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Assessment of Measures for Reducing Harmful Emissions in Air from Soda Ash Producing Plant in Devnya, Bulgaria

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Abstract—The research has been done at Solvay Sodi JSC plant for the production of synthetic soda ash situated in the industrial region of Devnya. The paper deals with the assessment of emissions of harmful substances (CO, CO_2, NH_3, NO_x/NO_2, SO_x/SO_2, H_2S and PM_{10}) from the Soda Ash plant(Solvay Sodi JSC) after coming into force of the Integrated Pollution Prevention and Control (IPPC) Permitin 2006. Significant emission reduction is registeredforsome pollutants (CO, NH_3, SO_x/SO_2) during the period from 2006 to 2013, despite the fact that some annual emission levels considerably exceed the emission threshold established by the European Pollutant Release and Transfer Register (E-PRTR). The concentrations of pollutants do not exceed the limit values during the monitoring period. Appropriate measures for reducing the emission levels were appliedpursuant to the IPPC Permit for the Soda Ash plant.It isascertainedthat applying best available techniques (BAT) andstrictly complying the requirements of the IPPC Permit contribute to the low levels of emissions and improve the ambient air quality in the region.

Keywords—Air pollution, best available techniques (BAT),emission,industrial region Devnya,IPPC Permit, plants.

I. INTRODUCTION

Directive 2008/1/EC of the European Parliament and of the Council of 15 January 2008 concerning Integrated Pollution Prevention and Control and the referent Bulgarian Regulation,concerning procedures and conditions for issuing IPPC Permits, SG No 80/2009, amendments and supplements SG No 69/2012,establish the conditions and order for issuance of IPPC Permits for construction and operation of new and operation of already existing plants and facilities for various activity categories. The IPPC Permit requires the application of best available techniques (BAT) in order to reduce the adverse impact of plants upon the ambient air. Operators of industrial and combustion installations should apply appropriate measures, such as: modernization of technological processes and introduction of technologies with waste gas utilization; implementation of environmentally friendly technologies (best production practices); diversification of raw materials in order to reduce harmful emissions; introduction of new waste gas treatment facilities or upgrading the existing ones;application of appropriate technologies for waste gas treatment and reduction of emissions in the ambient air, etc.

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Many environmental documents, concerning soda ash production, establish the necessity of applying BAT at production plants in order to ensure reduction of the emission levels of major pollutants, such as CO, CO_2, SO_x, NO_x, NH_3 and PM_{10}, and improvement of ambient air quality and the health condition of the exposed population [1]-[7]. The paper is aimed at monitoring the emissions of harmful substances during the production of soda ash and assessing the emission reduction measurespursuant to the IPPC Permit for improving the ambient air quality.

II. MATERIAL AND METHODS

The study is carried out at the Solvay Sodi JSC plant for soda ash production, which is situated in the industrial region of Devnya, in the northeastern part of Bulgaria.The IPPC Permit No 74 of 27 December 2005 of Solvay Sodi JSC came into force on 14 January 2006. Major pollutants from the soda ash plant to be reported in the European Pollutant Release and Transfer Register (E-PRTR) are CO, CO_2, NH_3, nitrogen oxides NO_x/NO_2 and oxides of sulphurSO_x/SO_2. National information system has been developed for reporting pollutant release and transfer pursuant to the E-PRTR [8] complying the requirements of [3], [4]. The European Union member states are required to report the release of pollutants from all activities under Annex I of the Regulation, which includes 65 activities grouped into 9 sectors. In Annex II of the Regulation, 91 pollutants have been included, as limit value (threshold) for each of them has been defined. The study covers the period from 2006 to 2013 after coming into force of the IPPC Permit. The period is defined in order to assess the effect of applying measuresrequired by the Permit to ensure compliance with the emission limit values for various pollutants from point sources. The annual reports for implementation of the activities, for which the IPPC Permit of Solvay Sodi JSC is issued, have been used as data source [11].Data on annual emissions (kg/y) of major pollutants are used, as well as data from periodic monitoring of various harmful substances in waste gases for the entire period of observation (mg/Nm^3). The monitoring data on the major pollutants (NH_3, NO_x/NO_2, SO_x/SO_2, H_2S and PM_{10}) have been statistically processed by the means of variation analysis as differences have been estimated by using t-criterion of Student Fisher.

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III. RESULTS AND DISCUSSION

Solvay Sodi JSC is a producer of synthetic soda ash (*dense* and *light*) and sodium bicarbonate as a by-product for industrial and food processing consumers. The plant's capacity is 1.5 million tons soda ash light, 1.3 million tons soda ash dense and 30 000 tons sodium bicarbonate per year. The research results indicate that emissions of carbon monoxide CO, one of the main pollutants from the plant, considerably exceeds the emission threshold (500 000 kg/y) according to E-PRTR for the entire period of observation and varies from 13 087 000 to 25 091 520 kg/y (Fig.1). During the 2006 – 2008period right after the enforcement of the IPPC Permit, the emission levels of CO are much higher- from 23 950 580 to 25 091 520 kg/y. In 2009, there was a significant emission drop, probably due to decreased soda ash production (835.9 thousand t/y), compared to the previous years (Table I). During the2010 – 2013 period,the production capacity is increased, but the emission levels of CO decreased from 23 222 150 kg/y in 2010 to 13 087 000 kg/y in 2013. The significant emission drop registered after 2009 is due to renovation of the soda ash plant equipment – renovation of lime kilns, which are major source of carbon monoxide emissions, reconstruction of the kilns housing, installation of a new feeding device and replacement of scrubbers and electro-filters.

Table I
Production (thousand t/y) of synthetic soda ash for the 2006 -2013 period

Year / Product	2006	2007	2008	2009	2010	2011	2012	2013
Light	1172.5	1220.3	1254.6	835.9	1251.2	1452.3	1448.6	1350.9
Dense	790.9	841.6	859.6	610.7	954.6	1177.6	1264.2	1019.6
Sodium bicarbonate	20.3	20.7	21.6	16.5	20.2	20.8	22.8	25.1

The results from periodic monitoring of emissions from a scrubber at lime kilns indicate that,after the enforcement of IPPC Permit (2006), the concentrations of CO vary from 1256 to 6250 mg/Nm^3 and decrease significantly in 2012 to1189.7 mg/Nm^3, and even lower to 749 mg/Nm^{3}in 2013.

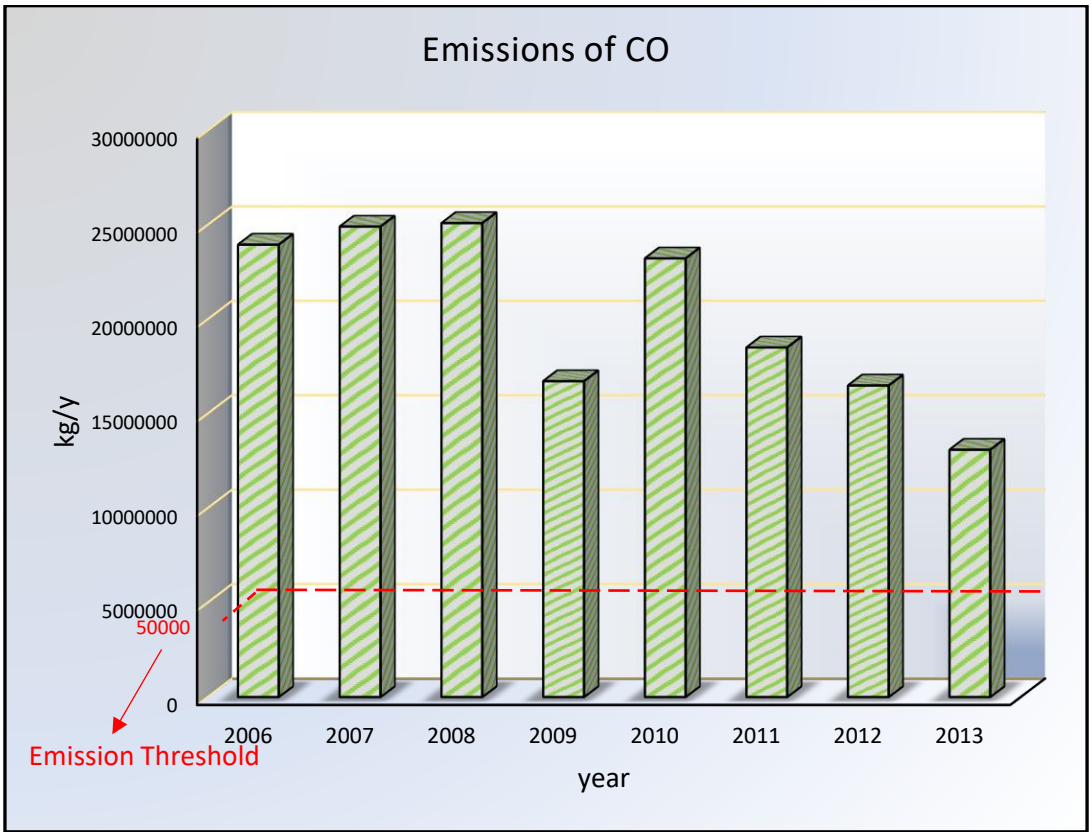

Fig. 1. Total Emissions of CO, kg/y

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Carbon dioxide CO_2 is a main pollutant from soda ash production, which is emitted from combustion of coke and decomposition of limestone[9]. Fig. 2 indicates that CO_2 emissions do not vary considerably during the monitoring period- from 460 629 523 kg/y to 533 197 270 kg/y. CO_2 emission drop in 2009 (296 439 368 kg/y) is due to decreased production capacity of the plant. Monitoring results indicate that the emission threshold for CO_2 (100 000 000 kg/y) has been significantly exceeded for the period after the enforcement of the IPPC permit. Such high emission levels require not only annual inventory of CO_2emissions, but also a short-term assessment of greenhouse gas emissions, pursuant to environmental legislation [10]-[14], [15] and applying measures for additional emission reduction.

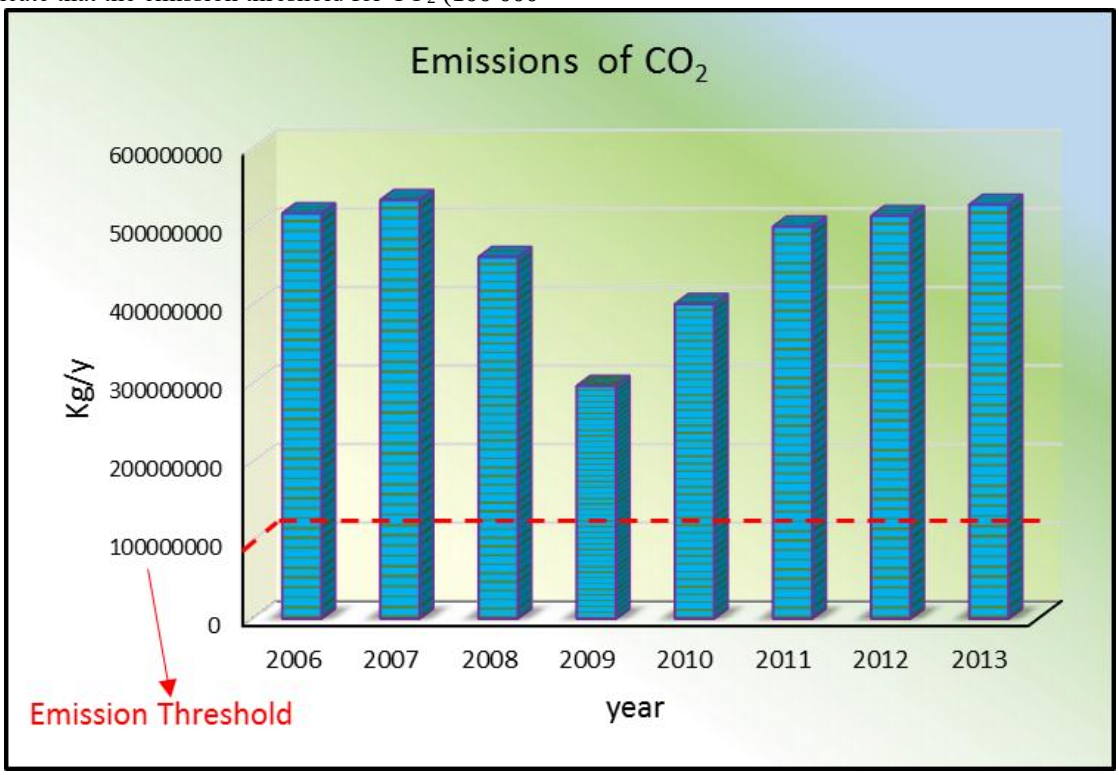

Fig. 2. Total Emissions of CO_2, kg/y

The major ammonia NH_3 emissions from soda ash production are caused by the precipitation of sodium bicarbonate and the filtration process. NH_3 emissions exceed significantly the emission threshold (10 000 kg/y), especially at the beginning of the period when the activities under the IPPC Permit started (2006-2007). The highest emission levels are measured in 2007 - 1 513 307 kg/y (Fig.3). In 2008 a considerable drop of NH_3 emissions is registered – 559 604 kg/y, which is almost 3 times lower than NH_3 emissions in 2007. From 2008 onwards, annual emission levels vary between 460 562 kg/y (2009) and 711 203 kg/y (2012). NH_3 emissions are significantly lower than that in 2007 onwards compared to those at the beginning of period after the IPPC Permit enforcement.

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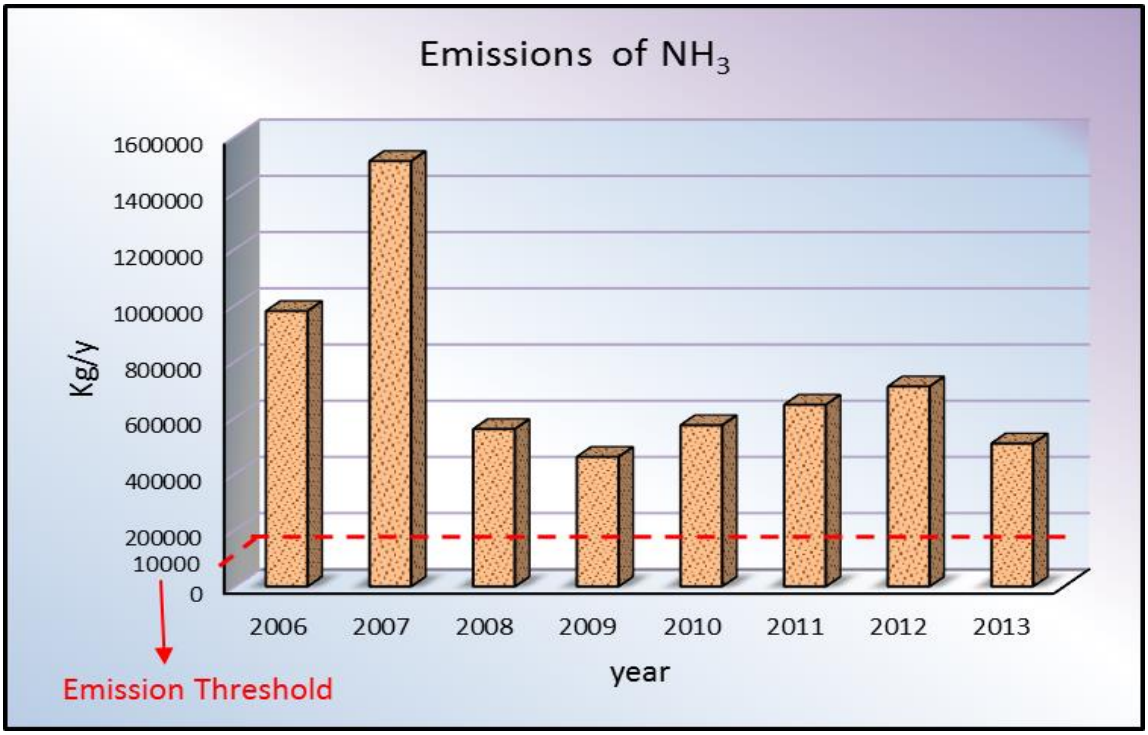

Fig. 3. Total Emissions of NH_3, kg/y

Monitoring results for NH_3 emissions from Absorption and Distillation section (Fig. 4) indicate that NH_3 concentrations in 2006 are 19.75 mg/Nm^3 and decrease gradually after the enforcement of IPPC Permit to 16.40 mg/Nm^{3}in 2013, as the differences are of strong statistical significance ($P < 0.05$). It is indicated that NH_3 concentrations do not exceed the emission limit value (ELV= 50 mg/Nm^3).

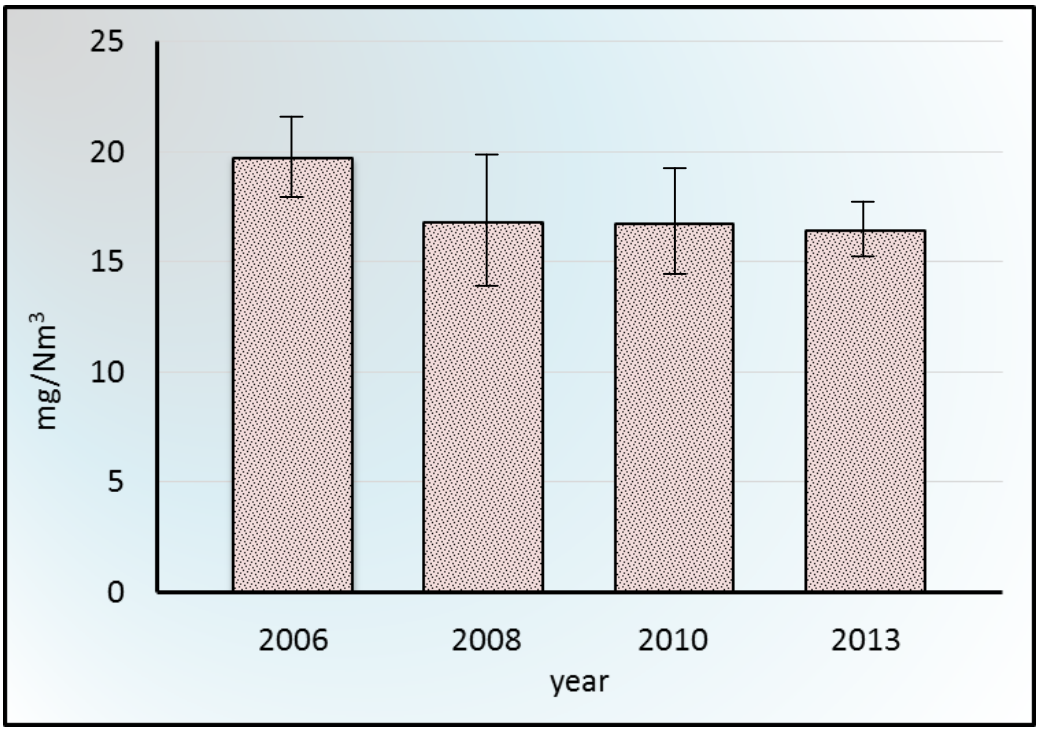

Fig. 4. Concentrations of NH_3 in waste gases from Absorption and Distillation Section

The emission levels of nitrogen oxides NO_x/NO_2 for the monitoring period are shown on Fig.5. A slight decrease trend of annual NO_x/NO_2 emissions is indicated from 2007 onwards – from 159 078 kg/y to 122 142 kg/y in 2012. The lowest emission levels are measured in 2009 – 97 880 kg/y, due to decreased production capacity. The emission threshold (100 000 kg/y) hasn't been exceeded in 2009only.

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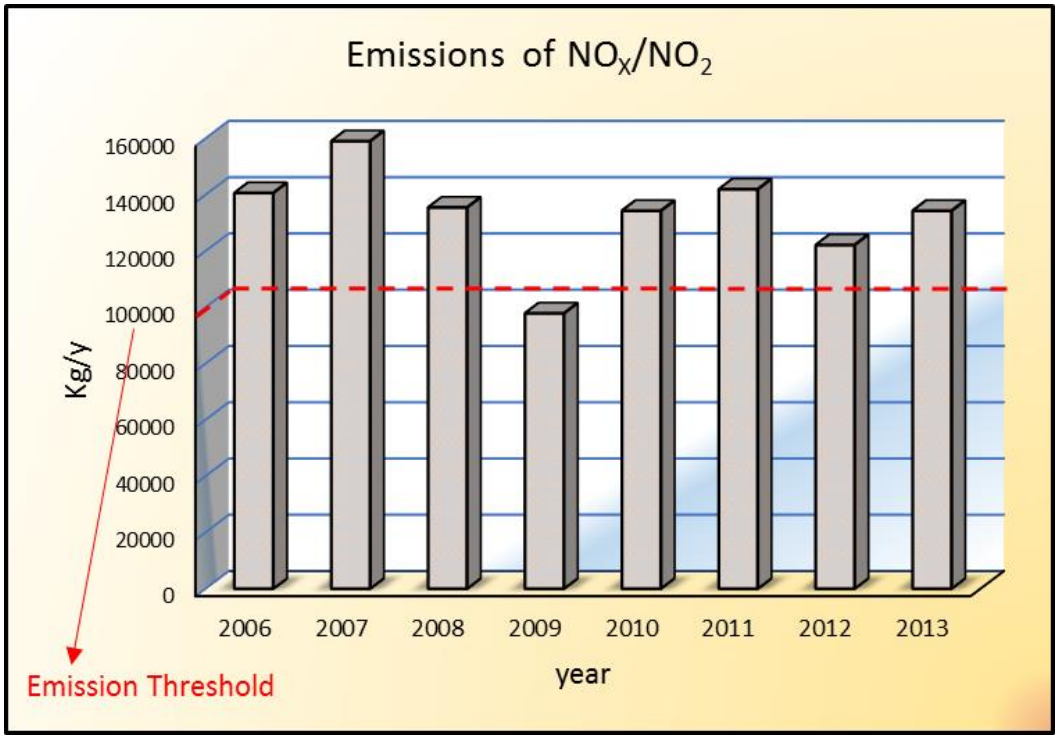

Fig. 5. Total Emissions of NO_x/NO_2, kg/y

Monitoring results for NO_x/NO_2 emissions from Absorption and Distillation section indicate that concentrations of NO_x/NO_2 are 159.35 mg/Nm^3 in 2006 (Fig. 6) and decrease gradually to 123.31 mg/Nm^3 in 2011, and to 111.24 mg/Nm^3 in 2013.The differences are of strong statistical significance ($P < 0.001$). During the 2008 – 2011 period, the decreasing NO_x/NO_2 concentrations are also of strong statistical significance ($0.002 \leq P \leq 0.05$). No registered concentrations of NO_x/NO_2 exceed the limit value for the entire monitoring period (500 mg/Nm^3 in accordance with the IPPC Permit till 2011 and 400 mg/Nm^3 afterwards).

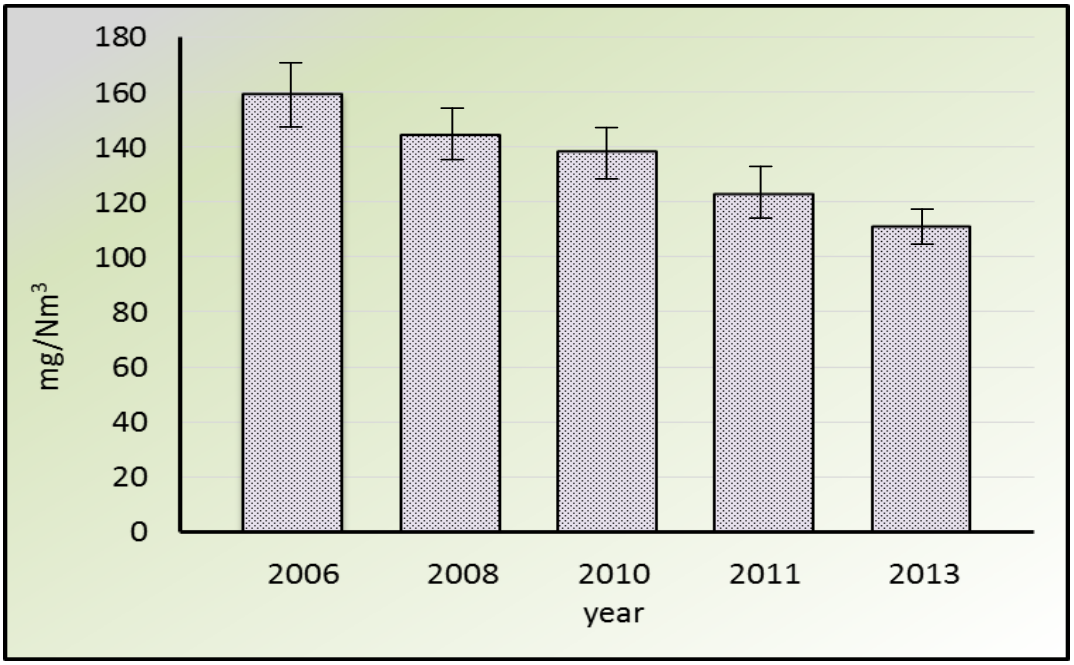

Fig. 6. Concentrations of NO_x/NO_2 in waste gases from Absorption and Distillation Section

Emissions of oxides of sulphurSO_x/SO_2 from synthetic soda ash production result from oxidation of sulphur-containing compounds in limestone and coke. The annual SO_x/SO_2 emissions for the entire period of research are below the emission threshold (150 000 kg/y). From 2009 onwards, a considerable emission drop is registered (Fig.7). During the 2006-2009 period the emission levels are close to 130 000 kg/y. In 2010 significantly less SO_x/SO_2 emissions have been declared – 57 013 kg/y, and in 2011 a minimum value of SO_x/SO_2 emissions is registered – 48 485 kg/y.

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Results from periodic monitoring SO_x/SO_2 emissions from lime production indicate that after the enforcement of IPPC permit (2008) concentrations of SO_x/SO_2 are 163.42 mg/Nm^3, decrease gradually to 93.58 mg/Nm^3 in 2011 and to 66.50 mg/Nm^3 in 2013 (Fig. 8). The differences are of strong statistical significance ($0.001 \leq P \leq 0.05$). The concentration limit values for SO_x/SO_2 haven't been exceeded (500 mg/Nm^3 until 2012 and 400 mg/Nm^3 for 2013, which isin accordance with the IPPC Permit).

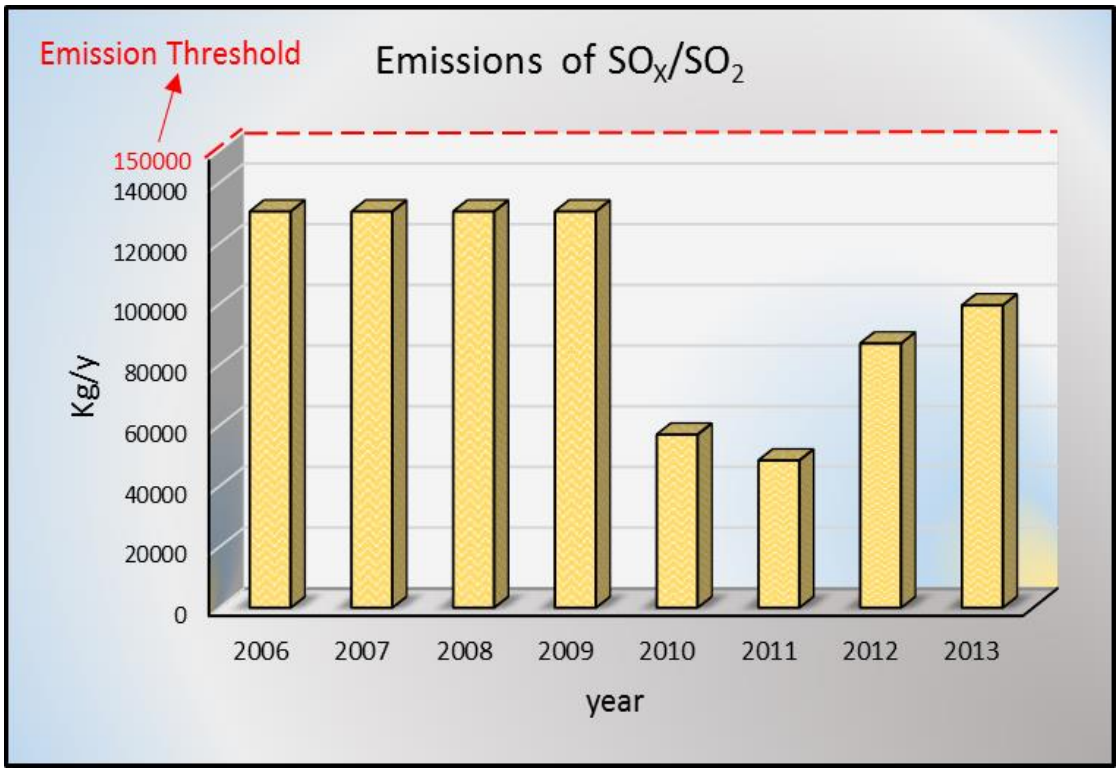

Fig. 7. Total Emissions of SO_x/SO_2, kg/y

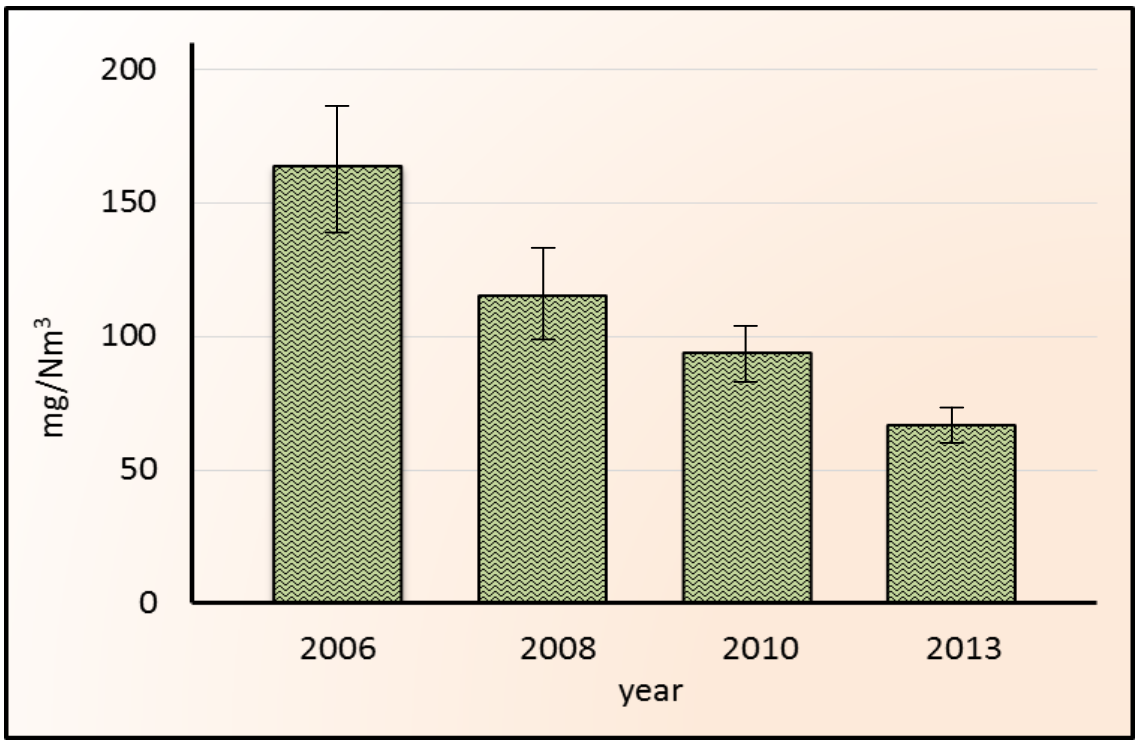

Fig. 8. Concentrations of SO_x/SO_2 in waste gases from Lime production Section

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With regards to the pollution of ambient air by hydrogen sulphide H_2S, the data from periodic monitoring of H_2S emissions from Absorption and Distillation Section indicate that after the enforcement of the IPPC permit (2006) concentrations of H_2S are 4.65 mg/Nm^3 and gradually decrease from 2008 onwards – from 2.87 mg/Nm^3 in 2011 to1.73 mg/Nm^3 in 2013 (Table II). The differences between the measured concentrations of H_2S during different periods are with strong statistical significance ($0.001 \leq P \leq 0.05$). The emission limit values for H_2S, required by the IPPC permit, haven't been exceeded for the period of research (6 mg/Nm^3 until 2012 and 5 mg/Nm^3 in 2013).

Table II
Concentrations(mg/Nm^3) of H_2S in Absorption and Distillation Section, (Average value ± SD)

2006 (1)	2008 (2)	2011 (3)	2013 (4)
4.65 ±0.52	4.79 ± 0.14	2.87 ± 0.40	1.73 ± 0.31
$P_{1.2} > 0.05$	$P_{2.3} < 0.001$	$P_{3.4} < 0.05$	
$P_{1.3} < 0.01$	$P_{1.4} < 0.001$		
$P_{1.4} < 0.001$			

Solvay Sodi JSC performs periodic monitoring of emissions of dust from several sources at Calcination section, but till the end of the monitoring period (2013) it is not required to report the annual emission values of this pollutant, according to the European Pollutant Release and Transfer Register [8].After the enforcement of new IPPC Permit of Solvay Sodi JSC in January 2015, the plant operator should report the annual values of dust emissions pursuant to the E-PRTR and the Guidance for minimum requirements concerning the type, place and content of clauses in the IPPC permits under article 117 of Environment Protection Act[2], [3]. Monitoring results for emissions of dust from a bag filter at Calcination section indicate that concentration of dust in 2006 is 26.33 mg/Nm^3, decreases significantly in 2010 to 16.68 mg/Nm^3 and to 7,02 mg/Nm^3 in 2013 (Fig. 9). The differences are of strong statistical significance ($P < 0.001$). Similar trend is registered regarding concentrations of dust from a scrubber at Calcination section (Fig.10). In 2006, the concentration of dust is 25.98 mg/Nm^3 and gradually decreases to 8.72 mg/Nm^3 in 2013 ($0.01 \leq P \leq 0.05$).The concentration limit values for dust does not exceed 80 mg/Nm^3 till 2011 and 20 mg/Nm^3 for 2012-2013, which is in accordance with the IPPC Permit requirements.

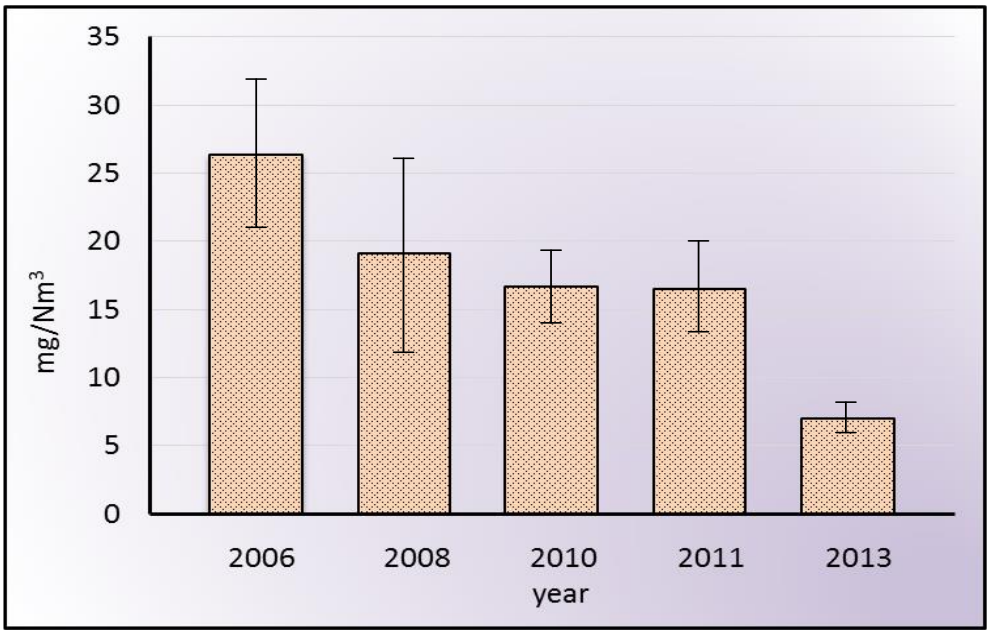

Fig. 9 Concentrations of dust in waste gases from Calcination Section (bag filter)

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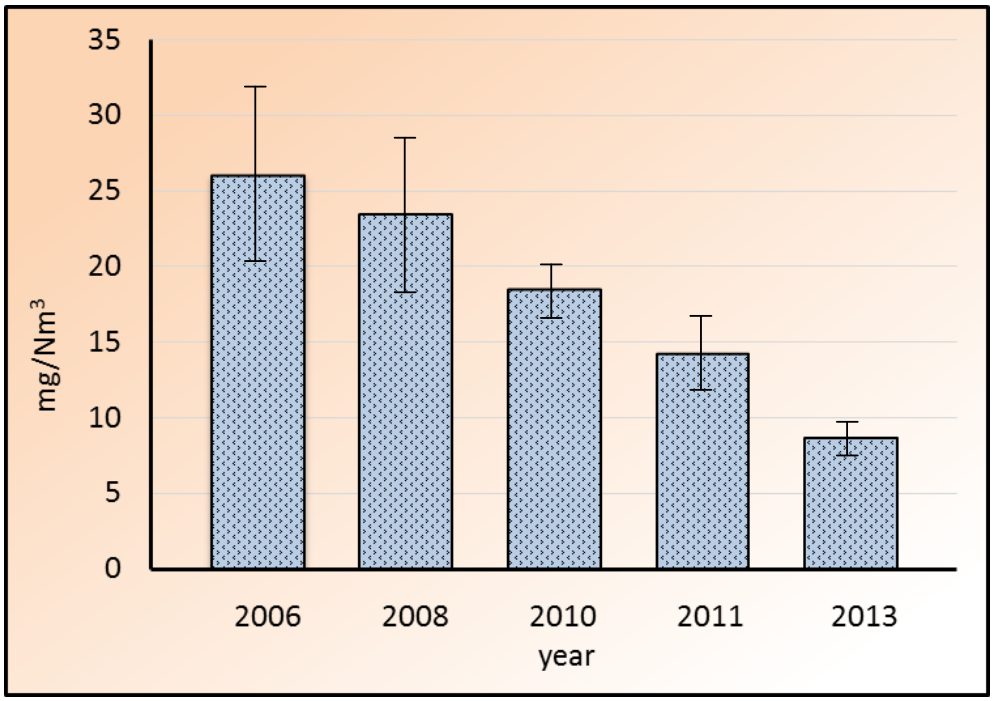

Fig. 10 Concentrations of dust in waste gases from Calcination Section (scrubber)

IV. CONCLUSION

Solid decrease trends regarding the annual emissions of most pollutants from soda ash production were indicated in the 2006 – 2013period (after the enforcement of the IPPC Permit No 74of 27 December 2005 of Solvay Sodi JSC). The registered decrease trends result from the application of emission reduction measures and the introduction of best available techniques at soda ash production. Both actions are required by the IPPC Permit. In 2007, an investment program at Solvay Sodi JSC started for implementing the measures required by the IPPC Permit in order to improve the quality of the ambient air. The main part of the funds is invested for the construction of a new line for dense soda ash production, introduction of a distillation column with low emission and low energy consumption, etc. Another important projects regarding environment protection are designing a new carbonizing column, lime kilns reconstruction, building up the dike of Padina slurry pond, modernization of waste gas treatment facilities: scrubbers and electro-filters. All newly designed facilities, as well as modernized existing equipment comply with best available techniques and environmental standards. The research results prove that implementing the EC Directive for integrated pollution prevention and control throughout enforcement (IPPC) for large industrial plants is a necessary requirement for improvement of ambient air quality. Other research documents provide reasonable conclusion that applying BAT and best production practices at various industrial plants is essential for improvement of ambient air quality at the industrial region of Devnya [13].

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