ASPECTS REGARDING MONITORING OF AIR POLLUTANTS EMISSIONS

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ABSTRACT: *From environment protection's point of view, industry represents the most important field of anthropic activity. As a result of industrial operations, substantial quantities of carbon oxide, carbon dioxide, sulphur oxides and mostly SO_2, nitric oxides (NO/NO_2), cove hydrocarbons, volatile salts (chlorides, fluorides, sulphates), water vapors and so on are released in the atmosphere. Given the importance of emission and immision levels for environment's and general population's health, it is mandatory that measurements of their levels are made by laboratories experienced and authorized in this field of activity. Considering all the above, the study outlines both the imperatives of the effective legislation regarding measurements of gas, dusts and hydrocarbons (VOC), as well as the elements that must be taken into consideration when performing determinations for quantifying environment quality in industrial areas, using efficient equipments.*

***KEYWORDS*:** *emissions, monitoring, pollution, environment, fixed sources*.

1. LEGAL BASIS AND NATIONAL MEASUREMENT PROVISIONS, COMPARISON WITH EU.

At EU level, a series of regulations on air quality have been developed, namely:

Air Quality Framework Directive 2008/50/EC of the European Parliament and the Council of 21 May 2008, on ambient air quality and cleaner air for Europe

Its main objective is to assess ambient air quality in Member States, based on common methods and criteria, with the purpose of protecting human health and the environment as a whole, by regulation of measures to improve air quality or by maintaining it where it complies with the air quality objective.

Air Quality Framework Directive was followed by the so-called "daughter directives", which set numerical limit values and target values for each of the identified pollutants. The "daughter" directive objectives are to consolidate monitoring can be found in the Government Emergency Ordinance no. 152/2005 concerning integrated pollution prevention and control, additional amended by Law no. 84/2006, with its later amendments and additions;

2.Directive 2001/80/EC on the limitation of emissions of certain pollutants into the air from large combustion plants (LCP Directive) - transposed into national legislation by Government Decision H.G. 440/2010 regarding measures limiting strategies, methods of measurement, calibration and estimation methods to reach air quality measurements comparable with those in EU and provide definitive information to the public.

The directive was transposed and implemented through Law 104/2011 on ambient air quality.

At EU level, the legal framework for industrial emissions is ensured by ***Directive 2010/75/EU*** on industrial emissions (integrated pollution prevention and control). This European regulation is the result of reuniting seven different directives into only one, through the reformation procedure.

Nationally, the seven directives on industrial emissions, included in Directive 2010/75/EU, are translated into relevant legislation, as follows:

1.Directive 2008/1/EC concerning integrated pollution prevention and control (IPPC Directive), as amended by Directive 2009/31/EC on the geological storage of carbon dioxide. The provisions of this directive

the emissions of certain pollutants from large combustion plants;

3.Directive 1999/13/EC on **the limitation of emissions of volatile organic compounds due to the use of** organic solvents in certain activities and installations (VOC Directive), with its later amendments and additions - transposed into national legislation by Government Decision **H.G. 699/2003** regarding measures to reduce emissions of volatile organic compounds due to the use of organic solvents in certain activities and installations, with its later amendments;

4.Directive 2000/76/EC on waste incineration- transposed into national legislation by Government Decision H.G 128/2002 on waste incineration, with its later amendments and additions;

5.Directive 78/176/EEC on waste from the titanium dioxide industry, amended by Directives 82/883/EEC, 83/29/EEC and 91/692/EEC;

6.Directive 82/883/EEC on procedures for the surveillance and monitoring of environments concerned by waste from the titanium dioxide industry, amended by Regulation (EC) no. 807/2003;

7.Directive 92/112/EEC on procedures for the harmonization of programs for the reduction of pollution caused by waste from existing titanium dioxide industrial establishments;

The main issues induced by Directive 2010/75/EU consist in extending the scope of the Directive by including new activities in its first appendix, obligation to apply the conclusions on best available techniques, imposing more stringent emission limit values for some pollutants and specific rules for inspection and control activities within its scope.

Fusion, respectively, **substituting** the above mentioned **directives** into a singular complex European document represent the legislative answer of the European Union institutions regarding a *higher level of environmental protection* through adequate establishment of a general framework for the control of the main industrial activities, in the context of preventing, reducing or eliminating pollution resulted from these activities, through an integrated approach regarding the prevention and control of the emissions found in air, water and soil, the waste management, the energetic efficiency and accident preventing.

Directive 2010/75/UE on industrial emissions (integrated pollution prevention and control), entered into force on 07.01.2011 was transposed into national legislation through Law 278/24.10.2013 which recalls Law 84/2006, HG 128/2002, HG 699/2003 (with its later amendments and additions), HG 440/2010 and the Order of the Ministry of Environment and Waters Management secretary, State Ministry and Ministry of Economy and Trade no. 751/870/2004.

2. EMISSIONS MONITORING

Monitoring of emissions is a necessary tool to control and implement specific requirements of effective regulations. It is also absolutely necessary mean of information on the contribution brought by different pollution sources and allows prioritization regarding downsizing pollution.

Determination of noxae content of emitted substances and measurement methods differ depending on the physical condition they are in, respectively gaseous (gas), liquid (vapors of different substances) or solid (sediment or suspended particles) [1].

• Major gaseous pollutants are: sulphur oxides; nitric oxides; carbon monoxide and dioxide; ammonia; fluorides; chlorides etc.

• Liquid pollutants are present in the form of fine particles or vapors of hydrochloric acid, nitric acid, sulphur acid and so on;

• Suspended particles are solid pollutants of smaller dimensions, but whose specific weight allows them to float in the air;

• Sediment particles are micrometer-sized solid particles that are involved in ascending movements by the ascending currents of pollution source (air shaft) or those of the atmosphere, but whom in calm atmospheric conditions are deposited on the ground.

Periodic measurements of emissions from stationary sources are widely used, especially where there are no available automated measurement systems, for permanent installation, or when the automatic measuring systems are considered inadequate because of technical or cost reasons.

These uses of measuring emissions from stationary sources, conducted for regulatory purposes include [1]:

-Measurements to determine compliance with emission limit values;

-Field-testing of automated measuring systems for conformity assessment;

-Acceptance studies for new plans to reduce pollution;

-Determination of emission factors for use in emission certificates trading and reporting records.

In order to prevent and improve air quality and also in order to avoid adverse effects on human health and the environment as a whole, an important role is played by the compliance of activities and facilities to European provisions on atmosphere protection. To that effect, ANPM's Atmosphere Protection Service monitors the implementation of European Union legislation on atmosphere protection, thereby ensuring alignment with international legislations and European Union regulations in the field of environmental protection.

3. DIRECT (DISCONTINUOUS) MEASURING METHOD

Monitoring of air pollutants emissions is performed mainly in order to verify compliance with the concentration limit values set by effective legislations or environmental authorizations.

Discontinuous emission measurements are necessary for punctually establishing, in a certain limited time, the emission behavior of an installation [4].

Emission monitoring can be performed by the operator (self-monitoring) or by specialized accredited laboratories. Measurements must be carried out based on procedures complying with regulations and standards related to general requirements for measurement.

In order for the results to be able to be used for calculating emissions or assessing emission factors associated with a facility, they must meet the following requirements [4]:

- The use of appropriate methods and use measuring equipment;
- Provision of measurements representativeness;
- Provision of time coverage and data capture indicated by effective regulations and standards;

- Implementation and compliance with procedures for ensuring and controlling measurements quality control.

Typically, during test measurements, compound concentration in the exhaust gases is measured. serves to convert concentration to emission rate (weight rate).

Pollutant weight rate is calculated by multiplying the measured concentration of the pollutant by effluent volume flow.

There are three types of techniques for discontinuous emissions measurement (test) [4]:

■ In-situ analysis, used for periodic campaigns - The equipment used is portable, being transported to the source location. Samples are taken from the effluent using a probe and analyzed in-situ.

■ Laboratory analysis of samples taken using online fixed samplers - These samplers continuously extract samples and collect them in a special container. From the container is extracted quantity which is analyzed, obtaining the average concentration in the total collected volume.

■ Laboratory analysis of instantaneous samples taken in sampling points - The amount taken must be sufficient to provide a detectable amount of the pollutant. Results represent instantaneous values, valid for the time when the sample was taken.

Continuous or discontinuous measurements of pollutants concentrations and process parameters are performed using relevant standards of the European Committee for Standardization (CEN) and ISO standards, national / international regulations meant to ensure the provision of data of an equivalent scientific quality.

Emissions monitoring / performing determinations involves the following steps: flow measurement, sampling, storage, samples transport and preservation, sample treatment, sample analysis, processing data, reporting data.

Total emissions from a facility or unit are given not only by normal emissions from shafts and pipes; diffused and fugitive emissions should also be considered. It is acknowledged that these emissions can cause potential health and environmental damage and that sometimes these flaws may have economic significance for a facility.

There are many reasons for performing discontinuous emission measurements. In addition to measurements required by relevant authorities, there also are measurements requested by station operators, such as measurements for self-monitoring or for optimizing the facility.

Discontinuous emission measurements are carried out:

a) In order to perform a test (proof of guarantee);

b) In order to certify compliance with the emission limit;

c) For monitoring, after the expiration of a predetermined time period, to check the status of the installation;

d) In case of complaints;

e) In order to initiate an approval procedure;

f) During own internal supervision;

Simultaneously, there are other measurements that have to be taken such as exhaust gases volume flow, which

g) For declaring emissions;

h) In the event of operating problems;

i) In case of check-outs related to technical safety.

4. MEASUREMENTS PLANNING

According to SR CEN / TS 15675:2009, a plan has to be developed before making measurements. It will contain all the technical data that must be known prior to measurement and will be completed by the person responsible for testing, along with the economic agent (facility operator) [5].

Generally, the duration of a discontinuous measurement should not exceed half an hour. By analogy with this, the measurement results must be usually sent as an average of half an hour.

5. PERFORMING MEASUREMENTS

Selecting the distance and surface of measurement

In order to perform a measurement of the emission values and obtain quality results, it is very important to carefully choose the measuring distance and surface. The place for performing trial measurement should be selected so that it reflects a representative measurement, in order to assess the emission behavior of the installation [6].

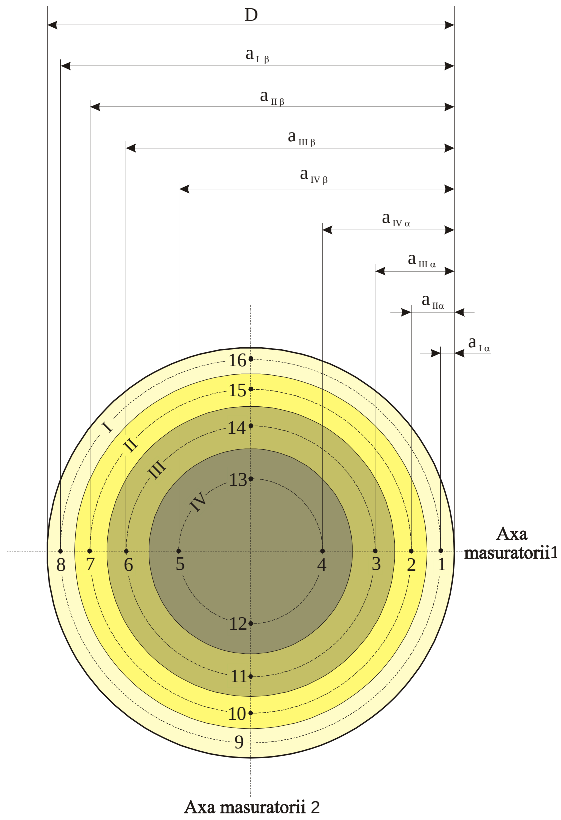
In choosing the measurement surface it is preferable to select the areas behind the suction fan, because there it is more likely to find a homogeneous mixture of residual gases that in front of the fan. Taking samples for the measurement of substances in particulate form, from horizontal residual gases ducts, should be carried out along a vertical measurement axis, as there is the possibility of sedimentation phenomena.

The measurements of the measuring range should be sufficient for the assignment (see Figure 1.1.) as follows:

- For accessing the measuring points it’s necessary to prepare a fixed or mobile platform, ensuring a sufficient working space, required electricity connections, attending to given specific measures of occupational safety;
- Measurements of the network have to be made, and then there must remain enough crossing space, in order to push the probes. One must keep in mind that the grid protection or guard rails should not be an obstacle in the way of the probe’s movement.

Working height from field measurement to measurement axes should be about 1.2 to 1.5 m.

Introduction of probes in measuring openings [4] must take place in conditions of maximum security and should not be prevented by grids or protective guard rails.



Sectiune transversala rotunda cu doua axe ale masuratorii
si cate opt puncte de masurare pe fiecare axa

Fig. 1.1 Model of measurement field from a perpendicular residual gas shaft, with 2 measuring axes and 4 measuring openings for performing crossing measurements

Access holes in the shaft and measuring places have to be dimensioned in such a manner that they allow good sampling.

5.2 Network measurements

In order to perform a network measurement, the measurement cross section is divided into several equal areas. Figure no 1.2 highlights the cross section of a rectangular duct, a round duct and dividing into equal areas. Rectangular cross-sections are divided in similar surfaces and round sections are divided into circular rings. The measuring points are found in the most important points of the divided surfaces.

In the case of circular cross-section, distances between measuring points and duct walls are calculated using equation (1), in close liaison with the number "i" of subdivided surfaces and ordinal number "n".

(1)

$$a_n = \frac{D}{2}\left(1 \pm \sqrt{\frac{2i-2n+1}{2i}}\right) = DK_n$$

i= number of subdivisions
n= ordinal number
D= chimney diameter

5.3. Isokinetic extractive sampling

Extractive sampling to collect particles, substances associated to particles or aerosols must be isokinetic sampling.

As isokinetic sampling corresponding to a previously established flow profile is concerned, at each measuring point the suction flow speed must be adjusted to the flow rate measured previously. Suction duration must be the same for each measuring point. A calculation of the density of different mass concentrations at different measuring points is automatically performed through the absolute sucked volume of the item being measured [7].

Automatic manual dusts sampling systems continuously measure flow rate or probe's pressure ratios and automatically adjusts the rate of suction.

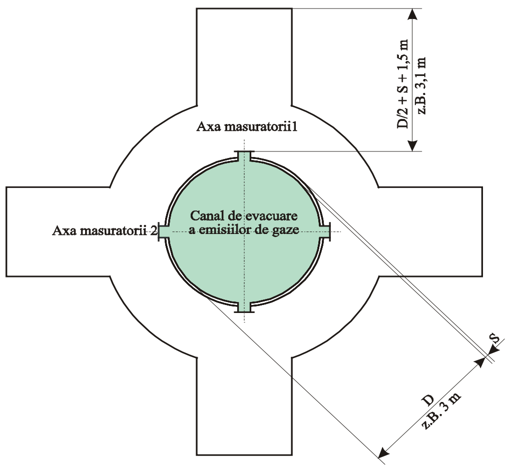


Fig. 1.2 Positioning of measuring points in cross, rectangular and circular sections of a culvert

5.4 Extractive sampling for gas measurements

Extractive sampling for gas measurement can be performed either in the form of network measurements (integrating the cross section), either punctual. Sampling from a measuring point in of the measurement range (punctual sampling) implies that the chosen measuring point has to be representative of the entire measurement cross section, regarding the mass density of the object's flow. This representativeness must be provable [5].

standard temperature and pressure. The measurement results refer to a time period of evaluation. Usually this time for evaluation corresponds the half an hour sampling period [6].

measurement assessment and measurement results interpretation (sampling site description, measurement methods, equipment used, stage of installation functioning during measurement, and so on).

6. MONITORING EQUIPMENT

Environmental Laboratory of INCD INSEMEX performs dust and exhaust gas emissions determinations (under RENAR accreditation) and measurements of volatile organic compounds emissions. The sampling equipment used to determine emissions of dust, gas and VOC is of the latest generation, with heated sampling lines to avoid condensation. All procedures regarding gas, dusts and volatile organic compounds emissions observe the effective standards, namely ISO / IEC 17025: 2005, SR CEN / TS 15675:2009, ISO 10396:2008, ISO 9096/2005, EN 12619: 2013 EN 15446: 2009.

Exhaust gases portable analyzer

Fig. 1.3 TESTO 350 XL multigas analyzer

The equipment is designed for determinations of nitrogen dioxide, nitric oxide, sulphur dioxide, carbon dioxide, carbon monoxide and oxygen concentrations in the emissions of stationary sources.

Multigas analyzer TESTO 350 XL (Fig. 1.3) is made of the sampling probe, analysis unit and control unit. For gaseous effluent sampling, a stainless steel metal probe is used, provided with an internal thermocouple to determine the effluent's temperature.

The method consists in determining the concentration of gas samples taken from stationary

In order to more clearly describe a gas flow, one must follow the following parameters of residual gases: density, moisture content, temperature, flow rate and static pressure.

5.5 Evaluation / Making the report

In order to make an assessment, the measurement values are generally referred to a dry-gas volume at

The measurement results are forwarded in the form of a test report. Besides the measurement results, the report also contain (according to EN 15259:2009) other adjacent information, important for the emissions sources emissions (industrial chimneys, pipes) using TESTO 350XL equipment.

Samples are taken with the help of an internal pump unit and gas is cooled on the suction route, water vapors being condensed with the direct result that NO2 and SO_2 have the lowest humidity absorption. Condense is pumped to the desiccator installed in the equipment's body [7].

In case of continuous technological flow, the duration of a measurement is 30 minutes. If the technological flow is discontinuous then the time of sampling / measurement is considered to be the period with relevant records of gaseous effluent concentration.

BASIC ISOSTACK system for determinations of dusts from emissions mass concentrations

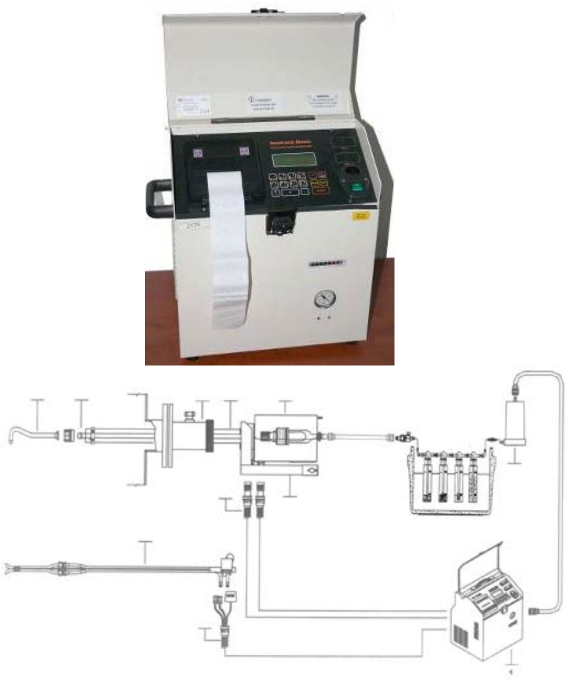


Fig. 1.4 BASIC ISOSTACK systems for dust and gas isokinetic sampling

This equipment is designed for measurements of dust and gases from stationary sources (chimneys). The ISOSTACK BASIC (fig.1.4) includes:

- ISOSTACK BASIC hot probe sampler;
- Quick gas entry connector;
- Raufilam suction tube, suction nozzle;
- Gas entry filter cartridge;

- Silica gel Box to absorb moisture;
- Power cable, sampling filters.

The device can be configured in two sampling lines:

- Isokinetic particulate sampling line;
- Isokinetic gas line.

The method consists in sucking through the filter a known volume of air with the help of a isokinetic probe and weighing the dust mass deposited on it.

Choosing the place where the measurement will be performed is made so that the results are representative for the emission behavior of the installation. Sampling site must meet the following criteria [7]:

Allow access to the sampling pipe / bin;

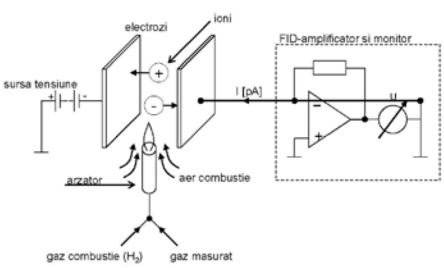
Determination of the total gaseous organic carbon mass concentration is performed by SmartFID analyzer (Fig. 1.5), using flame ionization detector, according to SR EN 12619:2013, which is intended for use as a reference standard method for measuring the mass concentration of gaseous or vapor organic substances (expressed as TVOC) in emissions from stationary sources.

Fig. 1.5 SmartFID hydrocarbons analyzer

Measuring technique used by the flame ionization detector (FID) is the ionization of organically bound carbon atoms in a hydrogen flame [7].

Flame ionization detectors (FID) are acknowledged as safe and robust analyzer to measure volatile organic compounds (VOCs) whose principle of operation (Fig. 1.6) consists in chemical ionization of organic substances in a hydrogen flame.

7. CONCLUSIONS



Availability of supplying the equipment with electricity;

There are adequate access holes for measurement and work platforms.

Gas flow profile is ordered and stable, without turbulence or gas return.

Extractive sampling for determination of pollutants concentrations can be carried out either as a network measurement (integrating the cross section) either punctual.

Sampling time for one sample is at least 30 minutes.

Analyzer for volatile organic compounds (VOC) determination

➢ Directive 2010/75/EU on industrial emissions (integrated pollution prevention and control) was transposed into national legislation by Law 278/24.10.2013;

➢ Compliance of activities and facilities to European provisions on atmosphere protection play an important role in prevention and improvement of air quality in order to avoid adverse effects on human health and the environment as a whole;

➢ Periodic measurements of emissions from stationary sources are widely used, especially where there are no available automated measurement systems, for permanent installation, or when the automatic measuring systems are considered inadequate because of technical or cost reasons;

➢ Monitoring of air pollutants emissions is performed mainly in order to verify compliance with the concentration limit values set by effective legislations or environmental authorizations.

➢ Environmental Laboratory of INCD INSEMEX performs dust and exhaust gas emissions determinations (under RENAR accreditation) and measurements of volatile organic compounds emissions.

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