Research on emissions from large combustion plants (LCP). Case Study.

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Abstract. According to Directive 2010/75 / EU and Law 278/2013 on industrial emissions, operators of large combustion plants (IMA) must carry out continuous self-monitoring of pollutant emissions and are required to perform parallel measurements with accredited laboratories, to certify results. Thus, INCD INSEMEX Petroşani, through the Testing Laboratories Group, accredited by RENAR, offers these type services to various economic agents. The curent paper presents measurements performed at two IMA exhaust chimneys, in two different stages, to quantify pollutants released into the atmosphere and to establish the impact they generate on health of population in the area of the study. Analysis of results showed that the maximum allowed value for the SO_2 was exceeded, so that, at the end of this paper, some recommendations were reviewed, according to BAT (Best Available Techniques), to support the economic agent and inhabitants of the area in haveing a cleaner environment.

1 Introduction

It's a well-known fact that industrial processes cause significant emissions of greenhouse gases into the atmosphere, which greatly contribute to the phenomenon of global warming. [1]. For this reason, reducing or stopping these emissions is also reflected in the European Green Pact, which highlights the new strategy of the European Union, without greenhouse gas emissions until the middle of the century, the year 2050 [2].

Air pollution is the number one environmental factor causing bad health and/or mortality worldwide, causing approximately 7 million deaths per year. In the 28 countries of the European Union, following exposure to particulate matter (PM), ozone (O3) and nitrogen dioxide (NO2), air pollution is thought to be responsible for 483,400 deaths per year [3] causing significant damage to the environment as well. Economic activity and its recent fast development, entails an even more accentuated pollution of all environmental components, directly affecting them.

Large combustion plants (LCP) have a total rated thermal input of 50 MW or more, regardless of the type of fuel used [4]. Emissions generated by these installations require

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periodic measurements in order to have knowledge of the degree of air pollution in the installation's area of activity.

For continuous monitoring of air quality in the area of interest, an activity plan must be drawn up and followed, including inspection, surveillance and regular evaluation.

Emissions monitoring can be performed by the economic agent (self-monitoring) or by specialized, accredited laboratories. Measurements shall be made following procedures in accordance with regulations and standards for general measurement requirements [5].

Operators of large combustion plants are required to monitor emissions from large combustion plants, legal obligations being provided by Law no. 278 of October 24th, 2013. If the LCP operators cannot comply with the emission limit values (ELV), they are obliged to comply with a certain desulphurisation rate, under the conditions of law 278/2013, art. 30 para. (3) and (4). To determine the desulphurisation rate, the SO2 concentration of the waste gas must be measured before and after the desulphurisation equipment.

Auxiliary, sulphur content of the fuel is determined on a regular basis, under the approval of the competent authority for environmental protection. Measurement of sulphur content in ash is also indicated [6].

As known, emission measurements are the direct responsibility of the polluting agent (economic agent), also being an indicator of the technological process.

2 Materials and methods

The method for determining gaseous components of industrial gas emissions is the direct method with multi-gas (type TESTO 350XL) analyser, which has a numerical display and determines flue gas concentrations (NO_2, NO, CO, SO_2) (fig. 1.a-b).

a. b.

Fig. 1.a. Flue gas sampling; **b.** Multigas analyser.

Assays are sampled with the help of the appliance's internal pump and on the suction route the gas is cooled, water vapours being condensed, directly resulting in the lowest absorption of humidity by NO_2 and SO_2. Condensation is pumped at equal intervals into the dryer installed in the body of the equipment.

The dry gas is passed through filters to retain suspensions. Filters also have the role of retaining water vapours. If the filters are soaked with water, their pores close and protect the pump and internal sensors. A very small part of the gas passes to the sensor membranes, where the gas concentration is transformed into an electrical signal, the excess gas being eliminated.

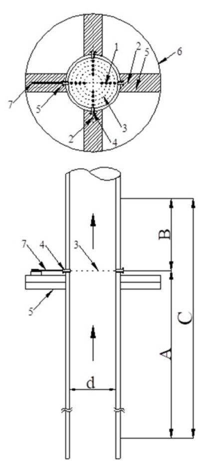
The TESTO 350 XL portable analyser (fig. 2) equipped with a sampling probe, control unit and analysis unit, has a stainless-steel metal probe for sampling, provided on the inside with a thermocouple for temperature determination.

Fig. 2. Testo type 350 gas component analyser.

Determination of gaseous components' concentration in industrial emissions is influenced by unfavourable weather conditions (strong wind, precipitation). Sampling location is chosen so that results are representative of the emission behaviour of respective installation, in accordance with the Standard SR EN 15259 of March 2008 [7] (fig. 3), respectively:

- lengths of the rectilinear sections before (upstream) and after (downstream) the location of measuring point should be at least 5, respectively 3 times the equivalent of the hydraulic diameter of the measuring section. Thus, the upstream section must be longer than the downstream section.

- the sampling probe is inserted into the chimney's measuring hole, so as to allow a representative emission sample to be taken in the sampling plan.



1 - measuring point;
2 - measuring line;
3 - measurement plan;
4 - measuring hole;
5 - free surface;
6 - place of measurement;
7 - measuring equipment;
A - input section;
B - output section;
C - measuring section;
d - the inner diameter of the basket.

Fig. 3. Place of measurement and related section.

3 Results and discussion

In 2021, the INCD INSEMEX Petroșani team, through the Test Laboratories Group, carried out measurement campaigns at a large combustion plant, working on fuel oil, which is a liquid fuel of petroleum origin used in thermal power plants, boilers and reservoirs. It's worth mentioning that the LCP installation is very old and has not undergone improvements for a long time, for economic and financial reasons. For objective reasons we will not provide the company's name.

The measurements were performed on two LCP chimneys, both 6.2 m high and having a diameter of 2.1 m, in two stages, in two different measurement days, namely between 15 ÷ 16.11.2021 and 14 ÷ 15.12.2021. During measurements, the combustion plant operated in nominal parameters. In order to be able to fully describe the gaseous effluent and to be able to report the monitored quantities under reference conditions provided by legislation, the following physical parameters of waste gases need to be monitored: temperature, pressure, humidity, speed, flow and oxygen and carbon dioxide concentration, from the source (chimney). Carbon content of the raw material used (fuel oil) for LCP is 87.26%.

Fuel oil fired boilers are operated according to thermal power plant's needs to produce thermal agent for the city's needs. Fuel oil burners are devices fitted on water heating boilers and run-on heavy liquid fuel. These systems have the ability to operate in stages and operate the same way as gas burners. They operate in several velocity steps with mechanical spraying and pressure drop.

According to requirements of the National Agency for Environmental Protection, the economic operator has the obligation to monitor gas emissions, through an accredited laboratory according to legal provisions. These measurements must be within the limits provided by law no. 278 of 24th October 2013 on industrial emissions which sets the emission limit values for large combustion plants for each gaseous component.

Table no. 1 shows the results of the first set of measurements performed on both boilers.

Table 1. Tests results of the first set of measurements

No.	Installation name	Measured component	Average value [mg/m^{3}N]	Maximum value [mg/m^{3}N]	Observations (Conditions of the process from which the waste gases result)
Test date 15.11.2021					
1	Exhaustion chimney: Boiler no. 1	NO_2	35,4	153,4	$T_{ambiental}$=16 °C; Tefluent=156,1 °C $P_{ambiental}$ = 1013,6 mbar CO_2 = 6,4 %vol; O_2 = 9,7 %vol v = 6,94 m/s; Q = 51.96672 m^3/h λ = 1,88; W = 31,2
		SO_2	547,4	644,7	
		CO	9	31	
2	Exhaustion chimney: Boiler no. 2	NO_2	413	472	$T_{ambiental}$=13,9 °C; $T_{efluent}$=154,7 °C $P_{ambiental}$ = 1016 mbar CO_2 = 7,4 %vol; O_2 = 8,0 %vol v = 9,23 m/s; Q = 115.030,35 m^3/h λ = 1,6; W = 38,6%
		SO_2	765,2	816,5	
		CO	SLD*	SLD*	

Test date 16.11.2021					
3	Exhaustion chimney: Boiler no. 1	NO_2	78,5	102	$T_{ambiental}$=13,4 °C; $T_{efluent}$=147,9 °C $P_{ambiental}$ = 1017,4 mbar CO_2 = 7,1 %vol; O_2 = 7,82 %vol v = 7,59 m/s; Q = 56.833,9 m^3/h λ = 0,98; W =41,0%
		SO_2	551,9	609,5	
		CO	36,3	77	
4	Exhaustion chimney: Boiler no. 2	NO_2	55,6	58,0	$T_{ambiental}$=15,1 °C; $T_{efluent}$=155,1 °C $P_{ambiental}$ = 1016,3 mbar CO_2 = 7,2 %vol; O_2 = 9,1 %vol v = 11,05 m/s; Q = 137.712,4 m^3/h λ = 0,99; W = 40,2%
		SO_2	442,6	539	
		CO	1,2	3	

Observations: *SLD – under detection limit
$T_{ambiental}$ – environmental temperature, $T_{efluent}$ – effluent temperature, $P_{ambiental}$ – environmental pressure, v – effluent speed, λ – excess air coefficient, Q – effluent air flow, W – air relative humidity.

Concentrations shown are related to normal conditions of temperature (273.15 K), pressure (1013 mbar) and reference O_2 (3%). Detection limits for the TESTO 350 XL are: CO = 5 ppm, NO_2 = 5 ppm, SO_2 = 3 ppm.

Measurement uncertainty: CO = ± 7,68 mg/Nm^3, NO_2 = ± 14,48 mg/Nm^3, SO_2 = ± 19,76 mg/Nm^3.

Emission limit values for furnaces fuelled by liquid fuel (fuel oil), according to Law 278/2013 are: CO = 170 mg/m^{3}N, SO_2 = 350 mg/m^{3}N, NO_2 = 450 mg/m^{3}N.

Measured concentrations for boilers 1 and 2 exceed emission limit values for parameter SO_2, having 1.2, 1.5 times higher values.

Table no. 2 shows results of the second set of measurements (14 ÷ 15.12.2021) performed on both boilers.

Table 2. Tests results of the second set of measurements

No.	Installation name	Measured component	Average value [mg/m^{3}N]	Maximum value [mg/m^{3}N]	Observations (Conditions of the process from which the waste gases result)
Test date 14.12.2021					
1	Exhaustion chimney: Boiler no. 1	NO_2	87,9	122,8	$T_{ambiental}$ = 8,6 °C; $T_{efluent}$ = 146,2 °C $P_{ambiental}$ = 1010,9 mbar CO_2 = 7,4 %vol; O_2 = 8,1 %vol; v = 6,96 m/s; Q=110.496,96 m^3/h λ=1,67; W = 39 %
		SO_2	594,3	629,7	
		CO	6	28	
2	Exhaustion chimney: Boiler no. 2	NO_2	197,8	216	$T_{ambiental}$ = 11,9 °C ; $T_{efluent}$ = 133,2 °C $P_{ambiental}$ = 1010,6 mbar CO_2=6,7 %vol; O_2=8,9 %vol; v = 8,31 m/s; Q = 131.929,56 m^3/h λ = 1,87; W = 43 %
		SO_2	715,8	794,8	
		CO	SLD*	SLD*	

Test date 15.12.2021					
3	Exhaustion chimney: Boiler no. 1	NO_2	72,1	89,5	$T_{ambiental}$ = 11,5 °C; $T_{efluent}$ = 149,3 °C $P_{ambiental}$ = 1012,4 mbar CO_2 = 7,9 %vol; O_2 = 8,2 %vol; v = 7,62 m/s; Q = 120.975,12 m^3/h λ = 1,72; W = 45 %
		SO_2	514,6	591,6	
		CO	7,4	9,2	
4	Exhaustion chimney: Boiler no. 2	NO_2	114,9	148,2	$T_{ambiental}$ = 9,3 °C; $T_{efluent}$ = 138,6 °C $P_{ambiental}$ = 1012,3 mbar CO_2 = 7,5 %vol; O_2 = 7,8 %vol v = 8,57 m/s; Q = 136.057,32 m^3/h λ = 1,61; W = 48 %
		SO_2	792,8	835,4	
		CO	SLD*	SLD*	

Observations: *SLD – under detection limit
Detection limits for the TESTO 350 XL: CO = 5 ppm, NO_2 = 5 ppm, SO_2 = 3 ppm.

As for the first set of measurements, the measured SO2 concentrations for CLP, boilers 1 and 2 exceed the emission limit values.

In order to be able to observe more clearly the evolution of sulphur dioxide (SO_2) concentrations during measurements, see fig. 4.

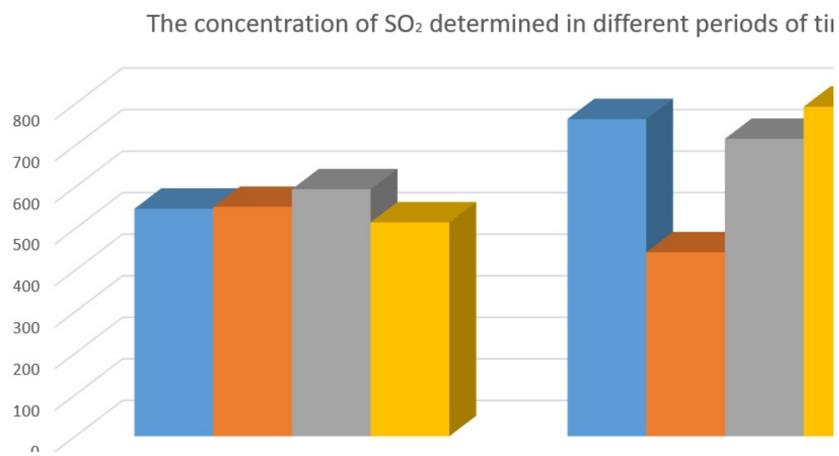


Fig. 4. Concentrations of SO_2 determined in different periods of time.

Fig. 4 shows that at the exhaustion chimney boiler no. 2, higher concentrations were measured than for boiler no. 1, and the highest value recorded was 792.8 mg/m^3N, a value 2.26 times higher than the maximum limit allowed by Law 278/2013 [8].

These exceedances have an impact on health of the population in the studied area, one of the reasons why we approached this topic.

Depending on concentration and exposure time, sulphur dioxide has different effects on human health. Exposure to a high concentration of sulphur dioxide over a short period of time can cause severe breathing difficulties. People with asthma, children, elderly and people with chronic respiratory diseases are particularly affected. Long-term exposure to low sulphur dioxide can result in respiratory tract infections. Sulphur dioxide can potentiate the harmful effects of ozone [9].

Finally, we refer to a recent ranking on air quality, where Romania ranked 15^{th} among the most polluted countries in Europe (global platform on air quality IQAir, 2020) [10].

In a similar paper, the authors concluded that SO2 emissions do not fall within the limit values set by Romanian regulations, because of the low calorific value fuel, and measured levels of NOx emissions were found to be within limit values, according to Romanian regulations, for the entire monitoring period [11].

4 Conclusions and recommendations

A major environmental issue are large combustion plants. Through chimneys, thermal power plants continuously and constantly release large volumes of flue gases into the atmosphere, containing concentrations of polluting gases and particles. Emissions generated by LCP must be measured periodically in order to have knowledge of the current degree of damage to air quality and thus to health of population.

Measurements were performed on two chimneys of a company's large combustion plant, being performed in two stages, in two different measurement days, each. The measured concentrations for boilers 1 and 2 exceed emission limit values for the SO_2 parameter (SO_2 = 350 mg/m^{3}N), having values 1.2 - 2.26 times higher.

Because of sulphur dioxide concentrations measured at the chimneys of the analysed plants, setting up a flue gas desulphurisation system is recommended, consisting of one or more emission abatement methods, namely the use of a dry, semi-wet or wet desulphurisation process, to comply with emission limits provided by national legislation. It is also recommended to use a more "sulphur deficient" fuel, to reduce SO_2 emissions at source.

In addition to these recommendations, the use of Best Available Techniques (BAT) solutions should be taken into account to support the economic operator and the locals in order to have a cleaner environment [12].

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