#### COMPARATIVE ANALYSIS OF METHODS FOR MONITORING CO<sub>2</sub> EMISSIONS FROM THE ENERGY SECTOR IN THE CONTEXT OF CLIMATE CHANGE

Mona Barbu; Elena Bucur; Mihai Bratu

#### National Research and Development Institute for Industrial Ecology – ECOIND, 71-73 Drumu Podu Dambovitei St., sector 6, Bucharest, Romania e-mail: ecoind@incdecoind.ro; poluare.aer@incdecoind.ro

#### Abstract

In an economy increasingly globalized a country's energy strategy is done in the context of changes and developments taking place worldwide. Due to increasing energy demand, energy has developed rapidly without the problem of environmental protection. [1]. Energy sector is the largest sector in the emissions of greenhouse gases, responsible for 66,44% of total emissions of greenhouse gases generated nationwide in 2009. Total energy demand in 2030 will be around 50% higher than in 2003 [1]. European Commission proposes a set of documents representing the new energy policy of the EU, increase emissions of greenhouse gases by 20% by 2020 compared to 1990. [2; 3]. Monitoring and reporting of greenhouse gas (GHG) is the basis for the (EU ETS) Romania, has undergone two phases of the EU ETS, first phase was performed in 2005-2007 in accordance with the Order of 1175/2006 and in the second first phase, in 2008-2012 in accordance with Decision 589/2007.[4; 5]. In the third phase of monitoring, starting on 01.01.2013 in all EU Member States nr.601/21.06.2012 rules apply. The paper proposes an application in the energy sector, for the evaluation of CO<sub>2</sub> emissions using both calculation data from those laboratory investigations and analytical determined bv direct measurement of the source. From research carried out it was determined the method for assessing CO<sub>2</sub> is direct measurement and for reporting and monitoring that both methods can be used (direct measurement and calculation), the results being comparable, the differences in uncertainty margin were under 10%, acceptable for automatic measurements [5; 6].

**Keywords**: monitoring, reporting, measuring emissions of greenhouse gases

#### INTRODUCTION

In an increasingly globalized economy, a country's energy strategy is done in the context of developments and changes taking place in the world. Due to the increasing demand for energy, this area has developed rapidly without solving the environmental question. [1] The energy sector is the largest sector in the emissions of greenhouse gases, (GES) being responsible for 66.44% generated nationwide in 2009.For 2030 it is expected that total energy demand will be about 50% higher than in 2003. [1]. The European Commission proposes a set of documents representing the new energy policy of the EU, reducing the emissions of greenhouse gases by 20% by 2020 compared to 1990. [2, 3].

According to the national inventory of emissions of greenhouse gases, produced by our country in 2012, GHG emissions related to the energy sector accounted for about 87% relative to 2010. The European Commission has approved, in March 2011, a reduction of greenhouse gas emissions by 80-95% by 2050 compared to 1990, which will prevent long-term negative consequences of climate change. [7, 8]

By the means of the adopted strategic document, energy will be ensured through the implementation of technologies based on low carbon. It is also encouraged the use of renewable energy to produce electricity and heat. [9, 10]

Monitoring and reporting of greenhouse gas (GHG) is the basis for the EU ETS GHG emissions trading (EU ETS). Romania, has undergone two phases of the EU ETS witch was performed in 2005-2007 in accordance with the order of 1175/2006 and in the second period, 2008-2012 in accordance with Decision 589/2007. In the third phase of monitoring, starting on 01.01.2013 the Regulations nr.601/21.06.2012 are applied in all EU Member States.

The report on carbon market presented in April 2012 in the European Parliament and the Council has revealed that over 10% GHG reduction in the second period of emissions trading is mainly due to the economic downturn and not measures implemented to reduce GHG emissions. Since 2013 the functioning of the carbon market will undergo some fundamental changes in order to strengthen and ETS scheme. [6].

GHG Reporting includes specific rules for the electricity sector and / or heat, as well as rules for energy efficiency improvements to the consumer. Monitoring and reporting of greenhouse gas emissions can be achieved using one of these methods [5]:

- Calculation method for determining emissions based on activity streams obtained through measurement systems and additional parameters from laboratory analyzes or standard factors
- Method for measuring emissions using direct measurement of CO2 concentrations and flue gas flow.

The calculation method, applied to the monitoring and reporting of CO2 emissions is based on establishing laboratory analyzes of the physicchemical and energy characteristics of solid fuels used in the installation, such as: technical analysis (moisture and ash), elemental analysis (C, H, S, N) and the determination of upper and lower calorific values. Also important is the determination of unburned carbon present in the slag and ash resulting from coal combustion in the boiler. Based on data obtained from laboratory investigations it is possible to calculate the emission factor and oxidation factor and thus CO2 emissions by using the following relationship:

CO<sub>2</sub> Emissions = Activity Data \* emission factor \* oxidation factor

Direct measurement method consists of gas analysis performed at the emission source with a gas analyzer equipped with special IR cell for measuring CO2 emissions and determination of the residual gas for the combustion installation. Applying the following relationship, CO2 emissions are directly obtained:

 $CO_2$  Emissions=  $CO_2$  measured (kg/Nm<sup>3</sup>) \* Q total gasses (Nm<sup>3</sup>) \* 10<sup>3</sup>

#### EXPERIMENTAL PART

In the experiments, measurements were made at a CHP unit boiler 420t / h steam (Installation category B / IMA 1). Over a period of 24 hours, a set of five separate samples of solid fuel (coal-lignite), slag and ash were taken and also direct measurements of CO2 were made at the dispersion installation, representing the average measurement zones. Solid fuel use at the date of measurement was 2859.44 t / day. In order to use the calculation based method, analytical investigations were performed on samples (solid fuel- slag, coal, and ash). Selected samples were subjected to specific laboratory analysis of solid fuels in order to calculate the oxidation factor, emission factor and to assess the level of CO2 emissions. Tests were performed in accordance with national and international standards in force in Romania recognized [13, 14, 15, 16, 17, 18].

To determine the elemental analysis (CH, S, N) a Thermo Fisher scientific Fleash elemental analyzer was used. EA 1112. Calorific value was determined using a Parr 6200 bomb calorimeter [15,16,17].

Direct measurement was performed in order to determine CO2 emissions in an industrial unit form the energetic sector, on a boiler of 420 t/h steam stray stack with the following constructive characteristics:

- Stack height: 220 m
- Stack diameter 7 m

The fuel used during the measurements was:

- Dual fuel (gas + solid fuel
- Type solid fuel: coal / lignite

CO2 concentration in the source / dispersion stack was measured with an analyzer TESTO 350 XL. [11, 12]. equipped with a special infrared (IR) cell to measure CO2 concentrations. For other gases, the equipment uses specific electrochemical cells for each of the gas.

## **RESULTS AND DISCUSSIONS**

## **Calculation approach**

The analytical results from the test samples were recalculated to the initial state of the fuel in accordance with the standard STAS 398/82. [18]. Table 1 presents the technique and elemental analysis on the 5 samples analyzed.

Droho	Wt	A <sup>i</sup>	Sti	C <sub>t</sub> <sup>i</sup>	Coi	H <sup>i</sup>	Osi	Q <sup>i</sup> i	Q <sup>i</sup> i
FIDDE	%	%	%	%	%	%	Kca	ıl/kg	kJ/kg
		28.4		18.5	14.3				
C-1	34.22	5	1.02	1	9	2.14	1791	1478	6190
		29.0		18.2	14.2				
C-2	34.29	0	1.09	5	4	2.19	1784	1469	6149
		29.2		18.4	14.1				
C-3	34.28	4	1.14	3	9	2.23	1781	1463	6125
		29.7		18.2	14.2				
C-4	34.30	5	1.17	3	3	2.21	1797	1480	6196
		28.9		18.0	14.2				
C-5	34.36	6	1.21	5	5	2.26	1779	1459	6107
		145.		91.4	71.3	11.0			3076
Total	171.45	41	5.63	8	0	4	8931	7348	7
		29.0		18.3	14.2				
Average	34.29	8	1.13	0	6	2.21	1786	1470	6153

 Table 1 – Technical and elemental analysis for solid- lignite fuel.

Also, the content of unburned carbon in the slag and ash samples was determined. The results are shown in Table 2.

Dula			Determined values		
Probe	Indicator	UM	Clay	Ash	
1	Unburned carbon content		6.03	3.38	
2		%	6.21	3.03	
3			6.26	3.31	
4			6.12	3.26	
5			6.05	3.30	
Total			30.67	16.28	
Average			6.13	3.26	

 Table 2 - Unburnt carbon content of slag and ash samples

Following the analytical laboratory investigations the oxidation factor, emission factor and the emission of  $CO_2$  were calculated, respectively, oxidation factor (fo) and emission factor (Fe) are directly influenced by the carbon content and net calorific value. The oxidation factor was calculated using the equation:

$$f_{o} = [1 - (A^{i}/C) * [0,2 * (C_{n}zg/100) + 0,8 * (C_{n}cen/100)]$$
(1)

The emission factor was calculated using the equation:

$$F_{e} = [(Mco_{2}/Mc)x(C^{i}/100)]/ [10^{6}/P_{CN}]$$
(2)

The CO2 emissions were calculated using the following equation [4]:

CO2 emissions = fuel flow (pt) \* calorific value / methane (kJ / kg) \* emission factor (tCO2/TJ) \* oxidation factor (t CO2) (3)

Also information was provided on CO2 emissions from the use of natural gas fuel as support for the combustion of coal. The quantities of CO2 emission from the combustion of natural gas were: Eco2 (natural gas) = 179.42 t CO2Applying the calculation method, using relations (1, 2, 3) we can calculate the oxidation factor (fo), the emission factor (Fe) and total emission of CO2. (Table no. 3).

Table 3 -	Assessment	of CO2	emissions	achieved	by	applying	the	calculation
method								

Nr crt	Fuel Quantity	Net Calorific Power (Qi <sup>i</sup> )	Emission Factor (Fe)	Oxidation	Eco <sub>2</sub> <sub>(</sub> comb)	Eco₂ (gaz nat)	E <sub>CO2</sub> total
	tone	KJ/kg	t CO <sub>2/</sub> TJ		t CO <sub>2</sub>	t CO <sub>2</sub>	(t/zi)
					1392.1		
1	2859.44	6188	85.27	0.9227	1	179.42	1571.53
					1383.6		
2	2859.44	6150	85.01	0.9254	4	179.42	1563.06
					1368.2		
3	2859.44	6125	84.94	0.9196	0	179.42	1547.62
					1372.4		
4	2859.44	6196	84.20	0.9199	3	179.42	1551.85
					1377.1		
5	2859.44	6109	85.54	0.9218	6	179.42	1556.58
Averag					1378.7		
е	2859.44	6154	84.99	0.9219	1	179.42	1558.13

## Measure based method

To measure the concentration of CO2, in the CET were performed 5 sets of measurements to the dispersion stack belonging to a boiler of 420 t / h for a period of 24 hours with a portable flue gas analyzer-TESTO 350 M / XL provided with a special infrared cell (IR) [6]. Measured values represent hourly average values. Measurements were performed for the following parameters: temperature, pressure and analysis of O2 and CO2. Gas analysis (O2, CO2) was performed according to ISO 10396:2008 standard.

Measured values presented in Tables 4 and 5 were recalculated to standard conditions of temperature (273.15 K) and pressure (101,325 Pa).

Moocuromont	Gas	ses	Amb	ient	Absolute
number	Tempe	rature	Tempe	rature	pressure
number	°C	K	оС	K	mbar
1	158.7	431.7	1.8	274.8	995
2	158.8	431.8	2.7	275.7	994.9
3	158.8	431.8	2.7	275.7	995.1
4	158.9	431.9	3.5	276.5	995.2
5	158.8	431.8	2.6	275.6	995.1
Average	158.80	431.8	2.7	275.7	995.1

**Table 4**-Physical parameters - temperature and pressure

Table no. 5 - Analysis of gases (O2, CO2) at the dispersion stack

Nr /Measurement	O <sub>2</sub> (%)	CO <sub>2</sub> (%)	CO <sub>2</sub> (ppm)	CO <sub>2</sub> (mg/Nm <sup>3</sup> )
1	12.99	6.96	69600	136416
2	13.05	6.69	66900	131124
3	13.29	6.76	67600	132496
4	13.1	6.75	67500	132300
5	13.11	6.79	67900	133084
Average	13.11	6.79	67900	133084

In order to assess the CO2 gas flow, the flow of the mixture of solid fuel and the gas flow for the natural gas were calculated.

Gas flow resulting from burning solid fuel mixture:

 $Q_{\text{gases comb}} = \text{Quantity of solid fuel } *2,9*1,4*10^3$  (4)

2,9 Nm<sup>3</sup>= amount of gases from burning solid fuel in a kg stoichiometric conditions

1,4 - air excess coefficient at solid fuel burn, which corresponds to  $O_2$  = approximately 6 % in burned gases[21/(21-6)]

 $Q_{gaze \ comb} = 2859,44^{*}2,9^{*}1,4 = 11609,32^{*}10^{3} \text{ Nm}^{3}$ 

Gas flow resulting from the burning of natural gas:

 $Q_{\text{gases natural gasl}} = \text{Natural Gas Quantity}^{11,70^{*}10^{3}}$  (5)

 $Q_{\text{gases natural gas}} = 72,82^{*}11,70^{*}10^{3} = 852^{*}10^{3} \text{ Nm}^{3}$ 

The total gas flow rate was calculated by the following relationship:

 $Q_{\text{total gases}} = 11609,32 \times 10^3 + 852 \times 10^3 = 12.461,32 \times 10^3 \text{ Nm}^3$ 

Total CO2 emissions were calculated using the equation:

 $E_{CO2} = CO_{2 \text{ measured}} (\text{ kg/Nm}^3) * Q_{\text{ total gases}} (\text{Nm}^3) * 10^3 \text{ t/zi CO}_2$ (6)

Equation (6) was applied in order to calculate the total CO2 emission by means of direct measurement of the dispersion stack. Final results of the measurements are shown in Table nr 6.

Nr			Total
/Measurement	Measur	Emission	
	(mg/Nm <sup>3</sup> )	(Kg/Nm <sup>3</sup> )	CO <sub>2</sub> (t/day)
1	136416	1699923.484	1699.92
2	131124	1633978.176	1633.98
3	132496	1651075.108	1651.08
4	132300	1648632.689	1648.63
5	133084	1658402.364	1658.40
Average	133084	1658402.364	1658.40

Table 6 - Direct measurement method for a	assessing the total CO2 emissions.
---	------------------------------------

Table. 7 shows the measurement results of CO2 emissions obtained by the two methods (measurement and calculation). Picture no. Chart 1 shows the variation of CO2 obtained by both methods.

Replied/	CO <sub>2</sub> emission (t/day)				
determination	Calculated	Measured			
1	1571.53	1699.92			
2	1563.06	1633.98			
3	1547.62	1651.08			
4	1551.85	1648.63			
5	1556.58	1658.40			
Average	1558.13	1658.40			



**Table 7-**CO<sub>2</sub> value by the two methodologies (measurement and calculation)

**Fig. 1** - Variation of the CO<sub>2</sub> emission obtained by the two methods (measurement and calculation)

Table. 8 and Figure 2 shows the average values obtained for total CO<sub>2</sub> emission and the graphical representation of data.

CO <sub>2</sub> Emission(t /day CO <sub>2</sub> )				
Calculation	Measurement			
1558,13	1658,40			

**Table 8** - CO2 Emissionobtained by the two methods:measurement and calculationat the source / dispersionstack - boiler 420 t / day



Figure 2 - Variation of  $CO_2$  emissions achieved by counting and measuring at the source / dispersion stack-boiler 420 t / day

# CONCLUSIONS

Monitoring and reporting of greenhouse gas emissions can be achieved using one of the following methods:

-The standard method of calculation which determines emissions from streams based on activity data obtained through measurement systems and additional parameters from laboratory analyzes or standard factors.

-Method for measuring emissions using direct measurement of CO<sub>2</sub> concentrations and flue gas flow.

- The experiments were performed in a CET unit on a boiler 420t / h steam. Five separate samples were taken of solid fuel-coal, slag and ash, and also 5 sets of direct measurements of  $CO_2$  were performed at the dispersion stack, for a period of 24 hours. • In order to use the calculation based method, analytical investigations were performed on samples of solid fuel- slag, coal, and ash. Test samples were subjected to laboratory analysis specific for solid fuels in order to calculate the oxidation factor, emission factor and evaluating the  $CO_2$  emission (Moisture, ash, carbon content, calorific value) in accordance with national and international standards.

• For using the measuring method of the concentration of  $CO_2$ , in the CET a portable flue gas analyzer-TESTO 350 M / XL was used, equipped with a special infrared (IR).cell.

Measurements were performed for the following parameters: temperature, pressure and analysis of O2 and  $CO_2$  Based on the data obtained the  $CO_2$  emission factor was calculated by applying methods based on both the calculation method and the direct measurement method.

• By comparing the results provided by the two methods (measurement and calculation) values obtained by direct measurement at the dispersion source / emission stack of CO2 (ECO<sub>2</sub>) were about 6% higher than those obtained by the calculation method.

• Research conducted showed that both methods can be used to monitor  $CO_2$  emissions, the results are comparable, the differences enrolling in the uncertainty of 10%, thus respecting the conditions in Annex VIII of Regulation 601/2012.

## REFERENCES

- [1] The Energy Strategy of Romania for 2007-2020
- [2] Annual Report on the state of the environment in Romanian-2010www.anpm.ro
- [3] International Energy Agency-IEA
- [4] Order 3420/2012 for the approval of the issuance of the permit for emissions of greenhouse gases in the period 2013-2020
- [5] Regulation 601/2012 on monitoring and reporting of greenhouse gas emissions pursuant to Directive 2003/87/EC of the European Parliament and of the Council
- [6] Report from the Commission to the European Parliament and of the Council, The European carbon market situation in 2012 - http://ec.europa.eu/

- [7] The European Environment Agency-www.eea.europa.eu;
- [8] Climate Change, UN Information Center-www.onuinfo.ro Romania;
- [9] Conference of the Romanian National Institute for development studies using energy sources, Reduction of CO2 emissions in the energy sector;
- [10] The analysis of greenhouse gas emissions -www.adrcentru.ro
- [11] ISO 10396:2008. Stationary source emissions. Sampling for the automated determination of gas concentrations. (O2, CO2, CO, SO2, NO, NO2, NO<sub>x</sub>)
- [12] SR EN 14181:2009 Stationary source emissions. Quality assurance of automated measuring systems
- [13] SR 5264:1995 Solid mineral fuels. Coal. Determination of moisture
- [14] ISO 1171:2010 Solid fuels. Determination of ash.
- [15] ISO 1918:2009 Solid mineral fuels. Determination of gross calorific value by the bomb calorimetric method and the calculation of lower calorific value.
- [16] ASTM D 5373:2008 Standard method for the analysis of carbon, hydrogen and nitrogen in a laboratory from samples of coal and coke.
- [17] ISO 351: 1996 Mineral fuels. Determination of total sulfur. High temperature combustion method.
- [18] STAS 398: 1992 Solid Fuels. Symbols for analytical characteristics and factors for recalculation of analysis results