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RESEARCH OF THE AMBIENT AIR POLLUTION IN THE INDUSTRIAL REGION DEVNYA, BULGARIA

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ABSTRACT: *The research has been done in the industrial region Devnya, Bulgaria for the term 2010 – 2014. The period has been chosen in accordance with the coming into force of the IPPC permits of the different operators of production installations. It has been used data from a monitoring station - automated measuring station "Izvorite" as the monthly and average annual concentrations of specific air pollutants were examined such as: SO_2, NO_2, O_3, CO, C_6H_6, NH_3 and PM_{10}. The results show that the annual average concentrations of air pollutants are below the emission thresholds for the protection of human health. For some pollutants (PM_{10}, SO_2, NO_2, CO and benzene) a seasonal dynamic was observed, associated mostly with emissions from combustion processes - heating in the residential and commercial sector. The results note also, that the measures, pursuant to the Integrated Pollution Prevention and Control permits (IPPC permits) of the industrial and combustion plants in the industrial region Devnya are effective and give their contribution for improvement of the ambient air quality.*

KEY WORDS: *ambient air quality, industrial region Devnya, monitoring station Izvorite, PM_{10}, pollution.*

INTRODUCTION

The research of the ambient air quality is current. This is due to the requirements of [7] for clean air in Europe. It is necessary the implementation of effective strategies to reduce the level of air pollution not only internationally but also locally.

In Bulgaria except the Clean Ambient Air Act, which requires an improvement of the ambient air quality [6], different kinds of environmental documents are implemented to demand of the operators of industrial and combustion plants to use the best available techniques in carrying out industrial activities and effective treatment facilities of harmful substances in exhaust gases. The ultimate goal is decreasing the concentrations of pollutants in

ambient air and reach the standards set to protect human health and the environment as a whole. Regulation No 12 of 15 July 2010 is a typical "tool" for achievement of the reduction of some pollutants in the ambient air by regulating their emission thresholds [15]. The assessment of the ambient air quality and its management [14] also requires the informing the population [12].

A number of authors study the air pollution in environmentally threatened areas in Bulgaria by establishing high concentrations of many pollutants (SO_2, NO_x, O_3, CO, benzene) [18] and especially of particulate matter PM_{10} in industrial and urban areas [2, 5, 11, 19].

The purpose of this study is the research of ambient air pollution in the industrial region Devnya, the concentrations of pollutants to be compared with the permitted limit values and the effect of coming into force of IPPC permits of operators of industrial installations for improving of the ambient air quality to be assessed.

MATERIAL AND METHODS

The study has been done in the industrial region Devnya. Devnya Municipality is located in northeastern Bulgaria, district of Varna, an area of 121.052 km^2. Devnya is approximately 30 km far away from the district city Varna, in a small valley, surrounded on all sides by low limestone hills as this favors the development of various industries in the area. In the industrial region Devnya there are plants with high rate of production, which are essential for the national economy. Solvay Sodi JSC is a plant for production of light and dense synthetic soda, and soda bicarbonate as a by-product for food processing and industrial consumers. Agropolychim JSC is one of the main leaders in production of fertilizers in the Balkans. There are two production lines at the plant – one for the production of nitrogen fertilizers which includes production of ammonia, nitric acid, ammonium nitrate and urea ammonium nitrate solution and another one for the production of phosphate fertilizers which includes phosphoric acid, sodium tripolyphosphate, triple superphosphate and compound fertilizers monoammonium phosphate and diammonium phosphate. Deven JSC is a co-generating power station for thermal and electrical power. The power station is based on separate combustion of imported coal, petroleum coke, natural gas and mazut. Produced thermal and electrical power is transferred to the other industrial plants in the region as part of the electrical energy is transferred to the national energy system. Part from the industrial region is also Devnya Cement JSC, which is plant for production of clinker and cement. The main raw materials for cement production are limestone, sand and marl.

In Devnya there is a monitoring station of the ambient air, which is part of the national system for monitoring and control – the automatic monitoring station Izvorite with nonstop operation. The station has been operating since 1990 and gives data about the concentrations of main pollutants, which are

essential for the ambient air quality in real time. The automatic monitoring station Izvorite which is placed in region "River Devnya" is classified as an industrial / urban background monitoring station with a range 10-100 m/100 m-2 km. The station is handled by Regional laboratory Varna, which is a department of the Executive Environmental Agency at the Ministry of Environment and Water. At the monitoring station concentrations of the following pollutants are measured: sulfur dioxide SO_2, nitrogen dioxide NO_2, carbon monoxide CO, ozone O_3, particulate matter 10 μm in diameter PM_{10}, ammonia NH_3 and benzene C_6H_6. The assessment of the ambient air pollution is based on a data from the monitoring station for the five-year period: from 2010 to 2014.

RESULTS AND DISCUSSION

The results from the monitoring about sulfur dioxide show that for the entire period of research the average annual concentrations of SO_2 are significantly below the limit value – average daily limit value for protection of the human health (125 μg/m^3) [15]. At the beginning of the research (2010) the average annual concentration of SO_2 is highest – 10.25 μg/m^3. There is a descending trend in sulfur dioxide pollution and in the end of the period (2014) the average annual concentration reaches 5.69 μg/m^3.

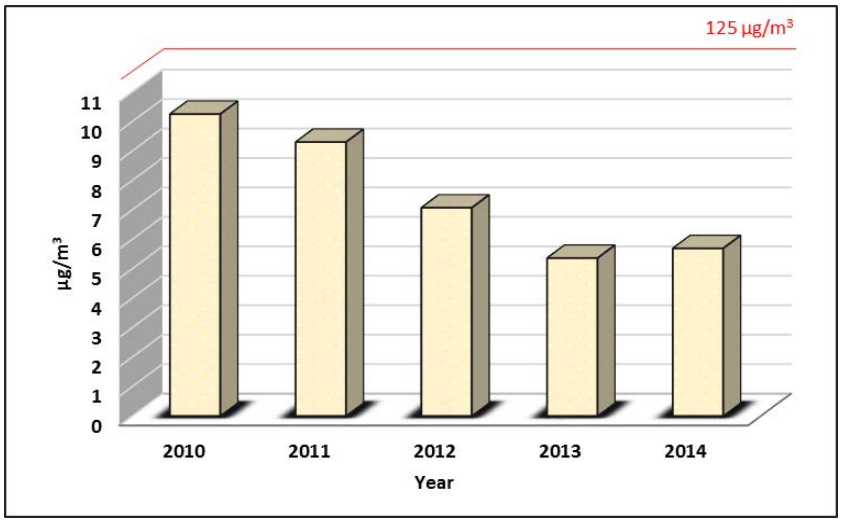
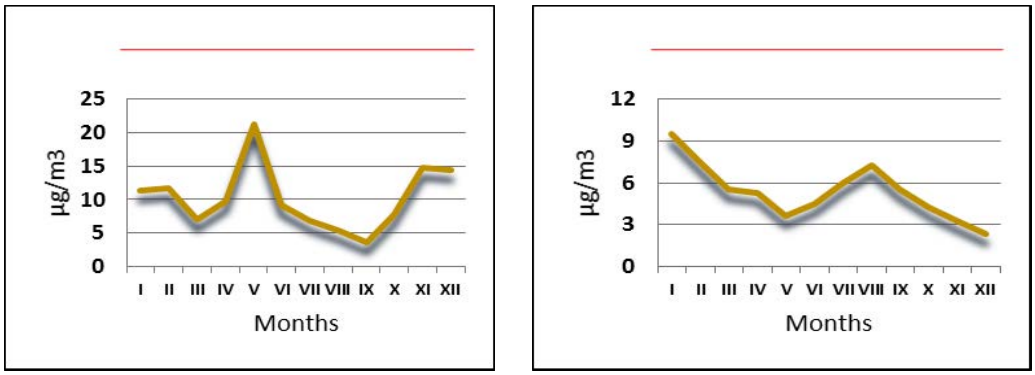


Fig. 1. Average annual SO_2 concentrations in the ambient air

The data for the monthly concentrations of sulfur dioxide for 2010 and 2013 is marked in fig.2. It is chosen the period when the highest and lowest annual average concentrations of the pollutant are registered (in fig.1).



2010 **2013**

Fig. 2. Average monthly SO_2 concentrations in the ambient air

It is obvious from the results that the average monthly concentrations of sulfur dioxide are the highest through the winter months. The highest values of sulfur dioxide are during the months November and December of 2010 – respectively 14.82 µg/m^3 and 14.42 µg/m^3 (the exception is observed in May – 21.14 µg/m^3). In 2013 again the highest concentrations of sulfur dioxide are reported through the winter months – January and February, respectively 9.46 µg/m^3 and 7.38 µg/m^3. We can summarize the higher concentrations of sulfur dioxide during the winter periods are not random, but they are a result of combustion processes and mostly of the residential heating in the use of solid and liquid fuels with high sulfur content.

The average annual concentrations of nitrogen dioxide for the period of research are presented in fig.3. It is very clear that concentrations of NO_2 are under the average annual limit value for the protection of the human health - 40µg/m^3 [15]. The highest average annual concentration of nitrogen dioxide is established at the beginning of the period of the research in 2010 – 20.0 µg/m^3. After 2011 the average annual concentrations gradually are declining from 18.2 µg/m^3 to 12.15 µg/m^3 (in 2014). It is typing a model of a descending trend.

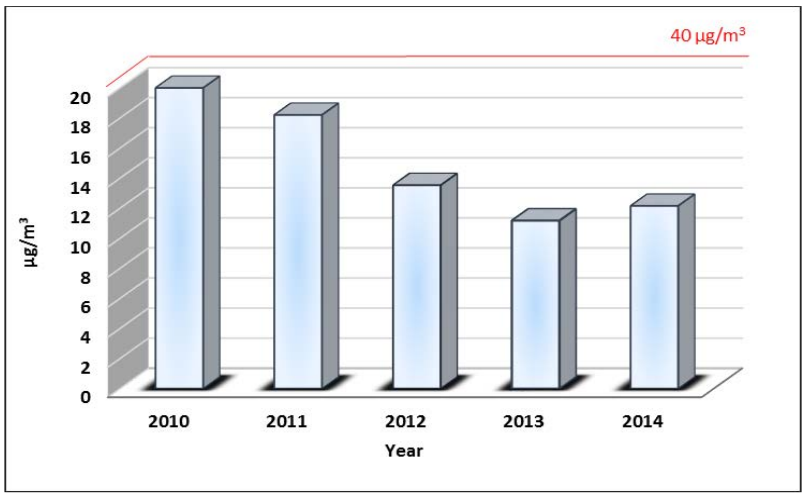
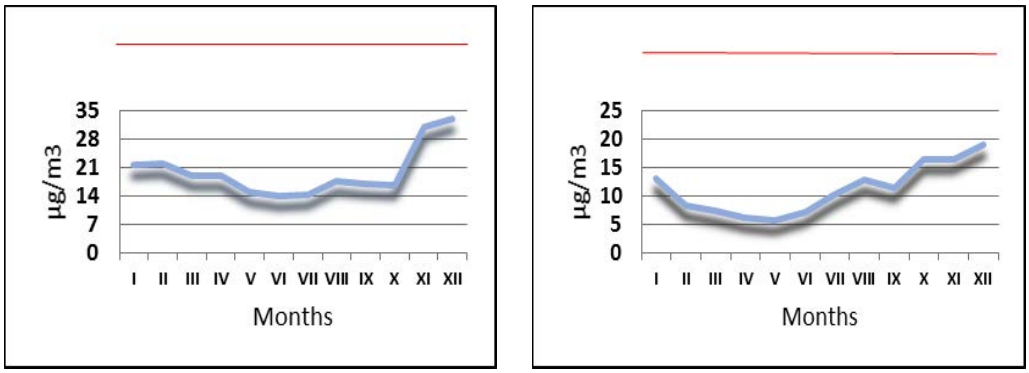


Fig. 3. Average annual NO_2 concentrations in the ambient air

In fig.4 the average monthly concentrations of nitrogen dioxide are represented in 2010, (at the beginning of the research) and in 2013, when there is a drop of the pollution by NO_2. The results show that the most contamination is during the winter months, but the limit value for the protection of human health for NO_2 (40 μg/m^3), is not exceeded. Through the two years of research, the highest monthly concentrations of nitrogen dioxide are reported during the period November – December. In 2010 the average monthly concentrations reach 31.05 – 32.96 μg/m^3, and in 2013 the highest monthly concentrations in December is 19.08 μg/m^3. The annual monthly concentrations express seasonal relation in the ambient air pollution by nitrogen dioxide. The impact on the higher values in concentrations during the cold months of the year is mainly caused by the combustion processes. In the industrial region Devnya it is places the co-generating power plant "Deven". Except this the heating in the residential and commercial sector contributes to pollution not only by sulfur dioxide but also by nitrogen dioxide.



2010 **2013**

Fig. 4. Average monthly NO_2 concentrations in the ambient air

The results about carbon monoxide show (fig.5) that the average annual concentrations of CO are below the limit value for protection of the human health – maximum eight hours average value for twenty-four hours (10.0 mg/m^3) [15]. There is no trend in the pollution by carbon monoxide. The average annual concentrations of CO vary from 0.30 mg/m^3 to 0.69 mg/m^3, as the highest value is established in 2012 – 0.69 mg/m^3. After 2012 there is a gradually descending of the concentrations and the lowest levels of CO are in the end of the research in 2014, when the average annual concentrations reach 0.30 mg/m^3.

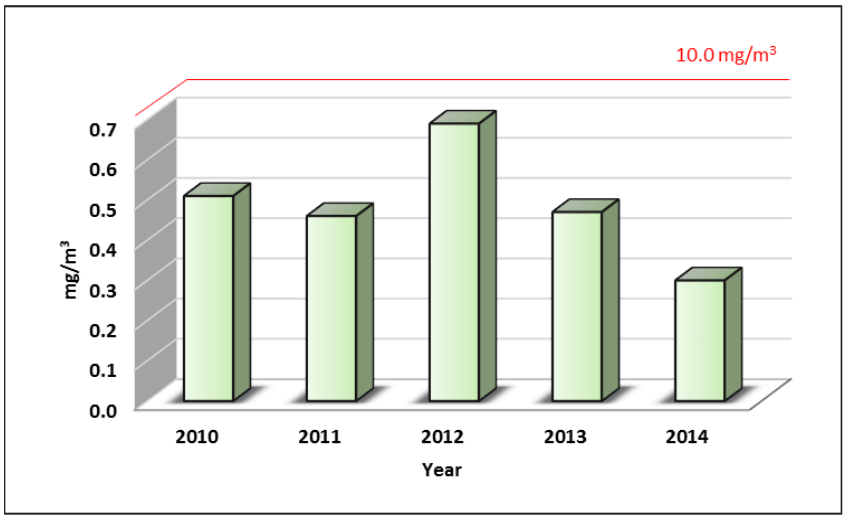
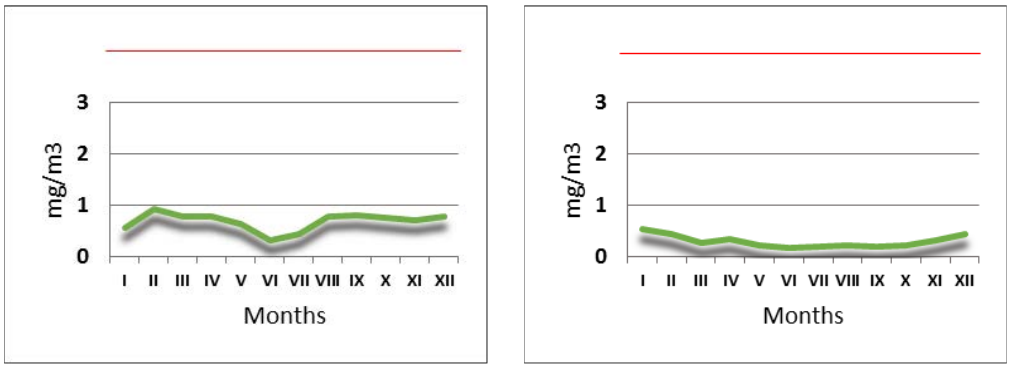


Fig. 5. Average annual CO . oncentrations in the ambient air

Data about the monthly concentrations of CO for 2012 and 2014 is indicated in fig.6, because during these periods there are the highest and the lowest pollution with carbon monoxide. From the results it is observed that the average monthly concentrations of CO are significantly below the limit values in both years. In 2012 the highest levels of the pollutant are established during the winter months – the highest average monthly concentration of CO is in February, which is 0.93 mg/m^3. In 2014 it is observed again high average monthly values of carbon monoxide during the heating season – the highest concentration of CO is in January – 0.54 mg/m^3. The seasonal relation of the pollution with carbon monoxide is caused by the residential heating during the winter period of the year mainly with burning of coals in home stoves.



2012 **2014**

Fig. 6. Average monthly CO concentrations in the ambient air

The ozone (O_3) is a powerful oxidant, which is a powerful pollutant. It does not emit directly to atmosphere. It is a result of the interaction of nitrogen oxides and volatile organic compounds under the influence of high temperatures and solar radiation. There are no significant variations in air pollution with ozone for the whole period of research. It is obvious from the data in fig.7, the average

annual concentrations of O_3 are below the limit value for protection of the human health – the maximum eight-hour value within the day (120 µg/m^3) [15]. The average annual concentrations of ozone range from 32.77 µg/m^3 (2012) to 44.42 µg/m^3 (2010).

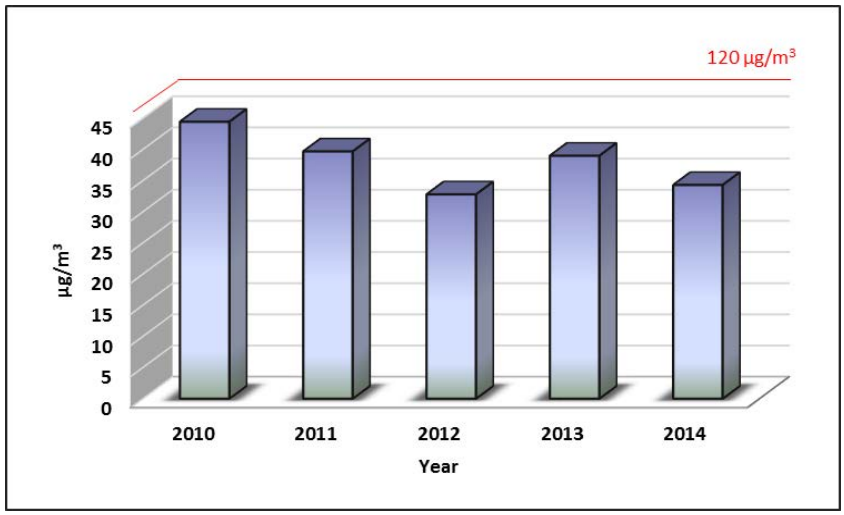


Fig. 7. Average annual O_3 concentrations in the ambient air

Data about average monthly concentrations of ozone in 2010 and 2012 are presented in fig.8 when there is highest and lowest level of pollution. It is clearly that the average monthly concentrations of ozone are higher during the spring – summer months of the year. In 2012 the highest average monthly concentration is founded in July – 44.49 µg/m^3. The seasonal relation and the higher levels of O_3 in the hot period of the year are due to the higher intensity of solar radiation during the summer months. As we mentioned earlier, the ozone is a secondary pollutant which is formed at high temperatures and under the influence of solar radiation.

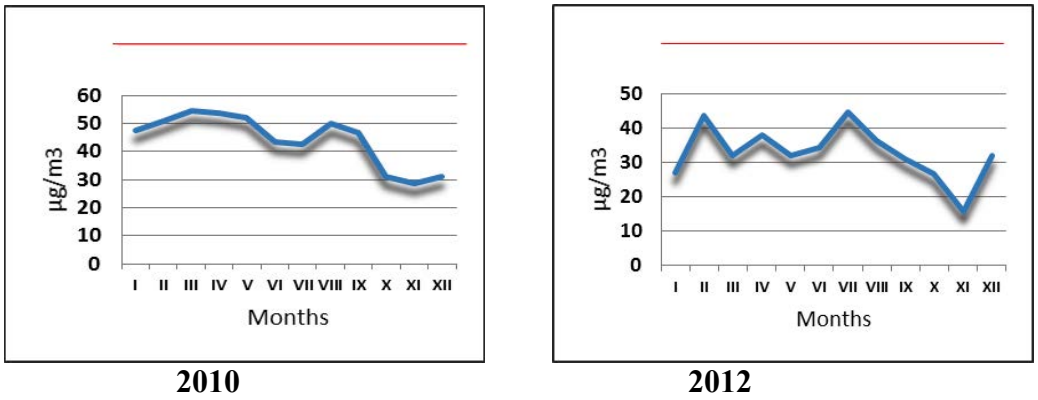


Fig. 8. Average monthly O_3 concentrations in the ambient air

Particulate matter can be transported in the atmosphere for thousands of kilometers, and the pollutants emitted in one country can affect PM_{10} concentrations in countries which are close or even far away from the source [10].

The average annual concentrations of PM_{10} for the period 2010 – 2014 are presented in fig.9. At the beginning of the study (2010 – 2011) the average annual concentrations are higher, respectively 23.31 μg/m^3 and 20.82 μg/m^3 and the average annual limit value for protection of the human health hasn't been exceeded (40 μg/m^3) [15]. After 2011 there is a gradually increase of the average annual concentration in 2014 which reaches 30.97 μg/m^3. There is an upward trend at the end of the research but the limit value is not exceeded.

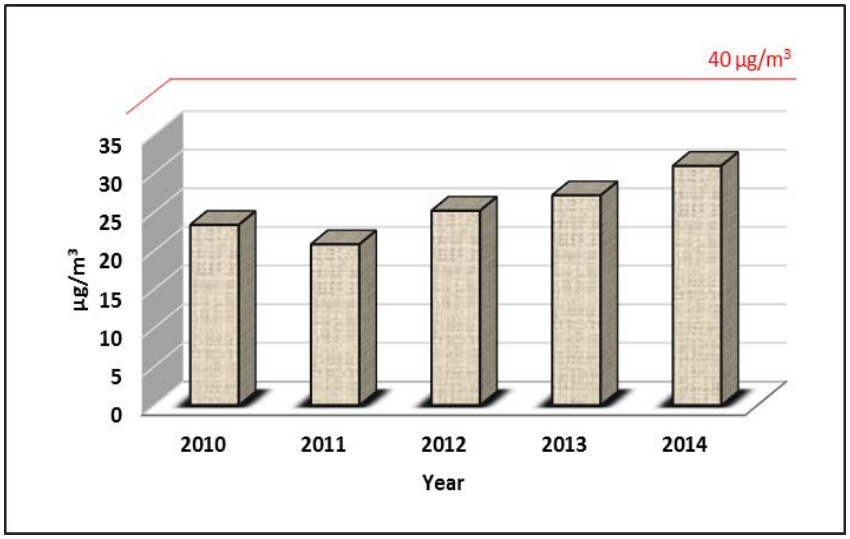


Fig. 9. Average annual PM_{10} concentrations in the ambient air

It is presented information about the average monthly concentrations of particulate matter about 2011 and 2014 in fig.10. It is chosen the period when there are the lowest and the highest levels of pollution with particulate matter.

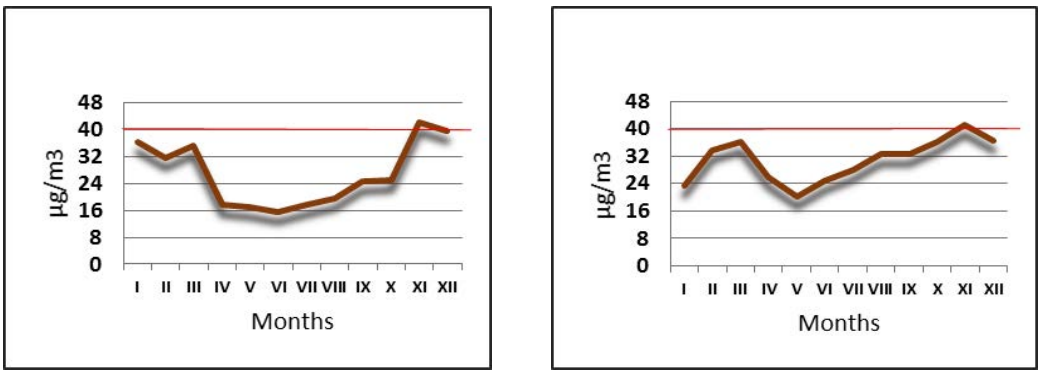


Fig. 10. Average monthly PM_{10} concentrations in the ambient air

The results in fig.10 show that the average monthly concentrations of particulate matter are significantly higher through the winter heating season. In 2011 the highest average monthly concentrations of PM_{10} at the station „Izvorite“ are registered in the period January – March (35.02 – 36.11 μg/m^3) and in November – December (39.38 – 41.96 μg/m^3). The average monthly concentrations of PM_{10} in November exceed the limit value for protection of the human health 1.05 times. During the summer months, the concentrations of particulate matter vary from 15.62 μg/m^3 (in June) to 19.67 μg/m^3 (in August).

In 2014 there are again higher concentrations during the winter months. The highest average monthly concentrations of PM_{10} are in November and December – till 36.65μg/m^3 and till 41.18 μg/m^3, as the average annual limit value for protection of the human health has been exceeded 1.03 times. The high concentrations of PM_{10} during the cold months of the year are mainly related to the residential heating with burning of coals and firewood. Another sources of particulate matter in the industrial region are the co-generating power plant “Deven” JSC, where there is mainly burning of coals, which creates a large amount of ash [1], “Devnya Cement” JSC – huge quantities of particulate cement dust is emitted, “Solvay Sodi” JSC for production of synthetic soda ash and “Agropolychim” JSC for production of granulated nitrogen and phosphate fertilizers.

Fig.11 displays the average annual concentrations of benzene in the ambient air. The benzene is with proven carcinogenic effect. The International Agency for research cancer (IARC) classifies the benzene in first group (proven carcinogens for people), as it causes benzene leukemia – cancer of the blood.

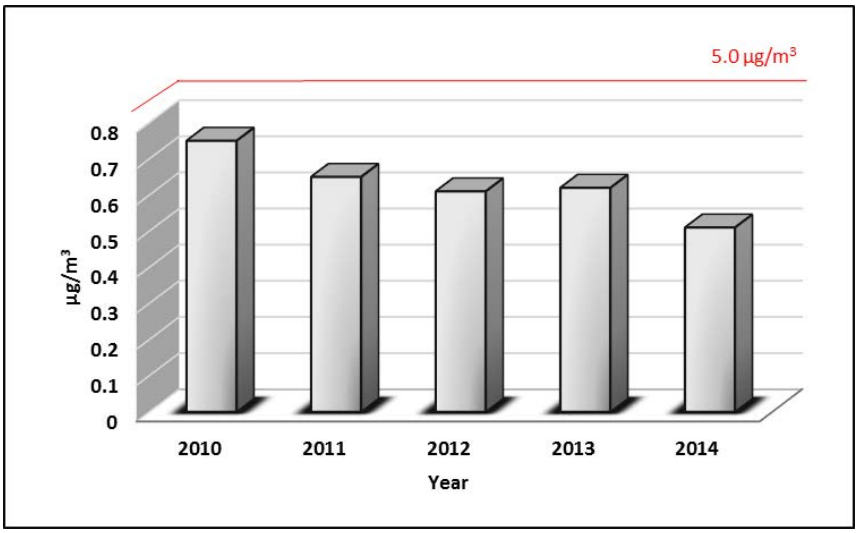
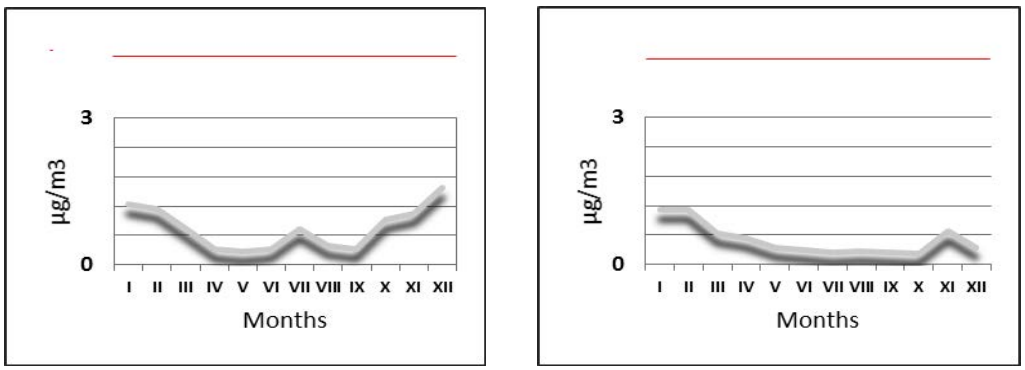


Fig. 11. Average annual concentrations of benzene in the ambient air

As it is visible by the data, there is no significant variations in the average annual concentrations of benzene through the period of the research and the

limit value for protection of the human health has not been exceeded (5 μg/m^3) [15]. The highest average annual concentration is in 2010 – 0.75μg/m^3. After 2010 there is a slight downward trend as the average annual concentrations of benzene reach 0.65 μg/m^3 in 2013 and 0.51 μg/m^3 in 2014.

The average annual concentrations of benzene in 2010 and in 2014 are shown in fig.12. There are chosen periods in which the concentrations of benzene are respectively highest and lowest.



2010 **2014**

Fig. 12. Average monthly concentrations of benzene in the ambient air

During the two years of research, the highest concentrations are in the winter months respectively in the periods January – February and November – December. In 2010 the highest average monthly concentration is in December – 1.57 μg/m^3, and in 2014 – in January and February (1.12 μg/m^3). This seasonal relation with higher values during the cold months and lower levels of pollution during the hot months of the year is mainly due to the combustion processes through the heating season by residential heating with burnings of firewood.

The average annual concentrations of ammonia for the period 2010 – 2014 are presented in fig.13. It is obvious from the results that the average annual concentrations of ammonia for the period of research are significant below the average daily limit (100 μg/m^3) [16]. The highest average annual concentration of NH_3 is in 2010 – 1.85 μg/m^3. After 2010 there is a drop of the pollution with ammonia. The average annual concentrations of NH_3 in 2012 and in 2013 are 0.6 - 0.7 μg/m^3 and low level of pollution is maintained. The concentrations of ammonia gradually increase in 2014 – the average annual concentration of NH_3 is 1.0 μg/m^3, but again it is highly below the limit value. The drop of the average annual concentrations of ammonia during the period of research is mainly due to the low capacity of some productions at Agropolychim JSC (reducing the production of ammonium nitrate, urea ammonium nitrate solution), in which NH_3 is the main pollutant.

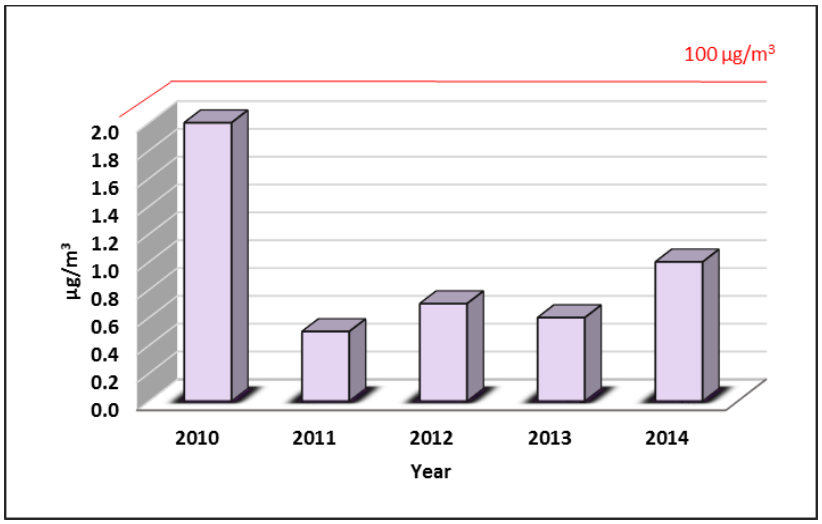


Fig. 13. Average annual NH_3 concentrations in the ambient air

The concentrations of the research pollutants in the ambient air in the period of study are below the limit values for protection of the human health in accordance with Regulation No 12 of 15 July 2010 relating to limit values for sulfur dioxide, nitrogen dioxide, particular matter, lead, benzene, carbon monoxide and ozone in ambient air. The low level of pollution is connected with the implemented IPPC permits of industrial and combustion plants in Devnya region for reducing of harmful emissions and improvement of the ambient air quality. The Directive which concerns the issues from the industry requires integrated pollution prevention and control upon all industrial activities. By this environmental act some special measures come into force, which aim prevention and there where it is not possible – to limitation of the emissions in the environment [8]. For many industrial and combustion plants which are with significant importance for the ambient air quality in some region IPPC permits are required in accordance with the Directive. By the Regulation on terms and methods for IPPC permits issue, the conditions and the order for coming into force of IPPC permits are regulated for construction and operation of new and operation of already existing plants and facilities for different kind of activities according to Annex № 4 of Environmental Protection Act [17, 9]. In some other studies we have analyzed the measures for reducing of the pollution and improvement the ambient air quality in the industrial region Devnya [3, 4, 13].

All plants in the industrial region Devnya have IPPC permits – IPPC permit № 68 of Agropolychim JSC since 14.01.2006; IPPC permit № 63 of Devnya Cement JSC since 14.01.2006; IPPC permit № 74 of Solvay Sodi JSC since 14.01.2006; IPPC permit № 93 of co-generating power plant Deven JSC since 06.05.2006. For implementation of the activities of the IPPC permits in the industrial plants there are introduced monitoring systems for nonstop control and operation of the emissions of harmful substances in the ambient air; at Agropolychim JSC there are implemented high efficiency facilities for tail gases

from dust and the plant for the production of ammonium nitrate is reconstructed; at the co-generating power plant Deven JSC there is a new circulating fluidized-bed steam generator with low level of issues; at Solvay Sodi JSC there is a new line for the production of soda ash dense, an issue free distillation column, a carbonizing column and a limestone blast furnace. As a result of the implemented measures for reduction of issues and introducing new technologies, the ambient air quality is improved.

CONCLUSIONS

As a result of the research it is established that the average annual concentrations of all pollutants in the ambient air in the industrial region Devnya are below the limit values for protection of the human health for the period 2010-2014. For the different pollutants there are different trends. For the sulfur dioxide and nitrogen dioxide it is observed a pronounced downward trend at the end of the period in 2014. The concentrations of PM_{10} increase gradually at the end of the research, but the limit value for protection of the human health has not been exceeded. About the secondary pollutant ozone there are no very marked variations in the period of the study and the average annual concentrations for the period of research does not exceed the threshold. The research indicates also that it exists a seasonal relation between the pollution of the ambient air and some pollutants as: PM_{10}, SO_2, NO_2, CO and benzene, as during the winter months there are significant higher average monthly concentrations as compared to the summer months of the year. The seasonal dynamic is associated mainly with emissions from combustion processes – the heating in the residential and administrative sector. The results from the research demonstrate that the measures implemented in the IPPC permits of industrial and combustion plants in the industrial region Devnya are effective and contribute to improvement of the ambient air quality.

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