

## STUDIES ON ENVIRONMENTAL POLLUTION IN LARGE INSTALLATION OF COMBUSTION IN REGARD WITH INTERNATIONAL STANDARDS

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**Abstract.** Industrial development and globalization forces the European Union to reconsider immediate needs for energy production and transport. The study refers mainly to the impact Craiova II Thermal Power Complex has on vector environment - air. Due to the technological process that pollutes the environment, we have monitored some of the physical-chemical parameters of the air, with implications for the ecosystems. The coal waste deposited in various places has a high content of pyrite. The most important environmental pollution problem is oxidation of the pyrite and the generation of acidity. The coal waste is deposited and constitutes an active source of acid ( $H_2SO_4$ ) that generates soil contamination at surface, as well as groundwater contamination, endangering ecosystems. Pyrite oxidation results in  $Fe^{2+}$ ,  $SO_4^{2-}$  and  $H^+$ . Coal mining drainages are characterized by low pH, a highly varied composition that prevail in high concentrations of sulphate, iron, manganese, aluminum and other toxic and radioactive ions, as well as solid particles in excess. This drainage is one of the oldest problems arising from coal mining.

**Keywords:** environmental protection, air quality, physico-chemical parameters, coal.

**Rezumat. Studiul privind poluarea mediului de către instalațiile mari de ardere în acord cu standardele internaționale.** Dezvoltarea industrială și fenomenul de globalizare obligă Uniunea Europeană la o reconsiderare imediată a necesităților de energie, producere și transport. Studiul se referă în principal la impactul pe care îl are Complexul Termoenergetic Craiova II asupra mediului-vector aer. Datorită procesului tehnologic care poluează mediu, am monitorizat o parte din parametrii fizico-chimici ai aerului, cu implicații asupra ecosistemelor. Deșeurile de cărbune depozitate în diverse locuri, au un conținut ridicat de pirită. Cea mai importantă problemă legată de poluarea mediului, o reprezintă oxidarea piritei și generarea de aciditate. Deșeurile sunt depozitate și constituie o sursă activă de acid ( $H_2SO_4$ ) care generează contaminarea solului la suprafață și a apei freatică, periclitanț ecosistemele. Din oxidarea piritei rezultă  $Fe^{2+}$ ,  $SO_4^{2-}$  și  $H^+$ . Drenajele din minele de cărbuni sunt caracterizate de pH scăzut, o compoziție foarte variată în care predomină concentrații mari de sulfați, fier, mangan, aluminiu și alți ioni toxici și radioactivi, ca și particule solide în exces. Aceste drenaje constituie una din cele mai vechi probleme ce apar ca urmare a exploatării cărbunelui.

**Cuvinte cheie:** protecția mediului, calitatea aerului, parametrii fizico-chimici, cărbuni.

### INTRODUCTION

In order to protect the atmosphere and improve air quality, measures are needed to control pollutant emissions. To assess the degree of air pollution, pollutant emissions are calculated and the quality of the ambient air is determined. Emissions are measured by appropriate assessment methods, specific to each pollutant, based on emission factors and activity indicators. The analysis of emissions at national level, sectorial distribution, spatial and temporal targets are key elements in setting environmental priorities, identifying the targets to be achieved and policies to be adopted, both locally and nationally. The indicators selected must meet the identification criteria and be relevant for the main issues related to the atmosphere.

The main objectives of environmental policy in Romania are designed to ensure a clean environment and aim to ensure a healthy life for the population, to lead to the elimination of poverty and environmental degradation, to regenerate the economy based on sustainable development principles and to harmonize national legislation on protection of the environment with that of the European Union.

Coal power plant in Romania and European countries are responsible for more than half of the negative effects on health. Directive 84/360 / EEC on the control of environmental pollution from industrial sources has been amended by Directive 2009/31 / EC on integrated pollution prevention and control and transposed into Law no. 205/2010 improved by Law no. 278/2013 on industrial emissions (\*\*\*, 2013). The objective of the above-mentioned legislation is to reduce pollution to the admissible limits. For the environmental factor air, there are considered measures to prevent, reduce, offset, as far as technically possible, measures generated by implementing legislation in force (RUSU & ROJANSCHI, 1980; BRÎNDUȘA & KOVACS, 2007; GAVRILESCU & POPESCU, 2012).

Different types of coal, coming from several basins, can have high sulfur content. In its structure, there are two forms of sulfur, namely: organic and inorganic sulfur. In coals from Russia and the United States, for example, inorganic sulfur is the major component, which is present between 0.5% and 11% in the form of iron disulfide (pyrite and marcasite). Other forms of inorganic sulfur are elemental sulfur and sulphides, which contain different metals. Organic sulphide is present in the form of thiols or mercaptans, sulphides or thioethers and thiophene compounds, these being integrated into the coal matrix. For the removal of organic sulfur from coal covalent bonds (C-S), which are resistant to the chemical treatment, must be broken (CISMAȘIU 2010a; b; TOMUȘ et al., 2015; ATHRESH et al., 2017).

## MATERIALS AND METHODOLOGY

The branch of Electrocentrale Craiova II is located in the N-E area of Craiova, about 1 km away from the lower railway passage between Bariera Vâlcii Street and Craiova-Filiași railway line. The main road access to the Craiova II SE is located on the eastern side, namely on Bariera Vâlcii Street. The thermal power plant SE Craiova II occupies an area of approx. 433,727 sqm. The site of slag and ash storage (landfill of non-hazardous waste - slag and ash) in the area of approx. 153 ha is located approx. 5.5 km of the premises (plant), S-E of the commune of Șimnicu de Sus, at approx. 1 km N-E from the village of Jieni (Fig. 1).

CE Oltenia SA - SE Craiova II, located on the southern platform of the city, produces electricity for the national energy system and heat for Craiova. The flow of fuel depends on the momentary load of the boiler and fuel quality. SE Craiova II using coal, 97% of the total quantity of fuel (Oltenia lignite basin) with the carrier oil (0.20% of the amount of fuel used) or natural gas (approximately 3% of the amount of fuel used) (Fig. 2).

Studies were undertaken during the period 2017-2018 and aimed at the following parameters: temperature, humidity, SO<sub>2</sub>, NO<sub>2</sub>, NH<sub>3</sub>, carbon monoxide, carbon dioxide, dust, fly ash, PM<sub>10</sub>, sediment particles, etc. As a result of the technological process within CET II Craiova, there results ash and slag, which represents debris consisting mainly of calcite, pyrite and other minerals, which during the combustion of coal subdivide and decompose. Clay particles remaining in the combustion zone for a sufficient time become complex silicates, molten glass-like. The mineralogical analyzes of slag and ash from the burning of inferior coals indicate the presence of compact, round aggregates along with other laced sponge aggregates.

In accordance with the mineralogical data, the ashes show a 10-16% crystalline phase (quartz, mullite, hematite and magnetite) and a vitreous phase of 50-70%. The global analyzes show that the lignite ash produced in the Oltenia carboniferous zone has the following oxidic composition: SiO<sub>2</sub> 48%, Al<sub>2</sub>O<sub>3</sub> 23%, Fe<sub>2</sub>O<sub>3</sub> 8,1%, CaO 9,2%, MnO 3%, SO<sub>3</sub> 3,7%, ash is silicoaluminous.

The research carried out on the level of radioactivity of the ashes produced by CET Craiova II is as follows (Eq/kg): Ra-226 137,4±16,5, Th-232 82,9±14,9, K-40 520,9±52, α-global 554±100, β-global 1070±117, which revealed a radionuclide content 2-4 times higher than standard materials (POPA et al., 2008).

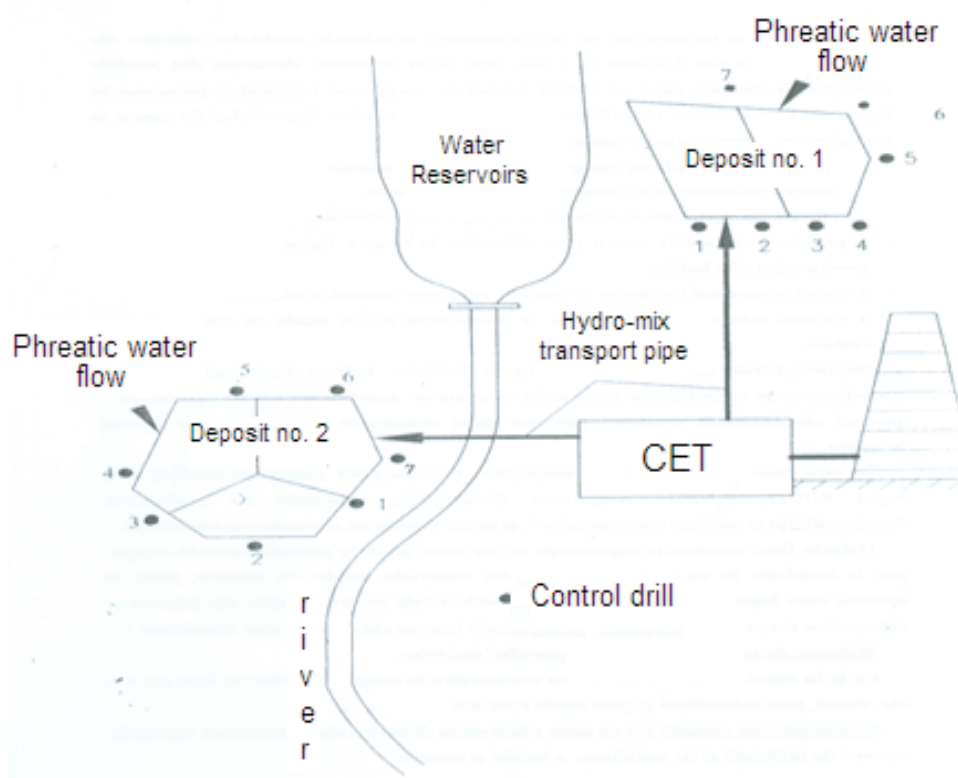


Figure 1. Scheme for operation of CET II Craiova Thermolectric Power Plant (after GAVRILESCU, 2007).



Figure 2. The satellite image of the Deposit from Monastery Valley within Oltenia Power Complex (from Google Earth, accessed: March 11, 2018).

## RESULTS AND DISCUSSIONS

The legal framework of environmental protection aims at preventing and reducing pollution of any kind, conservation and preservation of environmental quality, management responsible for natural resources and avoid overexploitation of ecological reconstruction of the areas affected by pollution from human activities and natural phenomena destructive and not last, maintaining a balance between the natural environment and the quality of life.

In the case of SE Craiova II, air quality indicators have been studied, this being the most important environmental factor for the transport of pollutants, as they represent the support of the fastest transport to the environment, so that air quality monitoring is on first place in monitoring activity.

The gaseous pollutant concentration determined in 2017 is influenced by the ability of plant tissue to convert  $\text{SO}_2$  into relatively non-toxic forms. Sulfite ( $\text{SO}_3^{2-}$ ) and sulphite acid ( $\text{HSO}_3$ ) are the main compounds formed by the dissolution of  $\text{SO}_2$  in aqueous solutions. Phytotoxic effects are diminished by their conversion by enzymatic and non-enzymatic mechanisms into sulfate, which is much less toxic than sulfite. As it can be seen, MAC (Medium concentration) in case of  $\text{SO}_2$ ,  $0.17 \text{ mg/m}^3$  is not found in any month (Tables 1; 2). Up to certain levels, nitrogen oxides (MAC- $0.30 \text{ mg/m}^3$ ) have a beneficial effect on plants, contributing to growth. In these cases, however, it has been observed an increase in the susceptibility to insect infestation and environmental conditions. As for ammonia, the values are below MAC- $0.30 \text{ mg/m}^3$ . Sometimes the gases can trap dust and other particles that reach the ground in dry form. Acid deposits can also occur at variable distances, generally difficult to sample (GAVRILESCU & GAVRILESCU, 2009; RODRÍGUEZ-MAROTO et al., 2009).

Table 1. Concentrations of gaseous pollutants determined in 2017.

Month for the determination of MAC	Gaseous pollutants – average samples for 30 min. – MAC ( $\text{mg/m}^3$ )		
	$\text{SO}_2$	$\text{NO}_2$	$\text{NH}_3$
Medium concentration I	0.0057	0.0103	0.0525
Medium concentration II	0.0048	0.0138	0.0467
Medium concentration III	0.0049	0.0070	0.0189
Medium concentration IV	0.0050	0.0046	0.0145
Medium concentration V	0.0049	0.0044	0.0220
Medium concentration VI	0.0037	0.0106	0.0528
Medium concentration VII	0.0036	0.0095	0.0779
Medium concentration VIII	0.0044	0.0045	0.0904
Medium concentration IX	0.0039	0.0082	0.0696
Medium concentration X	0.0050	0.0089	0.0466
Total MAC	0.75	0.30	0.30

Table 2. Content in sedimentable dusts recorded in 2017.

Harvesting points	Quantity of powders ( $\text{g/m}^2/\text{month}$ )									
	I	II	III	IV	V	VI	VII	VIII	IX	X
P1	3.76	3.62	7.39	6.73	14.66	6.41	7.89	8.07	10.02	6.17
P2	4.40	3.66	5.86	7.57	9.60	6.84	7.20	7.67	7.77	-
P3	5.32	3.31	12.48	11.26	18.08	11.55	22.82	11.99	18.52	6.57
P4	5.45	3.27	6.61	13.18	5.00	4.58	13.84	6.72	9.97	3.70
P5	14.66	2.85	11.33	8.72	13.63	15.12	25.29	5.70	14.15	7.34
MAC $17 \text{ g/m}^2/\text{l}$										

The maximum concentration of SO<sub>2</sub> recorded in 24 h to 200 m has a maximum CET of 18 μg/m<sup>2</sup>. As the distance from the source increases, the concentration decreases, reaching up to 2 μg/m<sup>3</sup>, exceeding the MAC 125 μg/m<sup>3</sup>.

The total concentration of particulate matter TSP does not exceed the maximum permissible concentration (MAC), but it is above the threshold for alert over a distance of 1000 m (NE and SW). The average concentration exceeds MAC 24 h over a distance of 1000 m (NE) and the alert threshold PA, the distance of 750 m SV (Table 3). The temperature is between 54.2 and 60.8°C and humidity between 10 and 13-19.2% V (Fig. 3).

Table 3. Total concentrations of TSP suspension powders.

Distance to the perimeter limit of the heap and the sector (m - sector)	Concentration / concentration range (μg/m <sup>3</sup> )	Health alert threshold (PA) (μg/m <sup>3</sup> )	Limit value = Health Intervention Threshold (VL/PI) (μg/m <sup>3</sup> )	Vegetation Protection Limit Value (VLV) / Ecosystems	Observations
	2	3	4	5	6
Mediation time 1 h					
0-500 m, NE	430-410	350	500	-	< MAC > PA
0-1000 m, SV	400-350				< MAC > PA
500-1000 m, NE	410-380				< MAC > PA
1000-1500 m, SV	350-320				< MAC < PA
1000-3000 m, NE	380-280				< MAC > PA
1500-2500 m, SV	320-260				< MAC < PA
> 3000 m, NE, >2500 m, SV	<280, <260				< MAC < PA
0-500 m NV, SE	400-350				< MAC > PA
500-1500 m, NV, SE	350-290	< MAC < PA			

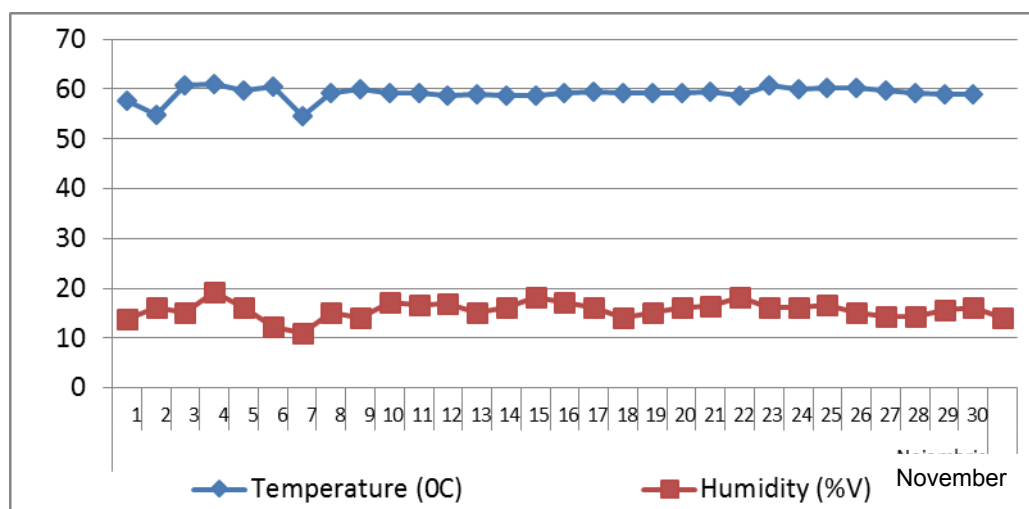


Figure 3. Temperature and humidity variation in November 2017.

NO<sub>x</sub>, SO<sub>2</sub> and CO do not exceed the allowed MAC (Fig. 4). CO<sub>2</sub> emissions from plant activity for the period 2013-2020 are covered by GHG Authorization (greenhouse gases) issued by the Ministry of the Environment and are within the permissible limits. Dust has values of 5.21-9.66 mg/Nm<sup>3</sup> (Fig. 5).

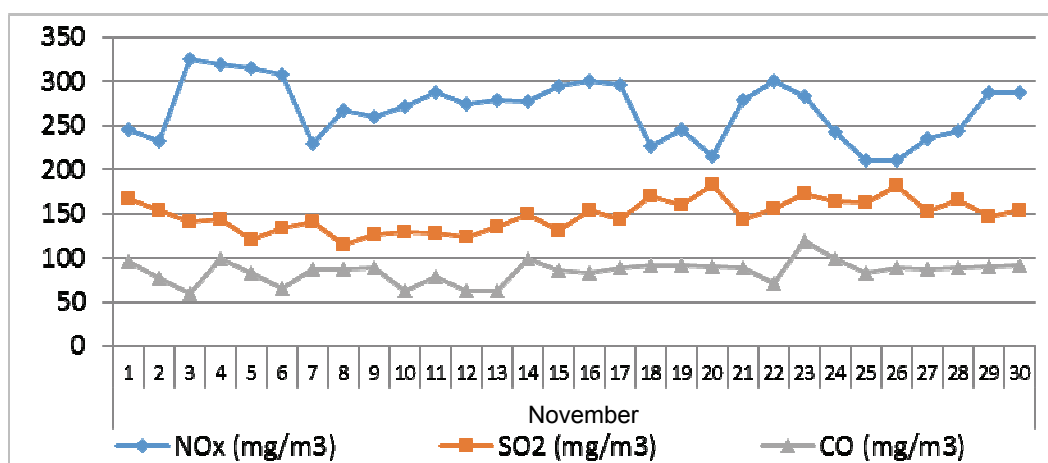


Figure 4. Variation of nitrogen oxides (NO<sub>x</sub>), SO<sub>2</sub> and CO sulfur in November 2017.

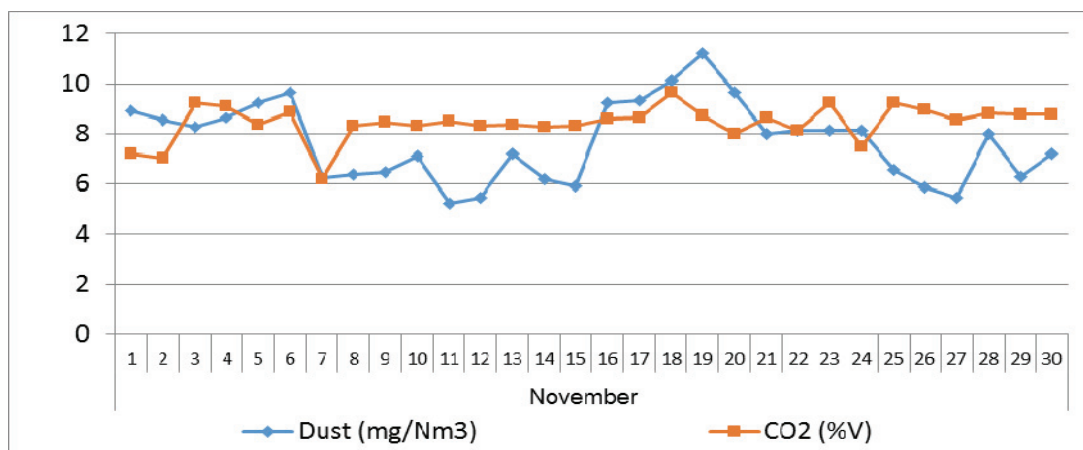


Figure 5. Variation of dust and CO<sub>2</sub> in November 2017.

Ash mg/Nm<sup>3</sup> is between 712 and 987 KNm<sup>3</sup>/h. Ash contains also PM<sub>10</sub>, which are aggregates containing hundreds of individual compounds, compounds with different chemical and thermodynamic properties. The main compounds found in PM<sub>10</sub> are sulfates, nitrates, organic carbon, elemental carbon, soot, ammonium, etc. However, neither the primary biological fraction should be omitted. Organic compounds represent about 21-39% of the material particles (Figs 6, 7).

The STAS 12574-87 provisions shall be complied with at the site of the power plant, as follows:

- sedimentable dusts - max. 17 g/m<sup>2</sup>/month;

- powder in suspension: 0.5 mg/m<sup>3</sup> at 30 minutes (short-term average) and 0.15 mg/m<sup>3</sup> at 24 h (long-term average).

Activity of the site or operation and exploitation must be carried out in such a way that the emission of pollutants that may affect directly or indirectly the soil quality at the site and in the immediate vicinity, to respect the maximum permitted concentrations of heavy metals (Co, Cr, Cu, Mn, Ni, Pb, Zn) provided by OMAPPM 756/1997.

The environmental impact assessment is based on: the quality indices of environmental factors (Ic), which method uses a matrix; the global pollution index (IPG), for which V. Rojanschi method is used (ROJANSCHI et al., 1997).

**The impact assessment matrix, the establishment of the Ic quality index.** The quality of an environmental factor or element environment fall against the permissible limits STAS - sites or regulatory basis, or expected effects of the project on the environment, based on - the size of which is determined taking into account the level of indicators quality that characterizes the effects. The impact assessment is thus carried out by a quantitative analytical method based on the Global Pollution Index (IPG), resulting from the ratio between the ideal state (natural) and the real (polluted) state (RUSU & ROJANSCHI, 1980; GAVRILESCU, 2007). According to the IPG, there is a quality scale (Table 4).

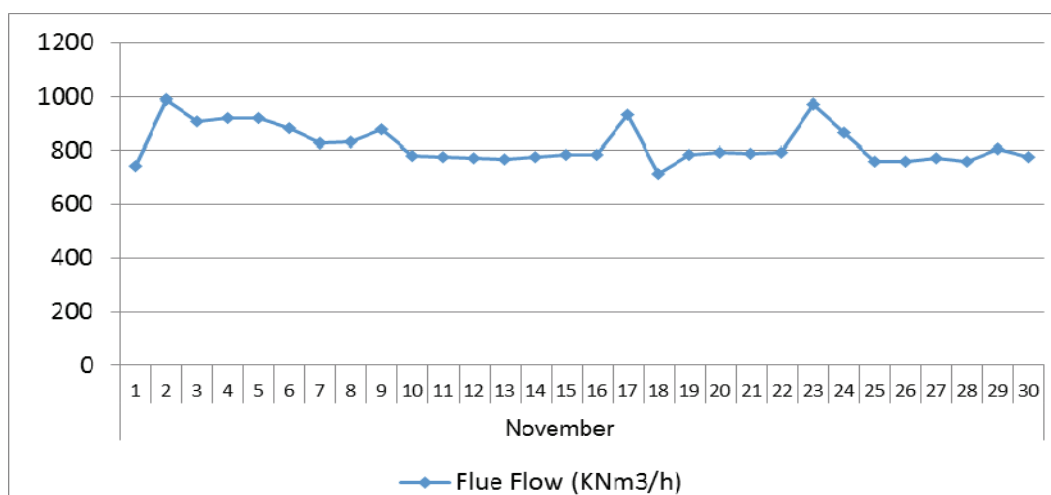


Figure 6. Variation of ash in November 2017.

Table 4. Emissions directed into the atmosphere.

Combustion plant	Pollutant	Fuel type	Location of the broadcasting point	Va Emission limit values ** (mg / Nm <sup>3</sup> ) according to Act 278/2013, Annex V, Part 1, P ≥ 300 MWt
IMA A 1 – existing installation (boilers K <sub>1</sub> , K <sub>2</sub> )	SO <sub>2</sub>	Sol Solid (lower lignite)	Co Dispersion point for the desulphurization plant	20
	PP Powders			20 (starting from 01.07.2020 to 30.06.2020)
				20

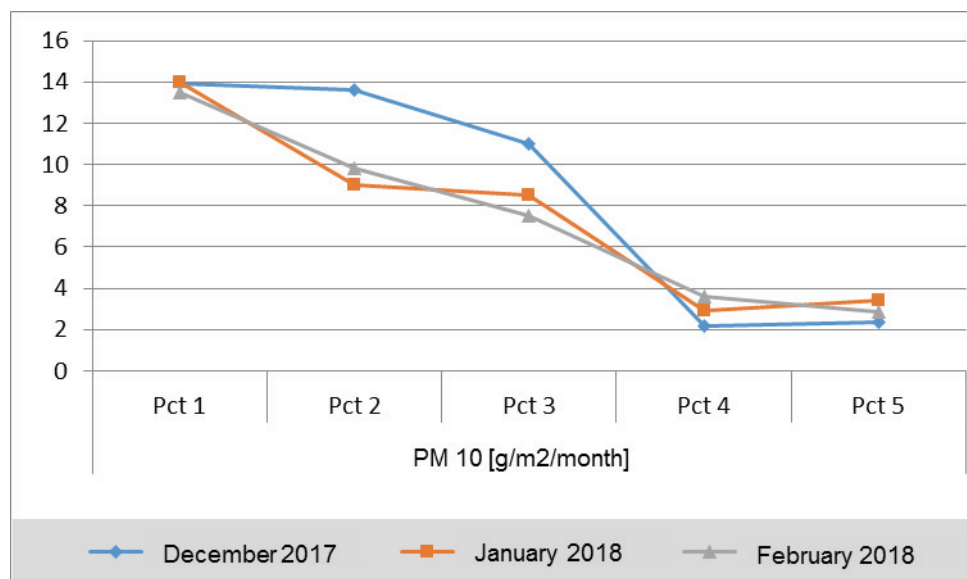


Figure 7. Variation of material powders in December 2017, January-February 2018.

**The air quality.** Emissions from work carried out in the installation must not cause air quality alteration by falling below the limit values set at Activity-Specific Indicators, in accordance with Law 104/2010 on air quality. The use of dense fluid deposition technology leads to a significant decrease in the surface of the slag and ash deposits of the dips. Measures will be taken to reduce spillages from the site by aspiration and / or spraying of dust deposition areas.

The STAS 12574-87 provisions shall be observed at the limit of the power plant location, as follows:

- sedimentable dust - max. 17 g/m<sup>2</sup>/month;
- powder in suspension: 0.5 mg/m<sup>3</sup> at 30 minutes (short-term average) and 0.15 mg/m<sup>3</sup> at 24 h (long-term average).

The microbiological process for removing sulfur from coal requires simple equipment, low amount of reagents, but a long time of water treatment resulting from this process, water with a high ferrous sulphate content. The microbiological process is feasible in reactors when the particle size is less than 0.5 mm. At particle sizes greater than 0.5 mm, coal can be treated in piles.

Conventional technologies for the removal of pyrite from coal have as a general principle the elimination of pyritic inclusions by physical separation, especially of large inclusions that require a long biodegradation period. Pyrite sulphide (FeS<sub>2</sub>) initially can be in the form of 2 crystal crystalline structures: pyrite and marcasite. They are different from the crystallographic point of view, chemically and thermodynamically, but have the same chemical formula. From the chemical point of view, pyrite is closer to the ideal formula FeS<sub>2</sub> than marcasite, in which sulfur deficiency was observed. At the same time, both pyrite and marcasite are attacked and decomposed by concentrated H<sub>2</sub>O<sub>2</sub>, but deposition of colloidal sulfur is observed only at pyrite.

In coal, pyritic sulfur is present in different amounts and forms, from very fine microscopic particles dispersed in the coal mass to large granules of several millimeters. The pyrite is deposited on the cleavage planes, fills the vertical cracks, spreads very finely, filling the walls and the inside of the fused cells. The larger particles, the coarser pyrite tend to sit or lie close to the roof of the coating. According to their appearance, the coal pyrites were classified as crystalline, massive, nodular, lamellar and globular. Pyrites have a high degree of purity; those unrelated to the carbon or silicon material are rarely found in coal and only in relatively large pieces. Massive nodules and additive pyrite can be easily removed by washing, while finely disseminated pyrite may be removed by the process substantially. This is particularly addressed to biological treatment.

Following the quality protection of all monitored environmental factors, we have proceeded to the elaboration of strategies and programs for the refurbishment and modernization of the electrical and thermal energy installations and equipment, respectively for the optimization of the existing technological processes or for the adoption and implementation of the best available techniques without the cost of excessive costs, as is provided by the current legislation (Romanian and European Union) (ZARNEA, 1994, ROJANSCHI et al., 1997, MIHAI et al., 2008, DEAK et al., 2009; GHIORGHITĂ et al., 2014).

In order to prevent the limitation and elimination of the impact of fossil fuel (coal, fuel oil, methane gas) production of electricity and heat, it is necessary to supply and use fuels, fuels and lubricants with low content of pollutants (S, N, P, heavy metals) according to standards and technical requirements, namely that of the legal regulations in force (quality standards, norms, specific instructions) (CISMAȘIU 2003; POPEA et al., 2004; CISMAȘIU 2010a, b; JACOB et al., 2018).

The optimization of the existing production processes consists of:

- upgrading and re-technologization the neutralization dilution and dispersion facilities and equipment in the environmental factors;
- adoption of best available techniques without involvement of excessive costs in the drafting and development programs;
- monitoring quality parameters of all environmental factors both on site and in its immediate vicinity;
- compliance measures and the conditions imposed by the competent organisms during checks conducted at the site, and the provisions of plans to prevent and limit pollution accidents, or of the action in the event of disasters or natural phenomena special (earthquakes, floods, abundant rainfall, strong winds, drought, etc.).

As possibilities to reduce the phenomenon of drifting, it is expected to stabilize dry crust that forms on the surface of the active deposit (bituminization, polymerization, silicification), during the periods when the conditions for the manifestation of this phenomenon are fulfilled, in order to prevent its occurrence. Placing trees around the deposition site is considered sufficient to reduce the amount of slag and ash scattered by the wind to a level that presents minimal health risks to the population. Also after the storage capacity is exhausted, the deposit area will be returned to the productive circuit, is the technical mining and the biological recultivation for the establishment of the tree plantations (\*\*\*. 2013; TOMUȘ et al., 2015; CIOBOIU et al., 2017; CISMAȘIU et al., 2017; GAVRILESCU et al., 2017).

### CONCLUSIONS

The emissions resulted from the activity in the system should not result in alteration of air quality by falling below the limit values set for activity-specific indicators, according to Law 104/2010 on air quality. Measures will be taken to reduce spillages from the site by aspiration and / or spraying of the soil deposition areas.

The provisions of STAS 12574 - 87 shall be respected at the site of the power plant: sedimentable dusts - max. 17 g/m<sup>2</sup>/month, suspension powders: 0.5 mg/m<sup>3</sup> at 30 minutes (short-term average) and 0.15 mg/m<sup>3</sup> at 24 h (long-term average).

The operation of large combustion plants is permitted in compliance with the special provisions and the conditions stipulated in Law no. 278/2013 on industrial emissions, which transposes Directive 2010/75 / EC.

The operator shall, in normal operating conditions, emission limit values for large combustion plant IMA 1-existing - type I set given by Law no. 278 / 24.10.2013 on industrial emissions. No emissions to the air shall exceed the set emission limit value. It is mandatory that there is no other significant air emissions to the environment except for those legally accepted.

The CO<sub>2</sub> emissions from the plant's activity for the period 2013-2020 are covered by GHG Authorization (greenhouse gases) issued by the Ministry of the Environment.

### ACKNOWLEDGEMENT

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