.12.09, with steam output of 400 t/h, and in this way *Deven Power Plant* has turned into the largest plant on the Balkans for producing both heat and electricity. The total production output of the installation for 2015 is 700 MW – burner No 1 for superheated steam with coal-fired steam generators Nos 2, 3 and 6 and a circulating boiling layer steam generator No 7.

The main pollutants from *Deven Power Plant* are: nitrogen oxides NO<sub>x</sub>/NO<sub>2</sub>, sulphur oxides SO<sub>x</sub>/SO<sub>2</sub>, CO, CO<sub>2</sub> and particulate matter. The estimate of harmful substance emissions from the thermal power plant covers the period 2007-2015, so that the level of emissions from the moment the IPPC permit of *Deven Power Plant* (No 93/2006) came into force, until now, can be traced [22]. Data have been used from the National PRTR Information System [23], and of pollutants which do not exceed the thresholds during the nine-year period of the survey, as well as harmful substance emissions from the emitting devices, the annual reports on the execution of the activities for which the IPPC permit was given, have been used [24]. The results have been statistically processed with the variation analysis method, and the differences estimated with A. Fisher's Student's t-distribution criterion.

## III. RESULTS AND DISCUSSION

The results from the pollutants reported to the PRTR show that the nitrogen oxides NO<sub>x</sub>/NO<sub>2</sub> in 2007 (immediately after the IPPC permit came into force) were 9169479 kg/yr and significantly exceeded the emission threshold (100000 kg/yr) [2]. Over the next year, NO<sub>x</sub>/NO<sub>2</sub> emissions gradually fell to 7919908 kg/yr and are significantly lower between 2009 and 2015, varying between 2783658 and 3574822 kg/yr, but still exceed the threshold (fig.1).

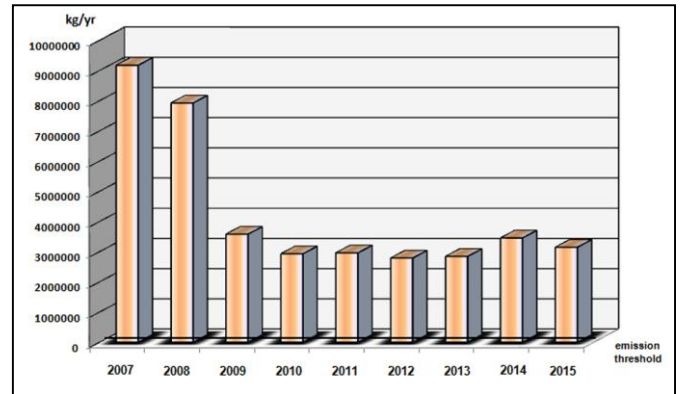


Fig. 1. Emissions of NO<sub>x</sub>/NO<sub>2</sub>, kg/yr

Sulphur oxides share the same trend. SO<sub>x</sub>/SO<sub>2</sub> emissions are the highest at the beginning of the survey period (2007) – 7782486 kg/yr and exceed significantly the emission threshold (150000 kg/yr) (fig.2). Over the following years a decreasing trend is observed in the quantities reported in the national information system: 5492188 kg/yr (2008); 3039675 kg/yr (2009) and 2547900 kg/yr (2010). After 2010, SO<sub>x</sub>/SO<sub>2</sub> emissions are even lower, with just a few exceptions, varying between 1752799 and 1847111 kg/yr. Notwithstanding the trend of decrease, the emission threshold is still significantly exceeded.

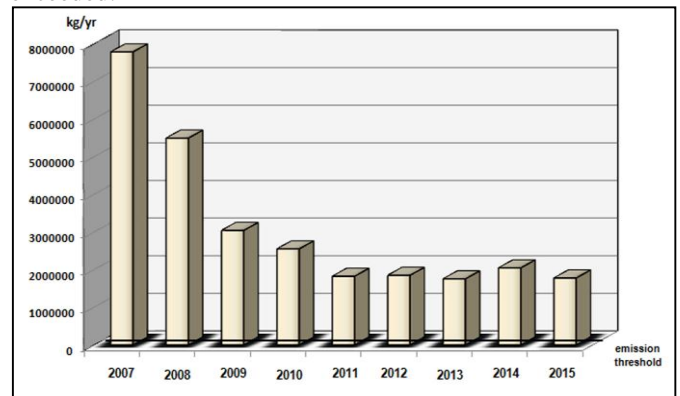


Fig. 2. Emissions of SO<sub>x</sub>/SO<sub>2</sub>, kg/yr

Carbon dioxide, though, falls into a different trend. The reported CO<sub>2</sub> quantities vary very little within the nine-year survey period and stay between 1454781000 and 1685183000 kg/yr, with the exception of 2009 (925045000 kg/yr), exceeding significantly the emission threshold (100000000 kg/yr) (fig.3).

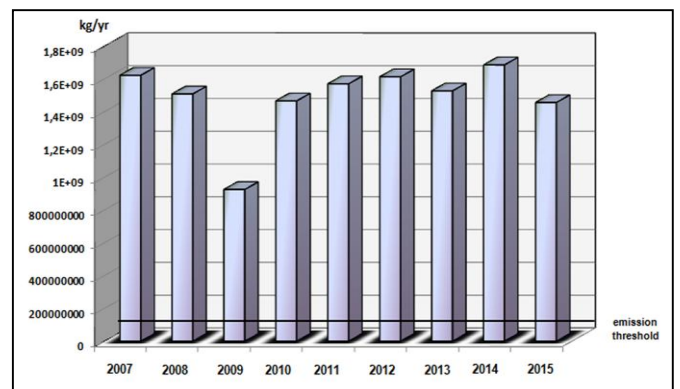


Fig. 3. Emissions of CO<sub>2</sub>, kg/yr

With particulate matter, a favourable trend is observed. In 2009, the reported quantities of PM are 582149 kg/yr and exceed the emission threshold (50000 kg/yr) (fig.4). Over the following year, a sharp fall in pollution is observed – up to 159406 kg/yr, which, with small variations, was maintained until 2014. It is important to note that in 2015 PM emissions fall below the threshold – 38383 kg/yr.

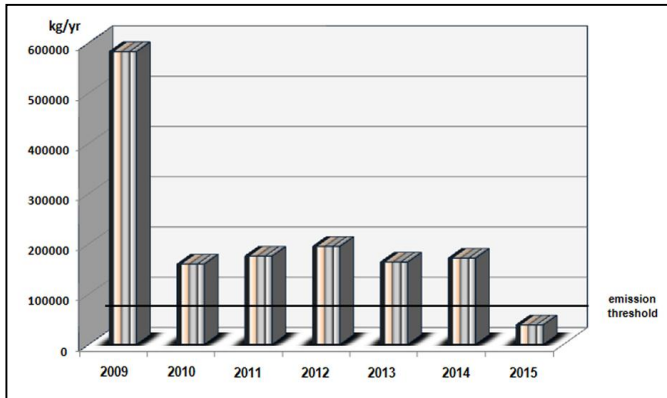


Fig. 4. Emissions of PM, kg/yr

As for carbon oxide emission, the released quantities are under the emission threshold (500000 kg/yr) and *Deven* Power Plant does not report them to the National PRTR Information System. The annual reports for the performance of the IPPC permit activities show that in 2010, CO emissions are 110837 kg/yr, rising slightly to 219090 kg/yr (2012) and going down to 103440 kg/yr (2015), not exceeding the threshold (fig.5).

The results from the continuous monitoring of pollutants in flue gas from the emitting devices at *Deven* Power Plant show that the concentrations of nitrogen oxides NO<sub>x</sub>/NO<sub>2</sub> from steam generators SG<sub>1</sub>-SG<sub>6</sub> into a common chimney were the highest in 2008 (immediately after the coming into force of the IPPC permit) – 1140.77 mg/Nm<sup>3</sup>, but do not exceed the emission limit value under IPPC permit (1200 mg/Nm<sup>3</sup>). Over the following years, the concentrations of NO<sub>x</sub>/NO<sub>2</sub> were lower – 923.98 mg/Nm<sup>3</sup> in 2009; 620.81 mg/Nm<sup>3</sup> (2010); 557.72 mg/Nm<sup>3</sup> (2012); 684.77 mg/Nm<sup>3</sup> (2015), and the differences as compared to the starting levels have high statistical significance (0.001 ≤ P ≤ 0.025) (fig. 6).

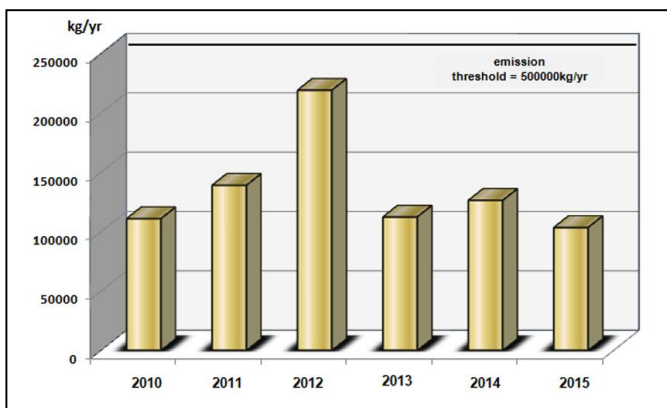


Fig. 5. Emissions of CO, kg/yr

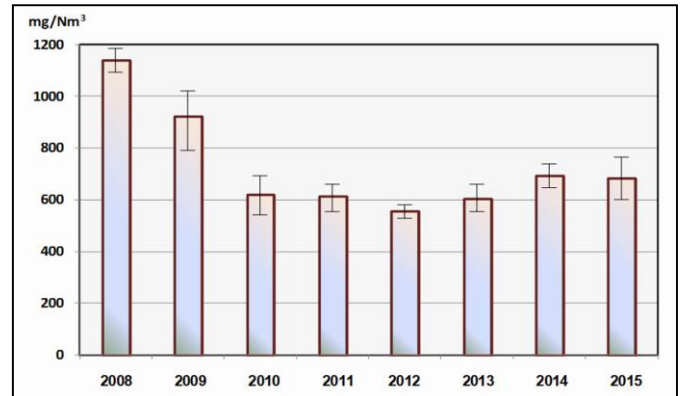


Fig. 6. NO<sub>x</sub>/NO<sub>2</sub> emissions from SG<sub>1</sub>-SG<sub>6</sub> into a common chimney, mg/Nm<sup>3</sup>

Fig. 7 shows that sulphur oxides concentrations from steam generators SG<sub>1</sub>-SG<sub>6</sub> into a common chimney were at their highest at the beginning of the survey 2008 – 795.90 mg/Nm<sup>3</sup>, and exceed the emission limit value under IPPC permit (400 mg/Nm<sup>3</sup>) approximately twice. Over the following year SO<sub>x</sub>/SO<sub>2</sub> concentrations were also high – 781.63 mg/Nm<sup>3</sup>, exceeding the emission limit value 1.95 times. After 2009, SO<sub>x</sub>/SO<sub>2</sub> concentrations gradually fall to 379.77 mg/Nm<sup>3</sup> (2011); 372.63 mg/Nm<sup>3</sup> (2013) and 387.43 mg/Nm<sup>3</sup> (2015), and the differences have high statistical significance (0.001 ≤ P ≤ 0.01).

The CO results show relatively constant no-variation pollution between 2008 and 2011, with concentrations from 806.23 to 886.93 mg/Nm<sup>3</sup>, which does not exceed the emission limit value under IPPC permit (1000 mg/Nm<sup>3</sup>) (fig.8). After 2011, CO concentrations gradually fell to 746.01 mg/Nm<sup>3</sup> (2012) and 684.31 mg/Nm<sup>3</sup> (2014), and the differences have high statistical significance (0.001 ≤ P ≤ 0.05).

From the results in Fig. 9 we can see that concentrations of dust from SG<sub>1</sub>-SG<sub>6</sub> in a common chimney were much higher at the beginning of the survey (2008) – 373.81 mg/Nm<sup>3</sup>, exceeding the emission limit value under IPPC permit (50 mg/Nm<sup>3</sup>) 7.5 times. Over the following year the concentrations gradually fell to 188.95 mg/Nm<sup>3</sup> and the emission limit was exceeded 3.8 times. After 2010, concentrations of dust are under the threshold and vary from 40.84 to 46.33 mg/Nm<sup>3</sup> and the difference from the start in 2008 has high statistical significance (P < 0.001).

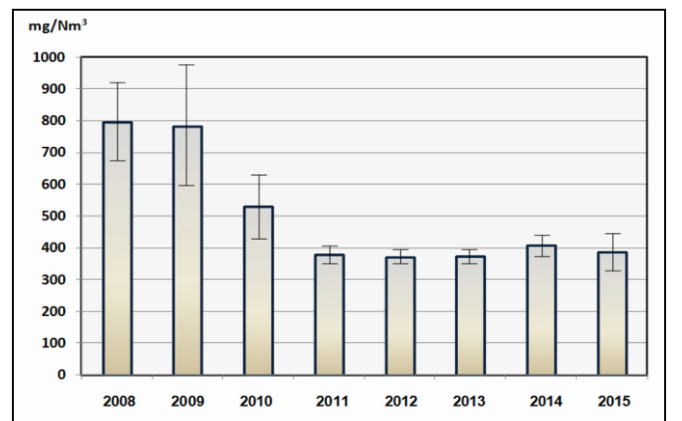


Fig. 7. SO<sub>x</sub>/SO<sub>2</sub> emissions from SG<sub>1</sub>-SG<sub>6</sub> into a common chimney, mg/Nm<sup>3</sup>

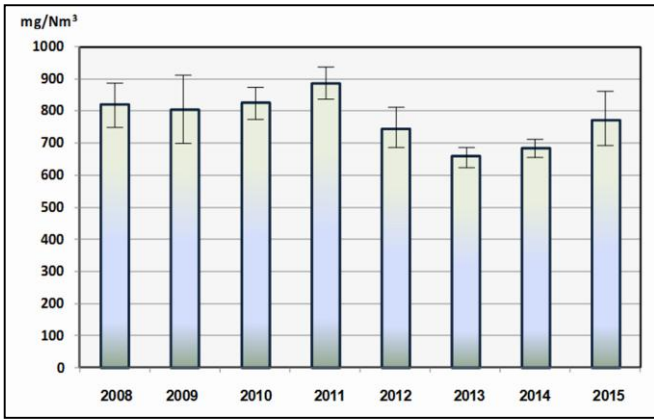


Fig. 8. CO emissions from SG<sub>1</sub>-SG<sub>6</sub> into a common chimney, mg/Nm<sup>3</sup>

The data for the steam generator with a circulating boiling layer CBL-SG show that the NO<sub>x</sub>/NO<sub>2</sub> concentrations were the highest during the first year of the introduction (2010) – 242.53 mg/Nm<sup>3</sup>, exceeding the emission limit value under IPPC permit (200 mg/Nm<sup>3</sup>) 1.21 times. During the following year the nitrogen oxides concentrations fell to 179.29 mg/Nm<sup>3</sup> (P < 0.001) and by the end of the survey, they got within the 166.53-180.05 mg/Nm<sup>3</sup> range (fig.10).

Sulphur oxides share the same trend. At the onset of the CBL-SG the SO<sub>x</sub>/SO<sub>2</sub> concentrations were the highest – 282.15 mg/Nm<sup>3</sup>, exceeding the emission limit value under IPPC permit (200 mg/Nm<sup>3</sup>) 1.41 times. After 2010, the concentrations fell to 159.06 mg/Nm<sup>3</sup> (2012) (P < 0.01) and to 130.23 mg/Nm<sup>3</sup> in 2014 (P < 0.001) (fig.11). In 2015, SO<sub>x</sub>/SO<sub>2</sub> concentrations rose slightly to 153.92 mg/Nm<sup>3</sup>, but did not exceed the emission limit value.

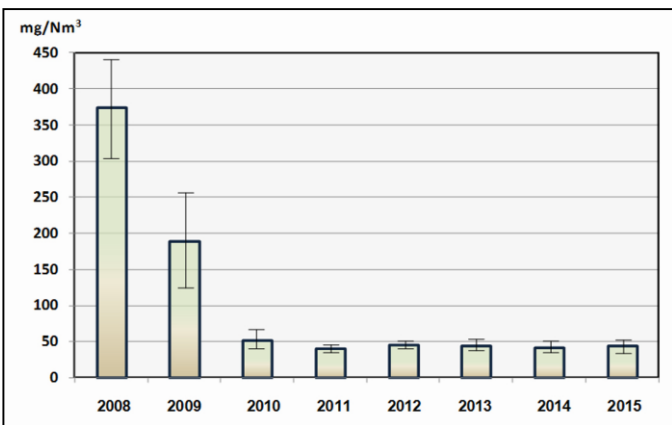


Fig. 9. Dust emissions from SG<sub>1</sub>-SG<sub>6</sub> into a common chimney, mg/Nm<sup>3</sup>

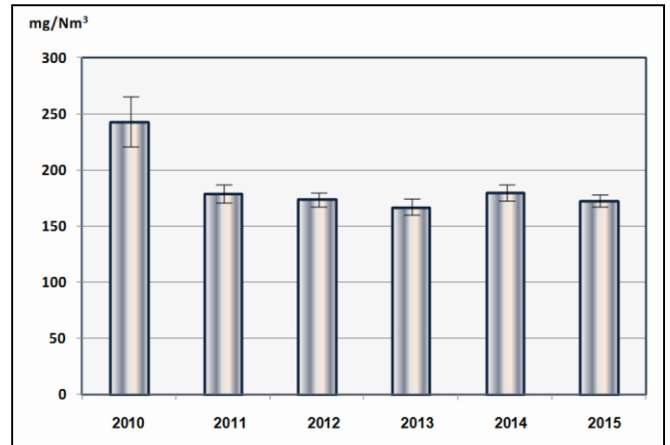


Fig. 10. NO<sub>x</sub>/NO<sub>2</sub> emissions from CBL-SG, mg/Nm<sup>3</sup>

From the results in Fig. 12, we can see that the CO concentrations are significantly lower than the emission limit value under IPPC permit (250 mg/Nm<sup>3</sup>) for the whole survey period. The CO levels are the highest at the onset of the study (2011) – 31.55 mg/Nm<sup>3</sup> and fell gradually, reaching in 2014 their lowest value – 10.88 mg/Nm<sup>3</sup>. The decrease in the concentrations has statistically veritable differences (0.001 ≤ P ≤ 0.01).

The dust results do not reveal any clear cut trend, but rather a wavy one. The concentrations vary slightly – alternating lower with higher ones and are significantly lower than the emission limit value under IPPC permit (30 mg/Nm<sup>3</sup>). In 2010, dust concentrations were 6.83 mg/Nm<sup>3</sup>, falling to 1.07 mg/Nm<sup>3</sup> in 2011, growing in 2012 to 9.86 mg/Nm<sup>3</sup>, falling to 9.14 mg/Nm<sup>3</sup> in 2013, growing to 14.90 mg/Nm<sup>3</sup> in 2014 and yet again falling to 11.48 mg/Nm<sup>3</sup> in 2015 (fig.13).

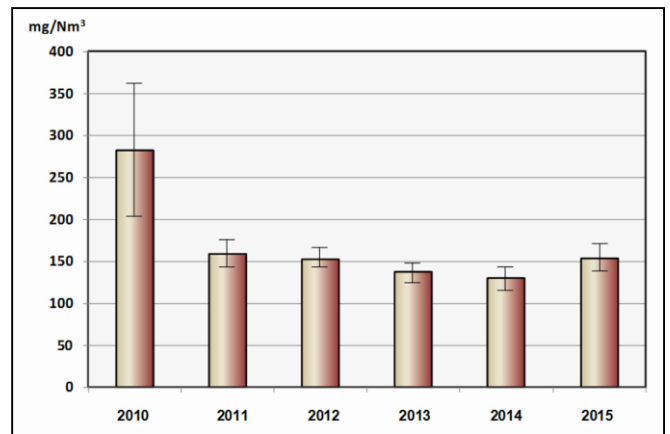


Fig. 11. SO<sub>x</sub>/SO<sub>2</sub> emissions from CBL-SG, mg/Nm<sup>3</sup>

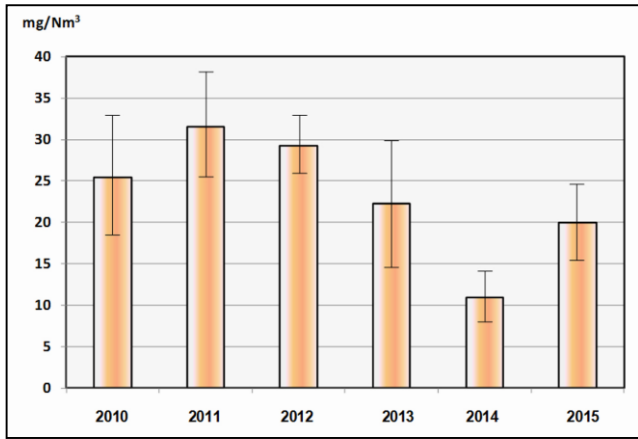


Fig. 12. CO emissions from CBL-SG, mg/Nm<sup>3</sup>

When reviewing the surveys of other authors of coal-fired small capacity thermal power plants (2 units with output capacity of 25 MW each), high concentrations of the basic pollutants have been established – 360 mg/Nm<sup>3</sup> for SO<sub>2</sub>, 237 mg/Nm<sup>3</sup> for NO<sub>x</sub>, 218 mg/Nm<sup>3</sup> for CO and 179 mg/Nm<sup>3</sup> for PM [25]. The most important parameter for the climate change, carbon dioxide, varies in the flue gas from smaller thermal power plants from 11.45 to 13.67%.

In order to reduce SO<sub>2</sub> emissions from coal-fired thermal power plants, sulphur purification installations for flue gas are fitted, and for particulate matter PM<sub>10</sub> and PM<sub>2.5</sub> – effective electrostatic filters [16, 20, 26]. In this way PM concentrations go down 30-40%, thus eliminating the formation of secondary sulphates and nitrates [12]. One of the options to reduce emissions is the more wide-spread use of natural gas as fuel. Harmful substance emissions from combined cycle natural gas thermal power stations are much lower than coal-fired thermal plants – for NO<sub>x</sub> with 40%, for SO<sub>2</sub> with 44%, and for CO<sub>2</sub> – with 22% [27]. These benefits for the improvement of ambient air quality and climate have to be compared against the increase of CH<sub>4</sub>, NMVOC and other emissions connected with the production, processing, storage and transport of natural gas.

Some authors offer suitable measures for the reduction of CO<sub>2</sub> emissions as a change in the fuel base, improvement in heating processes, changes in the permit for the activity of existing power plants, in order to limit emissions for given periods of time [28]. Some political approaches are also offered for reducing CO<sub>2</sub> emissions, such as applying stricter standards for CO<sub>2</sub> emission reduction from coal-fired thermal power plants, as well as installing pollution control systems [11, 29].

A very important factor for reducing harmful substance emissions is the combined production of electrical and heat energy. Such a cycle does not only reduce gaseous emissions of NO<sub>x</sub>/NO<sub>2</sub>, SO<sub>x</sub>/SO<sub>2</sub>, CO, CO<sub>2</sub> and particulate matter, but it also releases about 10 times less ash [13].

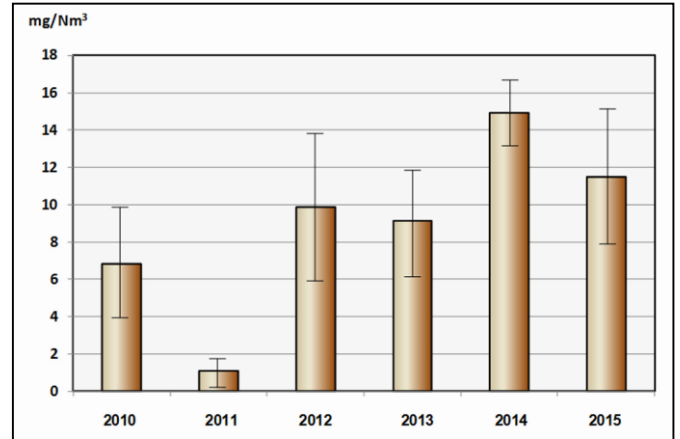


Fig. 13. Dust emissions from CBL-SG, mg/Nm<sup>3</sup>

The *Deven* Power Plant combustion installation for combined production of heat and electricity (cogeneration system) is defined as the most important option in the best available methods for reduction of greenhouse gas emissions. Cogeneration is the only sensible way for supplying the energy needed for the industry and especially for the chemical reactions in the production of synthetic industrial ash soda in the region of Devnya. In 2010, a new type steam generation was introduced, the No 7, with a circulating boiling layer. The new type of boiler is characterised by the fact that the fuel constantly circulates through the furnace and the separators with the aim of increasing the time the particles stay in the furnace, which allows for burning at a lower than usual temperatures. The result is an environmentally beneficial installation with very low emissions of nitrogen oxides (< 200 mg/Nm<sup>3</sup>), sulphur oxides, owing to adding limestone (< 200 mg/Nm<sup>3</sup>), very little dust emissions (< 30 mg/Nm<sup>3</sup>), simple construction, reliability, perfect efficiency, allowing for freedom and opportunities for burning a wide range of hard fuels, which allows for the better use of the primary energy resources.

#### IV. CONCLUSIONS

The existing cogeneration installation at *Deven* Power Plant allows for the achievement of utilisation of the energy from the fuel of up to 80%. The installation has been fully modernised and optimised with the introduction into operation at the end of 2009 of a new boiler with circulating boiling layer. The circulating boiling layer boilers have a number of advantages as compared to the conventional ones common within the network of the Bulgarian energy producing system. They have a wide range of fuel utilisation. The technology allows for the burning of many different types of fuels – black and lignite coal, petcoke, biomass, etc., as well as a calorific value of 2500 to 7500 kcal. They also cost less to operate and maintain. The boiler ensures a better mixture and recirculation of the boiling layer, thus guaranteeing a higher burning efficacy and lower emissions. An additional system for control of NO<sub>x</sub> has also been introduced. The developed scheme for cogeneration production of heat and electricity at *Deven* Power Plant achieves a high level of utilization of the energy from the fuels and a continuous enhancement of the environmental conditions via a controlled use of natural resources and limiting the amount of emissions into the atmosphere.

As a result of the measures appointed in the IPPC permit of *Deven* Power Plant and the introduction into operation of a steam generator with a circulating boiling layer, a reduction in the released quantities of NO<sub>x</sub>/NO<sub>2</sub>, SO<sub>x</sub>/SO<sub>2</sub> and PM for 2007-2015 was observed, and the PM emissions in 2015 were under the threshold. A decreasing trend is delineated in harmful substance concentrations from the emitting devices – not just from the CBL-SG but also from SG<sub>1</sub>-SG<sub>6</sub> into the common chimney, and the differences have high statistical significance ( $0.001 \leq P \leq 0.05$ ).

Other research of ours has established that between 2006 and 2007 (at the start of the application of the IPPC permits) for the industrial and combustion installations in the region of Devnya, the concentrations of a large part of the pollutants of the air significantly exceeded the limit values to protect human health [30]. At a later stage, after the implementation of the measures for reducing the release of harmful substances from the installations for the production of nitrogen and phosphorous fertilisers, as well as soda ash, the levels of the pollutants of ambient air in the industrial region of Devnya gradually started to fall [31-33].

The survey shows that the implementation of the IPPC Directive and the execution of the activities for integrated pollution prevention and control of large combustion installations is a necessary prerequisite for the reduction of emissions of harmful substances and the improvement of the quality of ambient air in the region.

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