

# Prognosis Modeling of Nitrous Oxide Emissions after Catalytic Reduction

Maya Stefanova, Rozalina Chuturkova, Evgeni Sokolovski, Nina Ilieva, Tsenislav Vlaknenski

**Abstract** – The present research aims to assess the effect of increasing the thickness of a secondary catalyst layer for  $N_2O$  emission reduction at a nitric acid plant in Devnya, Bulgaria upon the ambient air quality. A mathematical modeling is done for simulating the dispersion of  $N_2O$  emissions from the plant into the ground atmospheric layer taking into account the specific topographic and meteorological conditions of the region. Separate graphic models are done illustrating the dispersion of  $N_2O$  emissions at two main scenarios – at current thickness of the secondary catalyst layer of 60 mm and at future increase of the catalyst layer thickness to 90 mm. Modeling results indicate that under equivalent meteorological conditions the planned increase of the secondary catalyst layer thickness leads to 69 % reduction of the annual average  $N_2O$  concentration in the atmosphere. Maximum  $N_2O$  concentrations over specific periods of time (1 hour, 8 hours and 24 hours) are also reduced over 3 times within the outlines of the exposed areas. Research results provide prognosis on the impact of the increased thickness of the secondary catalyst layer as a measure for  $N_2O$  emission reduction upon the ambient air quality of the source region. Prognosis modeling provides a tool for assessing the contribution of  $N_2O$  emissions from nitric acid production to the overall greenhouse gas emissions in long-term future periods regarding the implementation of quantitative commitments under the Kyoto Protocol.

**Index Terms** - ambient air quality, greenhouse gas emissions, nitrous oxide, prognosis air dispersion modeling, secondary catalyst layer.

## I. INTRODUCTION

Nitric acid production is a major industrial source of nitrous oxide  $N_2O$  emissions, which is a greenhouse gas under the Kyoto Protocol and one of the main reasons for global warming effect [1]-[4]. There are 4 groups of measures developed for  $N_2O$  emission reduction at nitric acid production [5]-[9]. Primary measures are measures that affect the formation of  $N_2O$  during the catalytic oxidization of ammonia like modifying the geometry of the platinum gauze

which can lead to a higher conversion of ammonia to nitric oxide NO and/or a reduction in the formation of  $N_2O$ . Secondary measures are taken with regard to the process gas stream, produced in the process from the oxidisation catalyst, to the absorption tower (homogeneous decomposition, high temperature catalytic decomposition). Tertiary measures can be taken in the process that takes place between the absorption tower and the expansion turbine (low temperature catalytic decomposition, selective catalytic reduction with hydrocarbons, non-selective catalytic reduction of NOx with simultaneous  $N_2O$  reduction). Sequential (end-of-pipe) techniques are some of the techniques described as tertiary measures but placed behind the expansion turbine (selective catalytic reduction, catalytic decomposition). The implementation of a secondary non-platinum catalyst that decomposes  $N_2O$  to nitrogen and oxygen right after its formation in the reactor chamber is a secondary measure for catalytic  $N_2O$  emission reduction [10]-[24]. The efficiency of the reduction depends on multiple factors, most importantly on the specific technological conditions of the production process, the duration of the functional activity of the secondary decomposing catalyst and the thickness of the secondary catalyst layer [25]-[26]. Previous research indicates that catalytic reduction of  $N_2O$  emissions from nitric acid production affects positively the ambient air quality of the source region [27]-[28]. The present research's aim is to determine the prognosis level of additional  $N_2O$  emission reduction due to future increase of the secondary catalyst layer thickness at a nitric acid plant and its effect upon the ambient air quality taking into account the specific topography and meteorological conditions of the source region.

## II. MATERIAL AND METHODS

Within the present research data on  $N_2O$  emissions from a single point source is used – an industrial plant for nitric acid production with capacity 363000 tons per year, situated in the industrial area of Devnya, Bulgaria. A secondary decomposing catalyst for  $N_2O$  emission reduction is implemented at the plant in September 2005 [29]. The catalyst layer is equally disturbed directly on top of the support grid of each reactor separated on top and bottom by steel screens. The thickness of the catalytic layer in the basket is approximately 28 mm. In August 2012 a reconstruction of the ammonia burning reactors is done in order to mount a deeper catalyst basket that increases the catalyst layer thickness to 60 mm [26].

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\* Correspondence Author

Maya Stefanova\*, Department of Ecology and Environmental Protection, Technical University, Varna, Bulgaria.

Assoc. Prof. Rozalina Chuturkova, PhD, Department of Ecology and Environmental Protection, Technical University, Varna, Bulgaria.

Evgeni Sokolovski, PhD, Department of Engineering Ecology, University of Chemical Technology and Metallurgy, Sofia, Bulgaria.

Nina Ilieva, PhD, Department of Engineering Ecology, University of Chemical Technology and Metallurgy, Sofia, Bulgaria.

Tsenislav Vlaknenski, Department of Ecology and Environmental Protection, Technical University, Varna, Bulgaria.

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A mathematical modeling is done that simulates the dispersion of  $N_2O$  emissions from the plant. Due to the fact that  $N_2O$  is inert in the troposphere, all models simulating emission dispersion from area, transport or point sources at urban areas are not applicable [30]-[39]. A practically approved software product BREEZE AERMOD is used within this research for  $N_2O$  emission dispersion modeling, taking into account the topography of the region.

In order to ensure maximum reliability of the meteorological conditions of the region validated data is used in the form of hourly meteorological file containing information on 6 parameters – wind speed and direction, ambient air temperature, relative humidity, atmospheric pressure, sun radiation intensity.

In order to avoid obtaining of wrong results due to variation of the meteorological conditions  $N_2O$  emission dispersion modeling is done over two main scenarios. The first scenario simulates the dispersion of  $N_2O$  emissions from the nitric acid plant at current thickness of the secondary catalyst layer of 60 mm, and the second scenario simulates the prognosis dispersion of  $N_2O$  emissions at future increase of the catalyst layer thickness up to 90 mm. The dispersion of  $N_2O$  emissions at 60 mm thickness of the secondary catalyst layer is simulated using emission data in the form of hourly  $N_2O$  concentration file for 2013 and meteorological data for the same year. The dispersion of  $N_2O$  emissions at planned increase of the thickness of the secondary catalyst layer up to

90 mm is simulated using prognosis data file on the reduced hourly  $N_2O$  concentration [26] and meteorological data for 2013 in order to provide comparable results. Annual average and maximum average  $N_2O$  concentrations over specific periods of time (1 hour, 8 hours and 24 hours) are calculated for both simulated scenarios.

### III. RESULTS AND DISCUSSION

#### A. Topographic characteristic of the region

The nitric acid plant is located in the east industrial area of Devnya, Bulgaria. The terrain is mostly lowland with some hilly areas and the elevation varies between 0 and 300 m. The nearest settlement is Povevlyanovo, a suburb of Devnya (to the north). Other settlements in the area are Razdelna, Beloslav and Strashimirovo with elevation almost equal to the elevation of the source site. Settlements to the north from the source such as Slanchevo (elev. 145 m), Kipra (elev. 135 m) and Banovo (elev. 340 m) can be affected by the emission dispersion. These settlements are situated in hilly areas where displacement is higher or almost equal to the geometric height of the source and for that reason certain meteorological conditions may cause increased pollutant concentrations in the ambient air. Fig. 1 presents a topographic map of the source region.

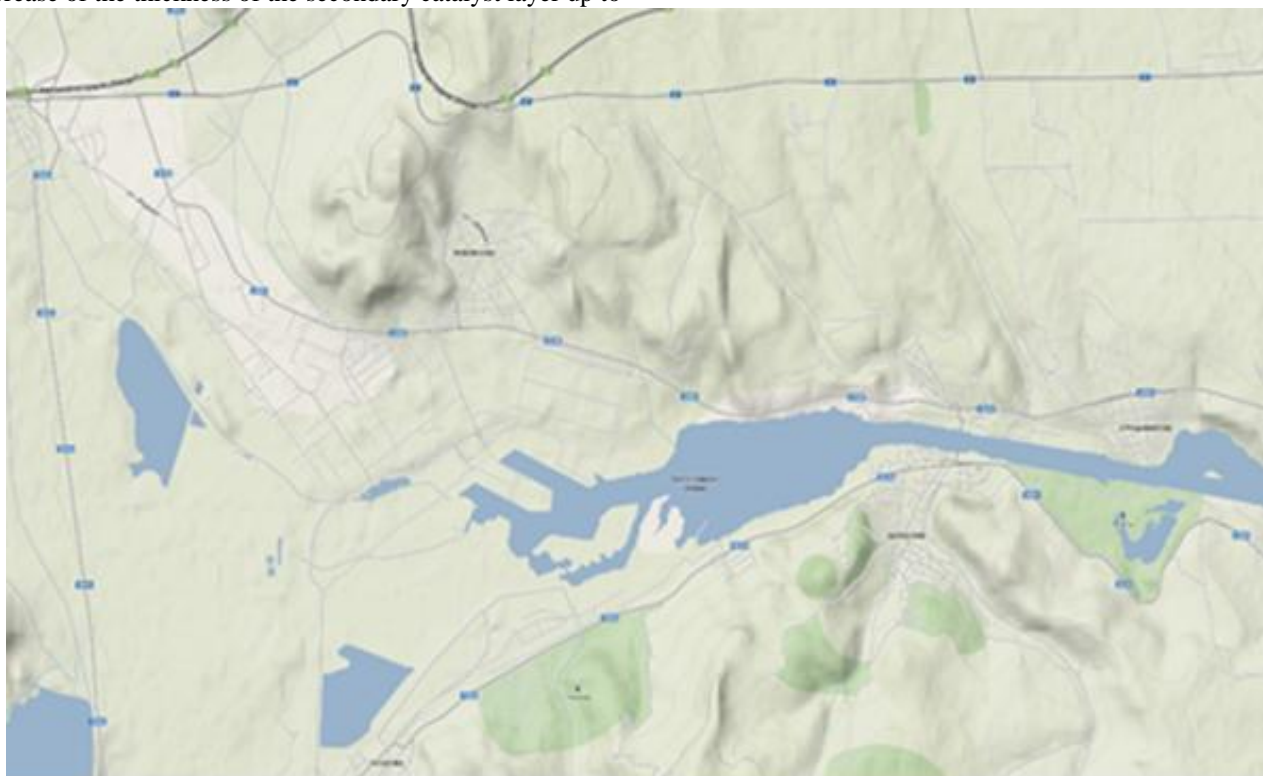


Fig. 1 Topographic map of Devnya region

#### B. Climate and meteorological conditions of the region

The climate of the source region is typical for its long and dry summer with high coefficient of windless periods and small rainfall quantity. Winter period is short and relatively soft with greater percentage and quantity of rainfall than summer period. There are breeze currents with east – west

direction circulating along Varna Lake and Beloslav Lake. At day time breeze currents come from east while at night they come from west. Moderate winds are typical for the region.

The terrain advantages the formation of inversion fog and considerable pollution of the ambient air. Statistical data indicates that about 60 % of calendar days are foggy.

Fig. 2 presents a wind rose built for the source region for 2013.

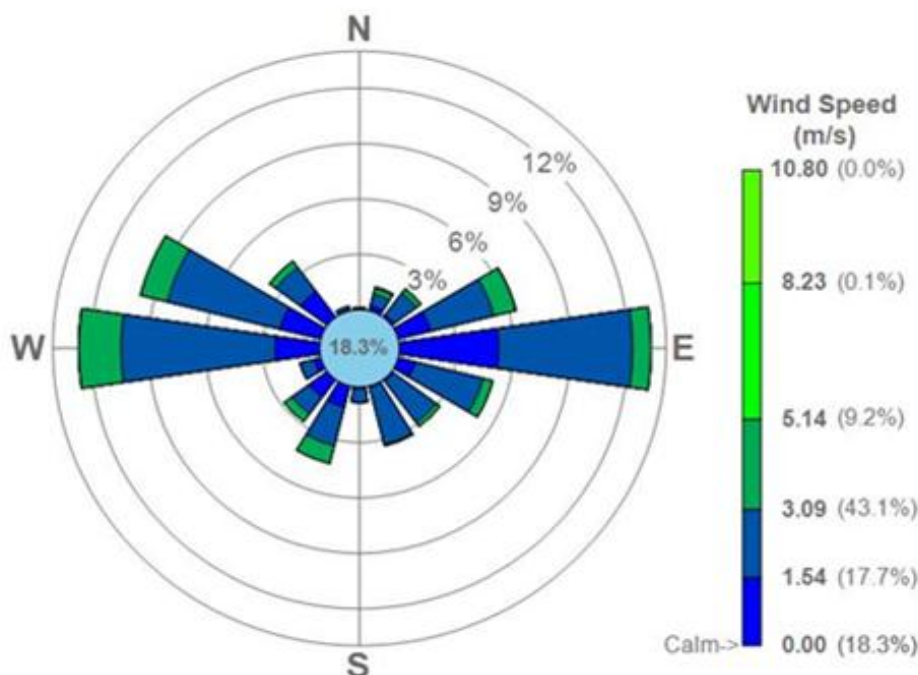


Fig. 2 Wind rose for 2013

As indicated on Fig. 2 most winds come from east and west with speed varying from 1.54 to 3.09 m/s. Winds with speed over 5.14 m/s are very unusual and mostly come from north-northeast. The percentage of windless periods is very low (18.3 %).

**C. Results**

Within the present research a single point source of N<sub>2</sub>O emissions is studied – P1 stack at the nitric acid plant. The height of the stack is 130 m and the diameter is 1.44 m. Due to the fact that N<sub>2</sub>O is inert in the troposphere and emissions are expected to expand on long distance from the source the study area is determined with the following proportions – 20000 m on west – east direction and 10000 m on south – north direction. The density of the recipients with a certain N<sub>2</sub>O concentration is 41 recipients on x (east) and 21 recipients on y (north) with 500 m step or the total amount of the recipients

is 861.

A simulation of N<sub>2</sub>O emission dispersion is done using BREEZE AERMOD software. Annual average N<sub>2</sub>O concentrations in the ground atmospheric layer are calculated as well as maximum average N<sub>2</sub>O concentrations over specific periods of time (1 hour, 8 hours and 24 hours) for both simulated scenarios.

Table 1 indicates calculation results for peak values of annual average N<sub>2</sub>O concentration, recipients’ coordinates and elevation and date/time when peak values are obtained. Fig. 3 and 4 present contours of annual average N<sub>2</sub>O concentration (µg/m<sup>3</sup>) in the ground atmospheric layer for both simulations – at 60 mm thickness of the secondary catalyst layer and at 90 mm thickness of the secondary catalyst layer.

Table 1 Peak values of annual average N<sub>2</sub>O concentration (µg/m<sup>3</sup>)

High	Avg. conc.	UTM		Elev. (m)	Hill Ht. (m)	Flag Ht. (m)	Rec. type	Grid ID
		East (m)	North (m)					
<b>Simulation at 60 mm thickness of the secondary catalyst layer</b>								
1ST	2.53	553791.95	4784610.56	155.40	209.00	2.00	GC	GC50C000

Simulation at 90 mm thickness of the secondary catalyst layer								
1ST	0.79	553791.95	4784610.56	155.40	209.00	2.00	GC	GC50C000

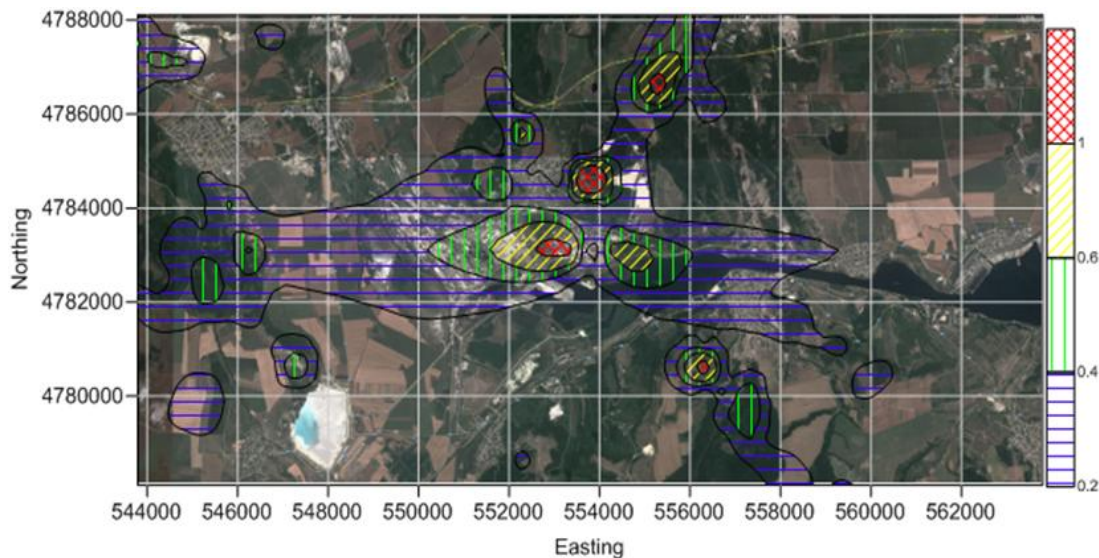


Fig. 3 Annual average N<sub>2</sub>O concentration (µg/m<sup>3</sup>) at 60 mm thickness of the secondary catalyst layer

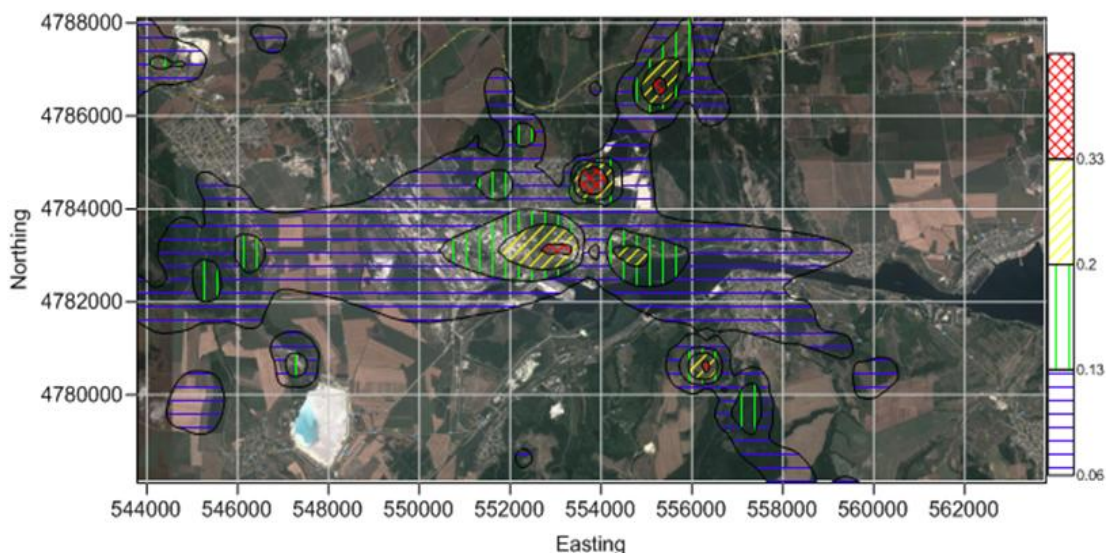


Fig. 4 Annual average N<sub>2</sub>O concentration (µg/m<sup>3</sup>) at 90 mm thickness of the secondary catalyst layer

Table 2 indicates maximum N<sub>2</sub>O concentrations (µg/m<sup>3</sup>) over specific periods of time (1 hour, 8 hours and 24 hours) for both simulated scenarios along with recipients' coordinates and elevation and date/time when peak values are obtained.

Table 2 Maximum N<sub>2</sub>O concentration (µg/m<sup>3</sup>) over specific periods of time (1 hour, 8 hours and 24 hours)

Avg. Per.	High	Type	Value	Date	UTM		Elev. (m)	Hill Ht. (m)	Flag Ht. (m)	Rec. Type	Grid ID
				YYMMDDHH	East (m)	North (m)					
<b>Simulation at 60 mm thickness of the secondary catalyst layer</b>											
1-HR	1ST	Avg. Conc.	1244.23	13/07/02/06	553791.95	4784610.56	155.40	209.00	2.00	GC	GC50C000
8-HR	1ST	Avg. Conc.	307.08	13/07/02/08	553791.95	4782110.56	155.40	209.00	2.00	GC	GC50C000

24-HR	1ST	Avg. Conc.	102.40	13/07/02/24	553291.95	4783610.56	155.40	209.00	2.00	GC	GC50C000
<b>Simulation at 90 mm thickness of the secondary catalyst layer</b>											
1-HR	1ST	Avg. Conc.	388.82	13/07/02/06	553791.95	4784610.56	155.40	209.00	2.00	GC	GC50C000
8-HR	1ST	Avg. Conc.	95.96	13/07/02/08	553791.95	4784610.56	155.40	209.00	2.00	GC	GC50C000
24-HR	1ST	Avg. Conc.	32.00	13/07/02/24	553791.95	4784610.56	155.40	209.00	2.00	GC	GC50C000

As indicated on Table 1, the planned future increase of the secondary catalyst layer thickness from 60 mm to 90 mm leads to prognosis reduction of the annual average N<sub>2</sub>O concentration in the ground atmospheric layer over 3 times – from 2.53 to 0.79 µg/m<sup>3</sup>. Similar reduction trend is observed regarding maximum N<sub>2</sub>O concentrations over specific periods of time (1 hour, 8 hours and 24 hours) as prognosis reduction of nearly 69 % is calculated for simulation at 90 mm thickness

of the secondary catalyst layer. Due to equivalent meteorological conditions at both simulated scenarios the maximum hourly N<sub>2</sub>O concentration is logically registered at the same recipient point on 2 July at 6.00 a.m. with south wind blowing with speed 2.82 m/s.

Fig. 5 – 10 present the contours of N<sub>2</sub>O concentration in the ground atmospheric layer over specific periods of time (1 hour, 8 hours and 24 hours) for both simulated scenarios.

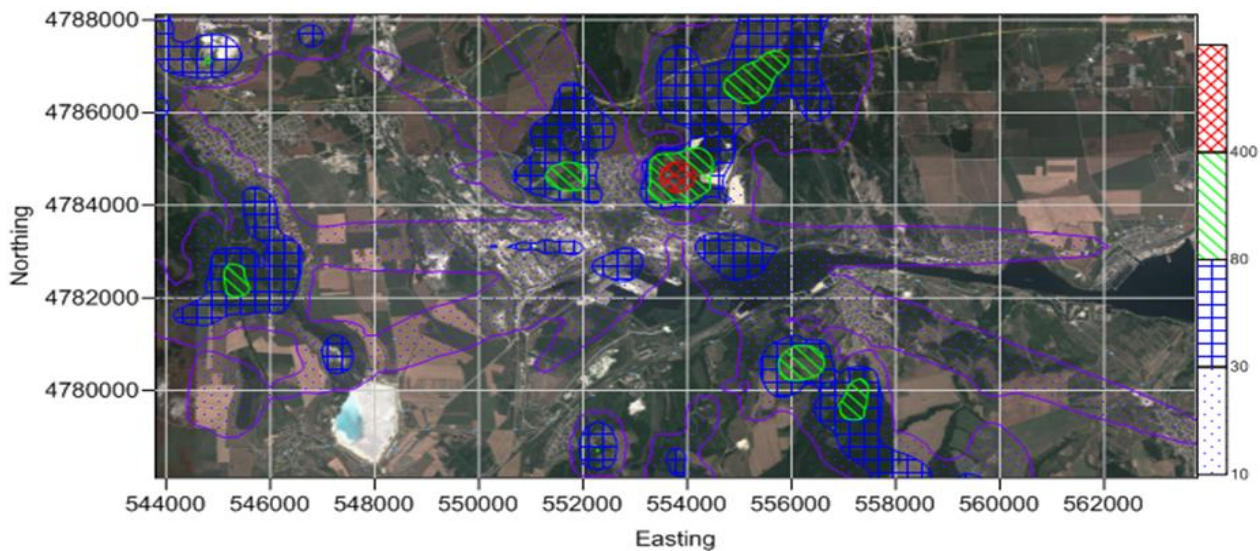


Fig. 5 Hourly average N<sub>2</sub>O concentration (µg/m<sup>3</sup>) at 60 mm thickness of the secondary catalyst layer

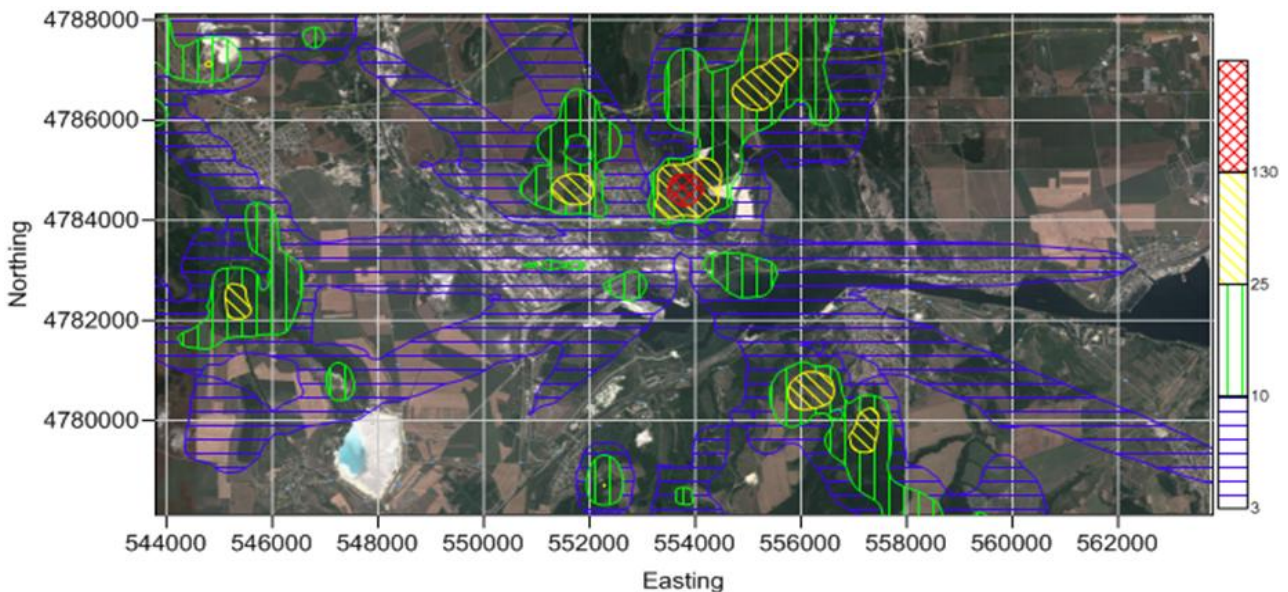


Fig. 6 Hourly average N<sub>2</sub>O concentration (µg/m<sup>3</sup>) at 90 mm thickness of the secondary catalyst layer

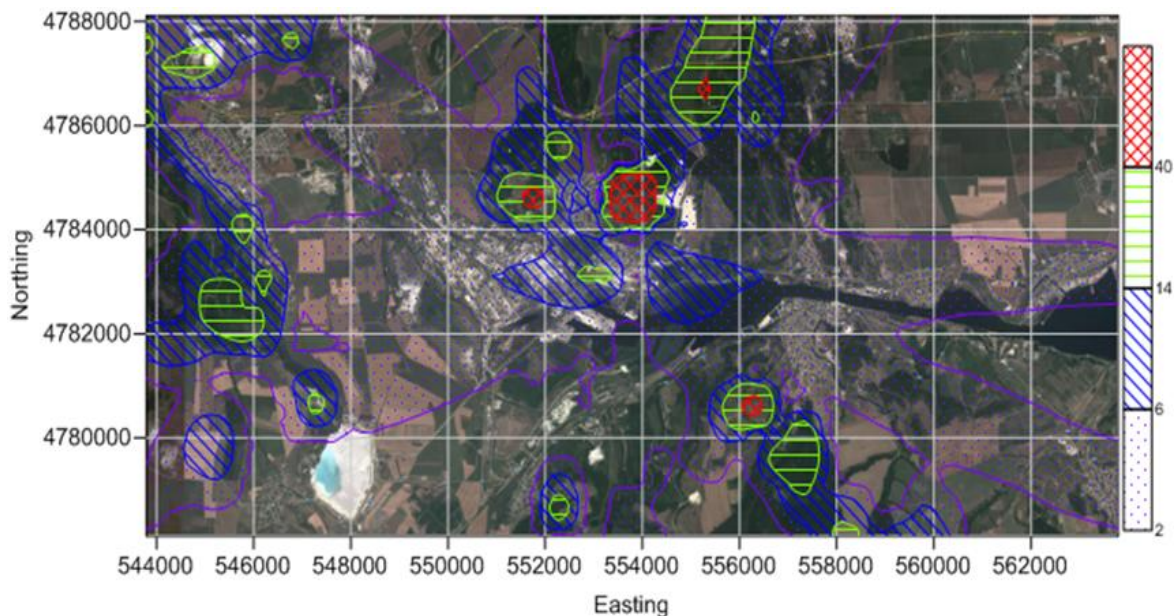


Fig. 7 Eight-hour average  $N_2O$  concentration ( $\mu g/m^3$ ) at 60 mm thickness of the secondary catalyst layer

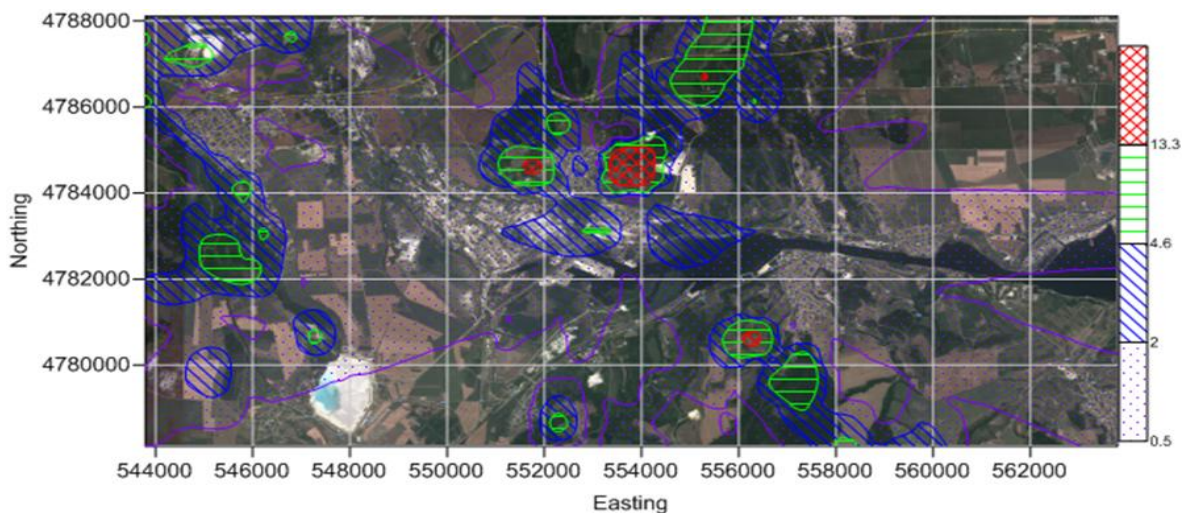


Fig. 8 Eight-hour average  $N_2O$  concentration ( $\mu g/m^3$ ) at 90 mm thickness of the secondary catalyst layer

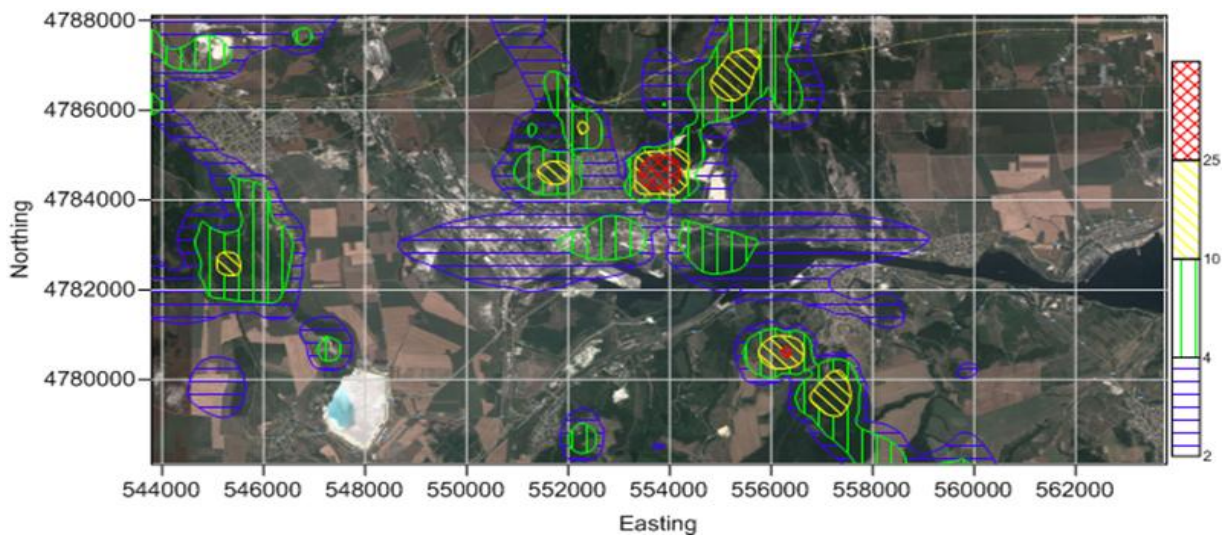
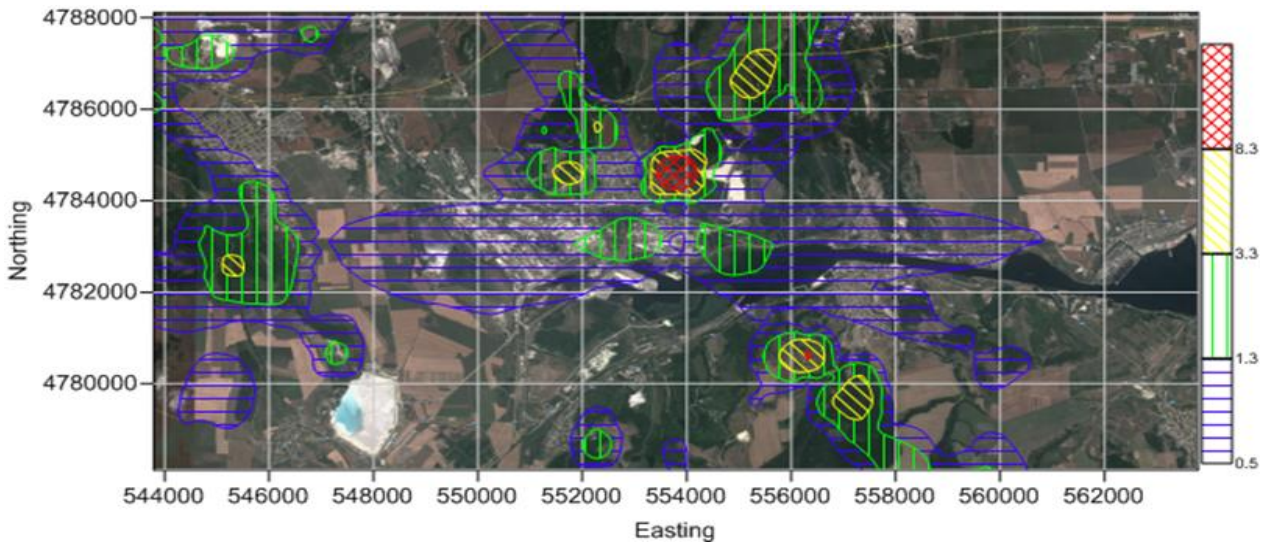


Fig. 9 Daily average  $N_2O$  concentration ( $\mu g/m^3$ ) at 60 mm thickness of the secondary catalyst layer



**Fig. 10 Daily average N<sub>2</sub>O concentration (µg/m<sup>3</sup>) at 90 mm thickness of the secondary catalyst layer**

As indicated on Fig. 5 – 10, the planned future increase of the secondary catalyst layer thickness up to 90 mm leads to reducing the hourly average, eight-hour average and daily average N<sub>2</sub>O concentrations in the ground atmospheric layer over 3.2 times due to prognosis reduction of N<sub>2</sub>O emissions from the nitric acid plant [26]. Within the present research validated meteorological data for 2013 is used for simulating both scenarios in order to obtain comparable results, therefore the assessment of the planned future increase of the secondary catalyst layer thickness upon the atmospheric N<sub>2</sub>O concentration is considered to not depend on the meteorological conditions.

As indicated on Fig. 3 and 4, the contours of the annual average N<sub>2</sub>O concentration outline equivalent areas for both simulated scenarios, but the simulation at 90 mm thickness of the secondary catalyst layer presents 3 times lower N<sub>2</sub>O concentration within the exposed areas (up to 0.33 µg/m<sup>3</sup>).

Calculations of the hourly average N<sub>2</sub>O concentration in the ground atmospheric layer also indicate equivalent contours of the exposed areas, but the simulation at 90 mm thickness of the secondary catalyst layer presents prognosis N<sub>2</sub>O concentration of 130 µg/m<sup>3</sup>, which is 3 times lower than the hourly average N<sub>2</sub>O concentration, simulated at 60 mm thickness of the secondary catalyst layer.

Analogical results are observed regarding N<sub>2</sub>O concentrations over the other specific periods of time. The simulation at future increase of the secondary catalyst layer thickness indicates prognosis reduction of N<sub>2</sub>O emissions over 3 times, which results in calculating maximum values of 13.3 µg/m<sup>3</sup> for the 8-hour average N<sub>2</sub>O concentration and 8.3 µg/m<sup>3</sup> for the daily average N<sub>2</sub>O concentration.

As indicated on Fig. 5 – 10 and Table 2, all peak values are registered at the same recipient point on the grid due to the specific local topography of the source region as the influence of the meteorological conditions is eliminated by using equivalent meteorological data for both simulated scenarios. All maximum N<sub>2</sub>O concentrations over specific periods of time (1 hour, 8 hours and 24 hours) for both scenarios are registered at the same point, situated near a hilly area, 760 m away on the east from the nearest settlement (Povelyanovo). Research results clearly indicate that typical

terrain like low hills and low valleys affect considerably the dispersion of atmospheric pollutants even at lowland areas.

#### IV. CONCLUSION

Analysis of the research results indicates a significant reduction of atmospheric N<sub>2</sub>O concentration for simulation at 90 mm thickness of the secondary catalyst layer – 69 % reduction of prognosis annual average, hourly average, 8-hour average and daily average N<sub>2</sub>O concentrations is calculated towards their relevant values at 60 mm thickness of the secondary catalyst layer, both scenarios being simulated under the same meteorological conditions. Besides the reduction of atmospheric N<sub>2</sub>O concentration, a considerable reduction is registered regarding the atmospheric areas of N<sub>2</sub>O emission dispersion. Both simulated scenarios outline equivalent atmospheric areas of dispersion but the simulation at 90 mm thickness of the secondary catalyst layer indicates that the maximum N<sub>2</sub>O concentrations over every specific period of time are 3 times lower than their respective values at 60 mm thickness of the secondary catalyst layer. Prognosis models indicate that all maximum N<sub>2</sub>O concentrations are registered at the same point, situated close to the nearest settlement therefore the prognosis reduction of atmospheric N<sub>2</sub>O concentration affects positively the ambient air quality in the region. Research results illustrate the direct connection between the thickness of the secondary catalyst layer and the reduction potential of the decomposing catalyst and may be used for prognosis assessment of atmospheric N<sub>2</sub>O concentration in the source region. The prognosis modeling of N<sub>2</sub>O emission reduction by increasing the thickness of the secondary catalyst layer is an assessment tool for calculating the contribution of N<sub>2</sub>O emissions from nitric acid production to the overall greenhouse gas emissions of all Member States to the United Nations Framework Convention on Climate Change over the following commitment periods [40].

Efficient reduction of N<sub>2</sub>O emissions from nitric acid production leads to reducing the concentration of N<sub>2</sub>O in the ground atmospheric layer and is proved to be an effective tool to combat the adverse changes of the climate system and move to a competitive low carbon economy [41]-[42].

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