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# <Literature review and practical experiences on continuous dust monitoring at low concentrations>

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# Contents

# 1. List of abbreviations

| | |
|---|---|
| AELs | Associated Emission Levels |
| AMS | Automated Measuring System |
| AST | Annual Surveillance Test |
| BAT | Best Available Techniques |
| BREF | Best Available Techniques Reference Document |
| CMR | Certified Measuring Range |
| IED | Industrial Emission Directive, EU Directive 2010/75/EU |
| ELV | Emission Limit Value |
| LCP | Large Combustion Plant |
| LCPD | Large Combustion Plant Directive |
| PEMS | Predictive Emissions monitoring system |
| PM | Particle Matter. Can be a synonym of "dust" or "TSP", but generally refers to a defined class diameter fraction of the TSP |
| QA | Quality assurance |
| QAL | Quality Assurance Level |
| SRM | Standard Reference Method |
| TSP | Total Suspended Particles, referred as "dust" in the EU standards |
| TÜV | Technischer Überwachungsverein (Association for Technical Inspection) |

# 2. Executive summary

This report reviews the techniques available for the measurement of dust from gas-firing combustion in refineries and identifies challenges associated with the continuous monitoring of these dust emissions at low concentrations.

A short background section is provided to show the contribution from gas-fired combustion sources to overall dust emissions in refineries. Information on the current emission limits and monitoring requirements for combustion units are also presented.

The findings from the literature review and practical experiences show that continuous dust monitoring and measurement at low concentrations are challenging. Furthermore, when dust concentrations are very low, it may not be possible to comply with the current standard requirement regarding the quality of the data (i.e. uncertainty requirement not possible to be met). When the measurements are not possible within a relevant uncertainty, the added value of a continuous monitoring for quantification purposes is questionable. In this case, a monitoring system will only act as a qualitative detection. When the process is expected to be stable in terms of combustion conditions and fuel, discontinuous measurements could be considered as a valid alternative for the quantification of the dust emissions.

# 3. Background

## 3.1. Dust emissions from the European refinery sector

According to the data in the Convention of Long Range Transboundary Air Pollution of UNECE database [1], the contribution of the oil refining sector to the total EU $PM_{10}$ emissions to air in 2013 was 7.6 kt. This represents only 0.4% of the total $PM_{10}$ emitted in EU (natural sources not included). Within refineries, the main emission sources of dust are catalytic cracker regenerators, process furnaces/boilers (mainly those fired with (liquid) heavy fuel oil), coke plants, incinerators, decoking and soot blowing of furnaces and flares [2].

Dust emissions from combustion sources are highly dependent on the type of fuel used. For example, burning fuels such as refinery fuel gas or natural gas lead to relatively low dust emissions while firing fuels such as refinery fuel pitch or refinery fuel oil produce significantly higher emissions.

The Refinery BREF [2] provides information on the particulate emission ranges for combustion installations. Table 1 below shows the table with the ranges of particulate concentrations that have been found in European refineries. Broadly, during gaseous fuel firing, emission concentrations have been < 5 mg/Nm$^3$.

**Table 1: Particulate emissions ranges from combustion units. Reference Refinery BREF [2].**

**Table 3.60: Particulate emissions ranges for existing installations**

| | Refinery fuel gas | Liquid refinery fuel |
|---|---|---|
| Process furnaces | 0.4 – 2.4 | 5 – 1 000 |
| Boilers | <5 | 5 – 500 |
| CHP | - | 2 – 3(1) |

NB: All figures in mg/Nm$^3$ at 3 % $O_2$ (monthly average).
(1) The flue-gas is treated in a $SNO_X$ plant. RFG and natural gas are co-fired with heavy fuel oil.
*Sources*: [ 151, Sema, Sofres 1991 ], [ 191, UBA Austria 1998 ] updated TWG NL 2010

Additionally, the database of large combustion plants [3] provides information on the mass emission of dust from plants above 50 MWth in refineries. According to the most recent data available in this database (2012), 37% of the Large Combustion Plant (LCP) units in refineries (Gas turbines excluded) were gas-fired, corresponding to 25% of the thermal capacity installed. The dust emissions from these units were only 5% of the total large combustion dust emissions in refineries (Figure 1).

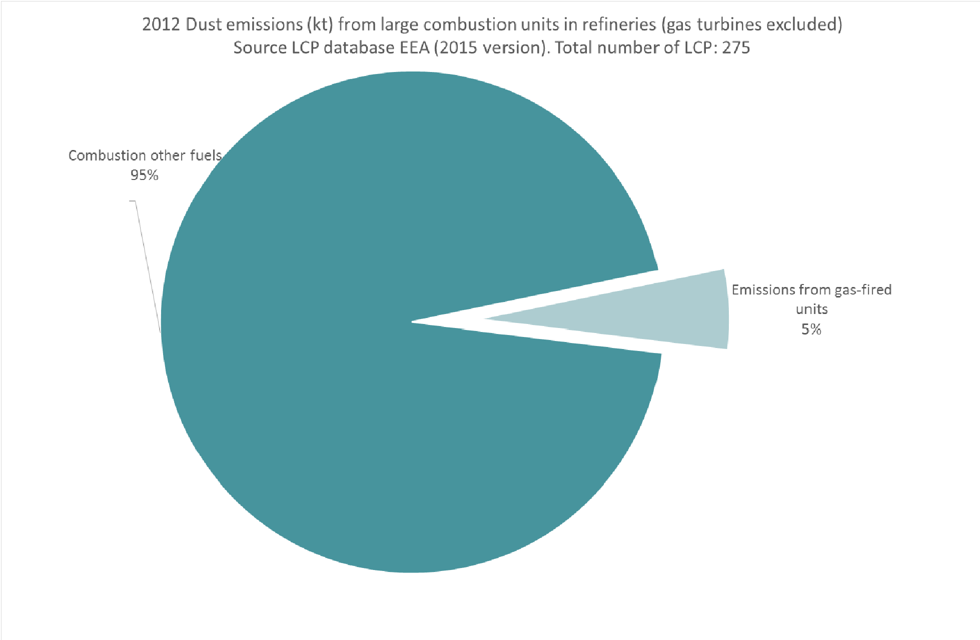



**Figure 1: Dust emissions from LCP in refineries. 2012 data [3].**

## 3.2. European Regulatory Framework

### 3.2.1. Best Available Technique (BAT) Conclusions

Refinery permits have to be reconsidered according to the European BAT conclusions for the refining of mineral oil and gas [4] which includes Best Available Techniques Associated Emission Levels (BAT-AELs) for some refining processes. For the dust emissions from combustion sources, the only process where BAT-AELs are provided is for multi-fuel fired combustion (BAT-35). See Table 2 below [4]. No dust BAT-AELs were adopted for gas-fired combustion units only.

**Table 2. BAT-AEL for dust emissions from multi-fuel fired units [4].**

*Table 12*

**BAT-associated emission levels for dust emissions to air from a multi-fuel fired combustion unit with the exception of gas turbines**

| Parameter | Type of combustion | BAT-AEL (monthly average) mg/Nm³ |
|---|---|---|
| Dust | Multi-fuel firing | 5-50 for existing unit (¹) (²) |
| | | 5-25 for new unit < 50 MW |

(¹) The lower end of the range is achievable for units with the use of end-of-pipe techniques.
(²) The upper end of the range refers to the use of a high percentage of oil burning and where only primary techniques are applicable.

The associated monitoring is in BAT 4.

The European BAT conclusions also require the monitoring of dust emissions from combustion units. For units equal or above 100 MWth, continuous monitoring by direct measurement is required. For units sized between 50-100 MWth, continuous monitoring is required using direct measurement or indirect monitoring.

*Note the definition of indirect monitoring as per BAT Conclusions: "Estimation of the emissions concentration in the flue-gas of a pollutant obtained through an appropriate combination of measurements of surrogate parameters (such as $O_2$ content, sulphur or nitrogen content in the feed/fuel), calculations and periodic stack measurements. The use of emission ratios based on S content in the fuel is one example of indirect monitoring. Another example of indirect monitoring is the use of PEMS".* [3]

It is worth noting that in the US, combustion units (boilers and heaters) firing refinery fuel gas are not subject to continuous emission monitoring for dust by US-EPA regulation.

### 3.2.2. Annex V Industrial Emission Directive (IED): Large Combustion Plants (LCP)

The Industrial Emissions Directive [5] Annex V sets a limit value of 5 mg/Nm$^3$ for combustion plants above 50 MWth using gaseous fuels.

*Note that the emission limit value of 5mg/Nm$^3$ applies to gaseous fuels in general. Blast furnace gas and gases produced by the steel industry which can be used elsewhere have different emission limits.*

It also requires continuous dust emission monitoring for units with a capacity above 100 MW; however, a competent authority may decide not to require continuous monitoring in the case of combustion plants firing natural gas.

Given the Emission Limit Values (ELVs) set by the IED and the data available in the Refinery BREF [2] (Figure 1 above) and the LCP database [3], it is reasonable to suggest that the dust emissions from most of the gas-fired units in refineries, are below 5 mg/Nm$^3$.

The following sections of this report review the techniques available for the measurement of dust from gas-firing combustion in refineries and identifies challenges associated with the continuous monitoring at these low concentrations.

# 4. European standards related to dust monitoring and measurements

In order to support the environmental legislations and more specifically the EU Industrial Emission Directive 2010/75/EU (IED), different European standards are published addressing different aspects of the dust monitoring and measurement for environmental purposes:

- the requirements regarding the measuring sections in the stacks;
- requirements for the manual method measurements performed by accredited laboratories for (i) the dust concentration determination and (ii) the calibration of the dust Automated Measuring System (AMS);
- technical characteristics of the dust AMS monitoring instruments themselves and quality requirements for the AMS manufacturers;
- the different Quality Assurance Levels (QALs) to perform during the AMS lifetime in order to confirm that the AMS measuring characteristics stay the same;
- the environmental data handling.

The most relevant European standards as well as the required QALs are listed below:

- **EN 15267-1, 2 & 3 Air quality - Certification of automated measuring systems**. It includes three published parts, and one part in preparation:
    - **Part 1: "General principles", 2009**, which states the roles and responsibilities of the involved stakeholders (AMS manufacturer, test laboratory, certification body).
    - **Part 2: "Initial assessment of the AMS manufacturer's quality management system and post certification surveillance for the manufacturing process", 2009**, which sets out the responsibilities of the AMS manufacturers.
    - **Part 3: "Performance specifications and test procedures for automated measuring systems for monitoring emissions from stationary sources", 2007,** which describes the tests (in laboratory and on site) that have to be carried out by an official testing laboratory (like TÜV in Germany or MCERT in the UK,) in order to deliver the QAL1 certificate. During the certification process, all the contributors to the measurement uncertainty are evaluated (such as the impact of power supply instability, interferences, etc.) and compared with the maximum allowed value. Results are summarized in the QAL1 certificate and allow for checking whether the AMS is suitable for the intended monitoring purpose.
    - **Part 4**: in preparation and will cover the same information as part 3 but for the instruments used as Standard Reference Method (SRM).

- **EN 15259- Air Quality - Measurement of stationary source emissions - Requirements for measurement sections and sites and for the measurement objective, plan and report.** This standard describes the location where the AMS has to be installed in order to provide results representative of the emissions, as well as the tests to perform in order to demonstrate the representativeness. The characteristics of the measuring section is of particular importance for dust measurements and monitoring.

- **EN 14181 Stationary source emissions - Quality assurance of automated measuring systems** (2014) in which the quality assurance (QA) procedures to perform after the AMS has been installed on site are described. The main objective of this standard is to demonstrate that the pollutant monitoring is performed with a measuring uncertainty not higher than the maximum uncertainty laid down in the IED, i.e. that at the emission limit value level, the values of the 95 % confidence intervals of a single measured result shall not exceed 30% of the ELV for dust. There are three QA levels:

  - QAL2: determines the calibration function by means of parallel measurements performed by an accredited laboratory using SRMs (Standard Reference Methods), and check that, by applying the calibration function, the monitoring results are within an uncertainty lower than the maximum uncertainty required by the directives.

  - QAL3: check on a regulatory basis of the AMS drift at zero and at a span point, keeping the drifts under control and under maximum values.

  - AST (Annual Surveillance Test): yearly check by an accredited laboratory of the calibration function and the measurement uncertainty.

- **EN 13284-1,2: Determination of low range mass concentration of dust**

  - **Part 1: Manual gravimetric method** (2002)

  - **Part 2: Automated measuring systems** (2004). This part amends the EN 14181 and provides guidance specific to dust measurements. It is only applicable in conjunction with EN 14181.

  The Technical Committee CEN/TC 264 "Air quality", working group WG5 is currently revising both parts of the EN 13284.

- **EN ISO 16911-1,2: Stationary source emissions — Manual and automatic determination of velocity and volumetric flow in ducts**

  - **Part 1: Manual reference method** (2012)

  - **Part 2: Automated measuring Systems** (2012)

- **DAHS: a new standard on Data Acquisition and Handling System** is currently being drafted by the Technical Committee CEN/TC 264 working group WG9. It will consist of three parts, like the EN 15267, and will state the minimum requirements for the environmental data management.

# 5. Measurement methods

## 5.1. Current requirements for dust monitoring

When a monitoring system is required for dust, as for other pollutants, the monitoring instrument (AMS) has to have a type approval (QAL1 certificate) delivered by an official testing laboratory (such as TÜV in Germany or MCERT in the UK) prior to its installation on site. After its installation on site, its suitability for the intended monitoring purpose has to be demonstrated according to the QAL2 procedure. During the QAL2, a calibration function is calculated from the comparison of AMS and SRM results of at least 15 pairs of values. SRM results are from parallel measurements performed by an accredited laboratory. The QAL2 has to be performed at least every 5 years. Between two QAL2, the validity of the calibration function has to be confirmed on a yearly basis in accordance with the AST procure by comparing five pairs of SRM and calibrated AMS results. The accredited laboratories are ISO17025 compliant and are acknowledged by the national or the local authorities, following processes depending on the country. During the QAL2, AMS calibration factors will be determined to correct the values provided by the AMS based on the SRM values. For this process, the SRM results from the accredited laboratory are supposed be real values. This is of course not the case, and uncertainties are also attached to SRM values. If the SRM values are of poor quality, so will be the calibration function as well as the calibrated AMS results.

Therefore, both reliable AMS monitoring devices and reliable SRM manual methods are needed to get a compliant monitoring system.

## 5.2. Units for dust emissions into the air vs. outputs of the AMS

In Europe, ELVs are expressed in mg dust/Nm³, dry basis at a reference $O_2$ concentration. The values from dust AMS for checking the compliance with the ELV have to be reported in the same units as the ELV. In the USA for example, the opacity is sometimes reported, instead of, or in addition to, mg/Nm³.

Most of the dust AMS are based on measuring principles that do not provide a direct estimation of the dust concentration, such as light scattering-based AMS. These AMS outputs only give an indication of the dust concentration. The exact correlation between AMS outputs and mg dust/m³ has to be established for each monitoring system, and also for each operating condition, since most monitoring devices are sensitive to the dust characteristics (colour, shape, size distribution) and flow rate variations. Nowadays, this correlation is included in the calibration function of the AMS determined during the QAL2 procedure.

## 5.3. Standard Reference Method

### 5.3.1. Description of the manual gravimetric method

Within the IED, dust AMS have to be calibrated, as with other AMS, at least every 5 years according to the procedure QAL2 from the EN 14181. Therefore, an accredited laboratory has to come on site to perform at least 15 dust measurements by using the SRM spread out on at least three days. Note that the standard recommends to perform 18 measurements in order to have some spare results. The dust AMS calibration function is derived from the comparison of AMS and SRM results and the uncertainty of the final results from the AMS will be checked against the maximum allowed uncertainty (i.e. 30% of the ELV). Note that normally the calibration function is linear, y=ax+b, but for dust AMS, quadratic calibration functions are allowed (cf. EN 13284-2).

The European SRM for dust measurement is described in the EN 13284-1:2001- Stationary source emissions - Determination of low range mass concentration of dust - Part 1: Manual gravimetric method. This standard addresses dust concentration lower than 50 mg/m³ (but is also suitable for higher concentrations).The standard reference method described in the standard is the gravimetric method.

The general principle of gravimetric dust measurements is the pumping of a known volume of flue gas through a filter. The sampling has to be isokinetic i.e. the velocity of the flue gases entering the sampling probe has to be the same as the velocity of the flue gases in the flue gas channel at the sampling point. The complete flue gas channel section has to be sampled which means that appropriate measuring ports and working platforms have to be available. By weighing the filter before and after the sampling, the total mg dust contained in one m³ of flue gas can be calculated.

Even if the gravimetric method looks to be quite simple, several factors can strongly influence the final result. The competencies and know-how of the accredited laboratory operators performing the SRM measurements have to be carefully checked.

Long enough sampling time (even longer than prescribed in the standard) have to be anticipated in order to reduce the measurement uncertainties. The filter can be inside the stack for dry flue gases as illustrated in Figure 2, or outside the stack for dry and for saturated flue gases as illustrated in Figure 3.

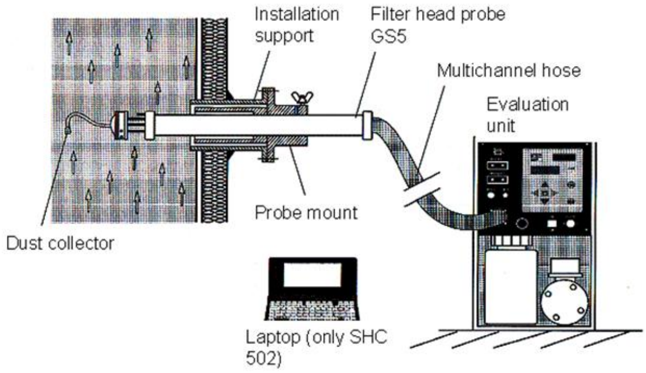



**Figure 2: Example of manual dust sampling with a GRAVIMAT (SICK company). The filter is located in the head of the sampling probe inside the stack.**

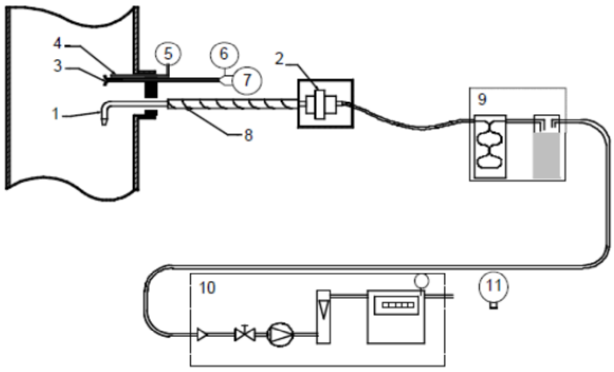


**Figure 3: Example of a manual dust sampling as illustrated in the EN 13284-1. The sampling line is heated and the filter is located outside the stack (2 on the scheme).**

### 5.3.2. Uncertainties of the standard reference method for dust measurements

Every accredited laboratory performing dust measurements must know the uncertainty associated with the measurements and must be able to reduce this uncertainty as much as possible. Generally, the uncertainties calculated by the accredited laboratory are optimistic. Several operators have complained about the poor quality of the measurements performed by accredited laboratories. In some cases, the provided results did not make sense and/or the measurement details were not communicated. Furthermore, measurement uncertainties were significantly higher than the laboratory calculations due to uncontrolled external factors which can increase the uncertainty. Dust measurements in the last year have shown that, with low dust concentrations, getting the dust monitors calibrated and validated in accordance with the EN 14181 is really challenging.

Some uncertainties are related to the technologies used by the accredited laboratory, such as the uncertainty associated with the sampled volume or with weighing of the filters.

Another source of uncertainty can be that the laboratory cannot sample across the complete section. This can occur when the stack diameter is too big and the laboratory does not use a sampling probe of the appropriate length. Sometimes it is impossible to manipulate the sampling probe due to the proximity of other equipment or due to the available space on working platforms. Swirling flue gas flow at the sampling plane and/or unstable process conditions during the sampling will further increase the uncertainty on the dust result.

| |
|---|
| In order to reduce the uncertainty of the dust measurement related to dust sampling, the requirements on the measurement sections and measurement ports described in the EN 15259 should always be respected. |

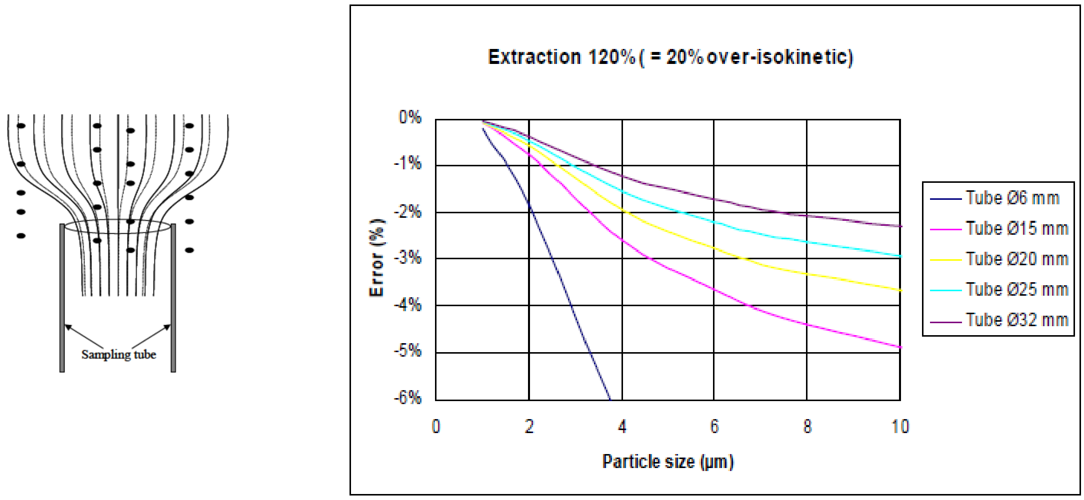
The EN 15259 recommends that the measurement plane is situated in a section of the flue gas where homogenous flow conditions and concentrations can be expected. This can mostly be achieved (1) when there is no disturbance of the flow stream, (2) in a section of duct with constant shape and cross-sectional area, and (3) in a section of the duct with at least five hydraulic diameters of straight duct upstream of the sampling plane and at least five hydraulic diameters from the top of a stack downstream (two hydraulic diameters when the flow stays in the flue gas channel and is not emitted into the atmosphere). The installation of measurement sections in vertical ducts should be preferred to installation in horizontal ducts. The standard EN 13284-1 specifies that, at all the sampling points, the gas stream should meet the following requirements:

- angle of gas flow less than 15° with regard to duct axis;
- no local negative flow;
- minimum velocity depending on the flow rate measuring method used (for Pitot tubes a differential pressure larger than 5 Pa);
- ratio of the highest to lowest local gas velocities less than 3:1.

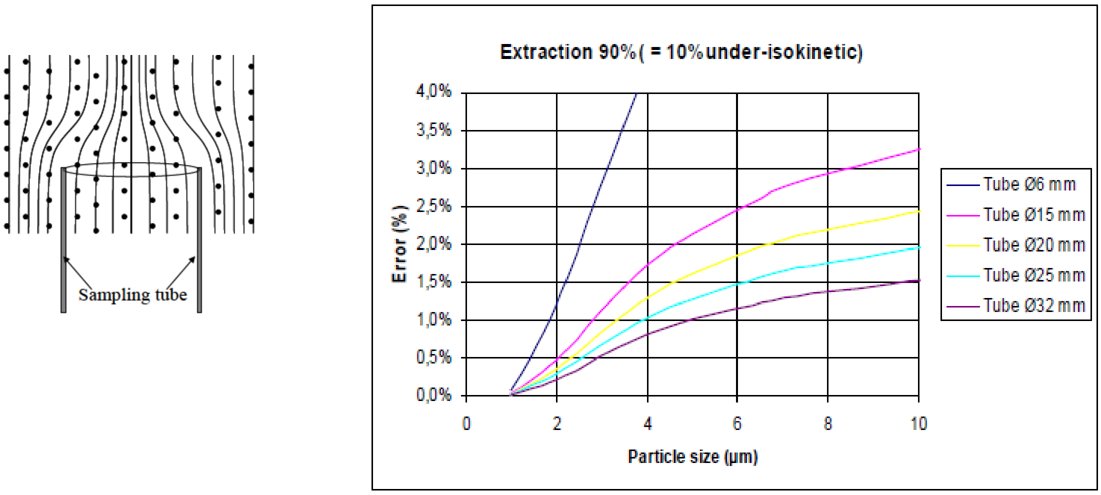
In order to increase the reliability of a manual dust measurement, the flue gas channel section where the samples are taken has to be as homogeneous as possible as stated above, while also ensuring that process conditions are kept as stable as possible.

| |
|---|
| During dust measurements (particularly when the measurements are used to perform a QAL2 or an AST) the process conditions should be kept stable when possible during each sampling. |

The standard EN 13284-1 also mentions that the sampling should be isokinetic. Sampling velocity should be within -5% to + 15% of the isokinetic velocity. The impact of non-isokinetic sampling is illustrated in Figure 4 below which shows the effect on particle trajectory with different size diameters if the sampled flue gases do not enter the sampling probe with the same velocity as in the flue gas channel.




a. Illustration of over-isokinetic sampling and its effects on final result




b. Illustration of under-isokinetic sampling and its effects on final result

**Figure 4 : Impact of over and under- isokinetic sampling on the final results of total dust quantification (Martin R Angelo, 2008).**

## 5.4. Automated Measuring Systems

Several measuring principles are available for dust monitoring at stacks. However only few of them are mature and /or suitable for low dust concentration monitoring.

The focus in this report will be on light scattering and beta-attenuation because the other measuring principles have been proven to be unsuitable for low dust concentrations and/or for dust quantification.

### 5.4.1. Uncertainty requirements for Automated Measuring Systems

All AMS have to be tested prior to their installation on site in order to prove that they are suitable for the intended monitoring purposes. The tests are performed by an accredited laboratory in accordance with the EN 15267, part 3, and the tests results are summarized in a QAL1 certificate. Individual contributors to the total measuring uncertainty are identified and quantified. Each of these has a maximum allowed value as presented in Table 3 and Table 4 below. The maximum allowable deviations (as absolute values) of the measured signals are given as percentages of the upper limit of the certification range.

**Table 3 : Performance criteria for dust monitoring AMS in laboratory tests (EN 15267-3).**

| Performance characteristic | Performance criteria |
|---|---|
| Response time | ≤ 200 s |
| Repeatability standard deviation at zero | ≤ 2,0 % [a] |
| Repeatability standard deviation at zero | ≤ 5,0 % [b] |
| Lack of fit | ≤ 3,0 % [a] |
| Zero shift due to ambient temperature change from 20 °C within specified range | ≤ 5,0 % [a] |
| Span shift due to ambient temperature change from 20 °C within specified range | ≤ 5,0% [a] |
| Influence of voltage at −15 % and at +10 % from nominal supply voltage | ≤ 2,0 % [a] |

[a] Percentage value as percentage of the upper limit of the certification range.

[b] Percentage value as percentage of the emission limit value.

*NOTE: The response time does not apply to batch-measurement techniques such as beta-ray-attenuation.*

**Table 4 : Performance criteria for dust monitoring AMS in field tests (EN 15267-3).**

| Performance characteristic | Performance criteria |
|---|---|
| Determination coefficient of calibration function, $R^2$ | ≥ 0,90 |
| Response time | ≤ 200 s |
| Lack of fit | ≤ 3,0 % |
| Minimum maintenance interval | 8 days |
| Zero drift within maintenance interval | ≤ 3,0 % |
| Span drift within maintenance interval | ≤ 3,0 % |
| Availability | ≥ 95,0 % |
| Reproducibility, $R_{field}$ | |
| — for concentrations > 20 mg/m$^3$ | ≤ 2,0 % |
| — for concentrations ≤ 20 mg/m$^3$ | ≤ 3,3 % |

*Note: "Lack of fit" refers to the linearity.*

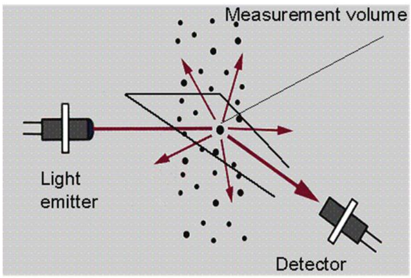
The Certified Measuring Range (CMR) can be no higher than 2.5 times the daily ELV for large combustion plants (1.5 times the daily ELV for waste incinerators). The global uncertainty, expressed as a % of the CMR, must be less than 75% of the maximum uncertainty allowed by the IED. The remaining 25% is left for other uncertainty sources such as the variation of the uncertainties between two AMS of the same type, the representativeness of the sampling point, the peripheral measurements like T°, $O_2$, etc. For dust, the maximum uncertainty allowed by the IED is 30% of the daily ELV. Consequently, the global uncertainty, calculated on the basis of the QAL1 certificate for dust AMS is 22.5% of the ELV given in mg/Nm³.

### 5.4.2. Optical automated measuring systems based on light scattering

#### 5.4.2.1. General principle

The most commonly used technologies for dust monitoring are based on light extinction or on more recently developed light scattering. A ray of light is sent through the flue gas. The light extinction or the light scattered caused by the dust is measured and allows for the evaluation of the dust content in the flue gas. Light extinction AMS have been developed for high dust concentrations (or for cases where opacity has to be reported like in the US). However, despite their (relative) low sensitivity, some manufacturers claim that with large stack diameters they might be also suitable for low dust concentrations. Laborelec, based on its experience, does not support this point. Therefore the focus of this report will be on light scattering instruments that were especially designed to measure low dust concentrations.

The dust has the property of scattering the light in all directions as illustrated in the Figure 5 below. Depending on the ratio particle size/ incident wavelength, the scattering can be described by three different laws presented in Figure 5 and illustrated in Figure 6.




Depending on the ratio particle size / incident wavelength, r/λ , the scattering can be described by three different laws:

- **Geometric optics** when **r/λ > 1**,
- **Rayleigh scattering** when **r/λ < 1**
- **Mie theory** when **r/λ ≈ 1**

**Figure 5 : Illustration of the light scattering by a particle in all the directions.**

A light scattering type instrument measures the amount of light scattered in a particular direction. Light scattering AMS can be "back", "side" or "forward" scattering depending on the position of the receptor toward the path of the light source. The intensity of the scattered light depends on the angle of observation, the size of the particle, its refractive index and shape, and the wavelength of the incident light.

Arguments have been made that the forward-scattering technique is the less sensitive to particle size changes or refractive index changes although this has not been well documented in source monitoring applications.

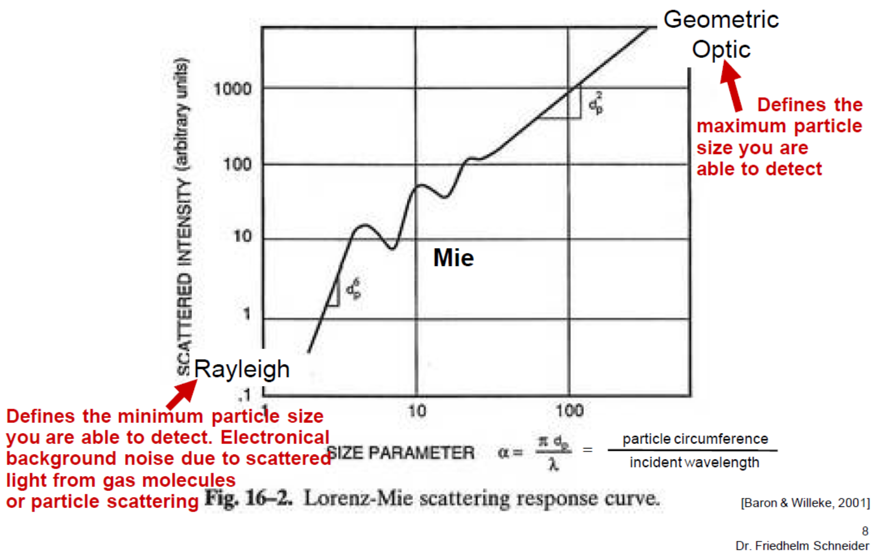
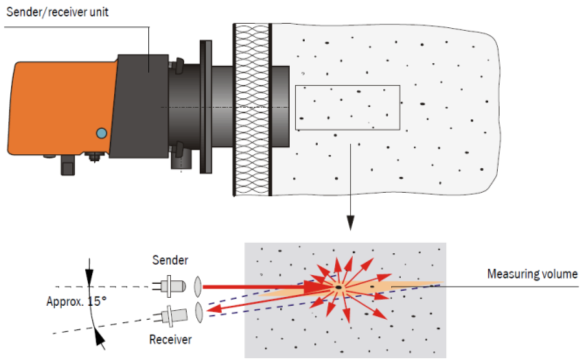



**Figure 6 Physical background- scattering intensity vs. particle size/wavelength (Grimm presentation).**

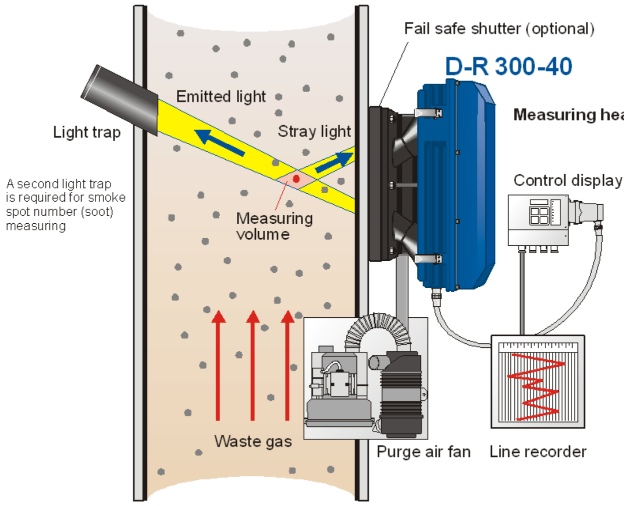
As a rule of thumb, the intensity of the scattered light for very small particles ($r/\lambda < 1$) is inversely proportional to particle diameter to the power 6. For most instruments, particles with a diameter size smaller than 0.1 µm are not detected.

Light scattering AMS can be in-situ or extractive. With in-situ AMS, the light beam crosses the flue gases inside the flue gas channel. For extractive scattering AMS, a sample of flue gas is extracted and is analyzed outside the flue gas channel section. With extractive AMS, flue gases are heated and/or diluted in order to avoid condensation, and the water droplets are evaporated before entering the measuring cell. Extractive AMS are therefore preferred for flue gases containing water droplets which would also scatter the light and interfere with in-situ measurement.

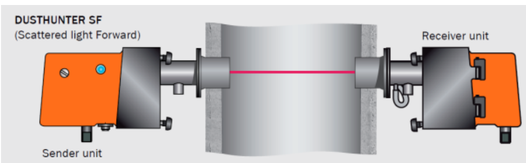
Scattering AMS of different types are illustrated in Figure 7 below.



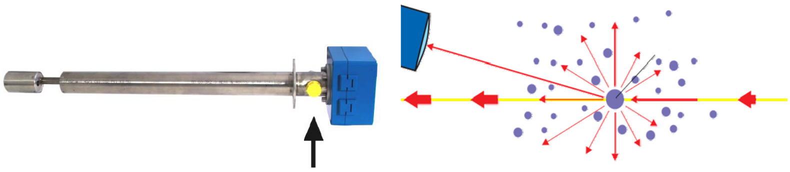

Back scattering wall mounted device (SICK Dusthunter SB)



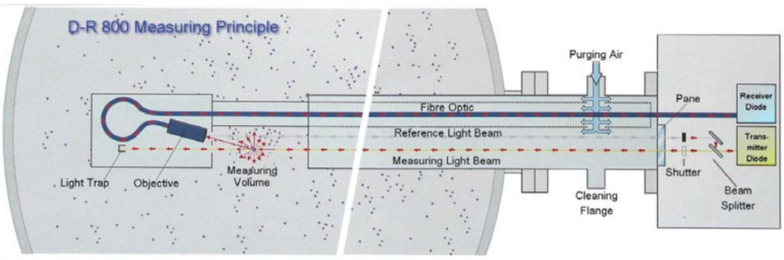

Back scattering wall mounted device (DURAG DR-300-40)

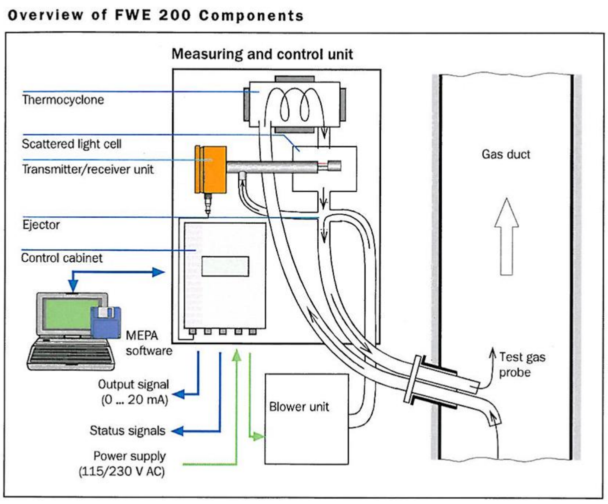



Forward scattering wall mounted devices (SICK, Dusthunter SF)

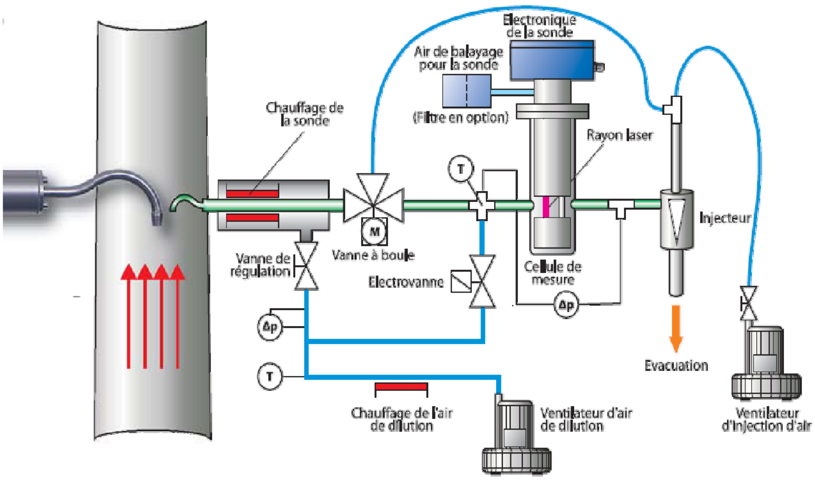


Forward scattering probe (DAURAG, DR800)

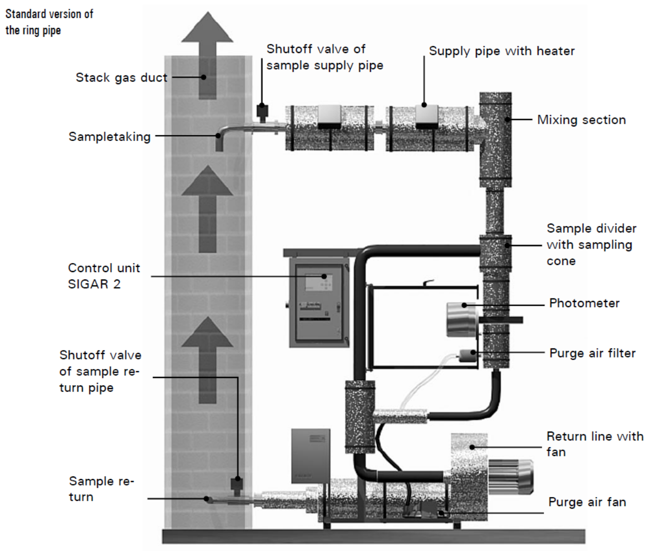



Extractive forward scattering device (SICK FWE200)




Extractive forward scattering from DURAG (D-R 820 F)




Extractive forward scattering from SIGRIST (SIGRIST - STACK GUARD)

**Figure 7: Illustrations of different light scattering devices.**

#### 5.4.2.2. Performances limitations of light scattering automated measuring systems

Several limitations of the light scattering instruments are listed below:

- they require a very homogeneous dust distribution;
- the instrument answer can vary with different dust;
- possible interferences caused by external light sources;

- possible deterioration of the optical part. Most of the AMS include air purging systems or blowers to keep the optical windows clean;
- the measuring point or the measuring line has to be representative of the average dust emission across the flue gas channel section;
- for extractive scattering instruments, issues related to non-isokinetic sampling + possible plugging of the sampling/heating systems.

### 5.4.3. Beta gauge automated measuring system

#### 5.4.3.1. General principle

**In β-radiation attenuation AMS, flue gas containing the dust is drawn through a glass fibre filter tape which collects the dust. The filter tape, wound on a roll, moves sequentially so that that a spot of dust is collected on the tape over the period that the tape is stationary. The filter tape is then automatically advanced to a C14 source, which emits β rays. The β radiation radiation is both absorbed and scattered by the collected dust on the tape, depending on the the amount and composition of dust present. The intensity of the attenuated radiation is measured by a detector, typically a Geiger-Müller tube. A reference measurement is provided provided by passing β radiation through an unexposed section of tape. The operating principle of a beta gauge monitor is illustrated in Figure 8 below. Pictures of the beta gauge commercialised by DURAG and by ENVIRONNEMENT SA are presented in**

Figure 9.

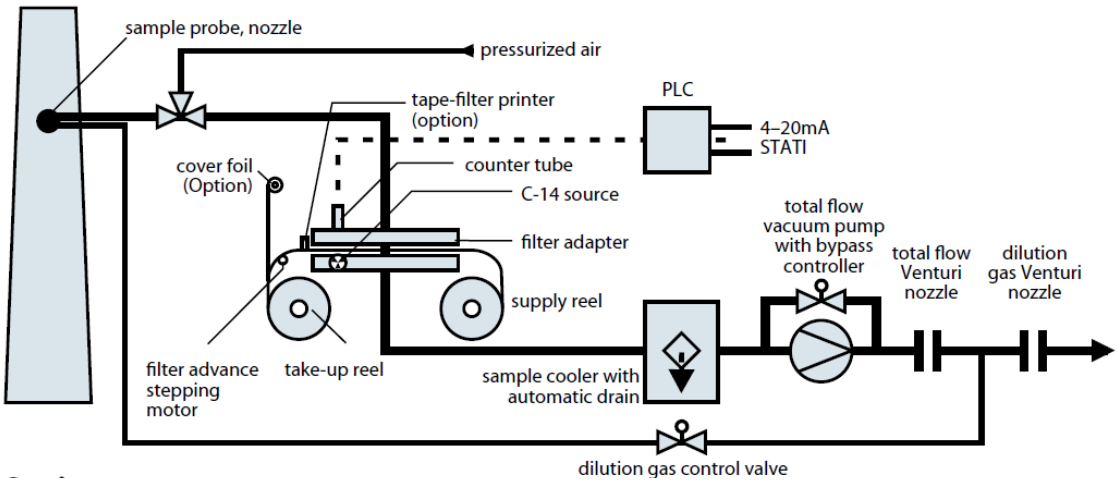



**Figure 8 : Operating principle of a Beta gauge monitor with dilution of the sample (Verewa Beta gauge monitor F-904-20, Durag catalogue).**

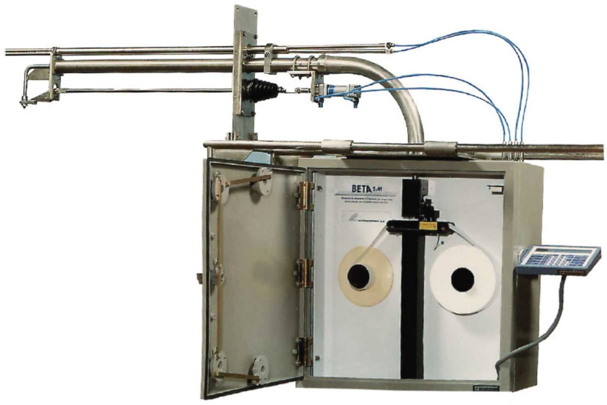
| | |
|---|---|
|  |  |
| Verewa Beta gauge monitor F-904-20 (Durag catalogue) | ENVIRONNEMENT SA Beta 5M Beta gauge monitor (Folder Environnement SA) |

**Figure 9 : Picture of two Beta gauge monitors.**

If the composition of the dust is relatively constant, the attenuation of the radiation can provide a measure of the thickness of the spot of dust, and hence the mass of collected material. The sampling system is usually heated; therefore wet gases can be sampled. The beta-gauge technique is essentially particle size independent, minimizing the effects of changing process conditions on the system calibration line.

The sampling time can be extended in order to get more dust on the filter and consequently to improve the measurement precision.

Afterwards, the chemical composition of the dust deposit on the glass fibre filter tape can be investigated if needed.

#### 5.4.3.2. Performances limitations of beta gauge automated measuring system

Several limitations of the beta gauge instruments are listed below:

- experience shows that these types of monitoring systems require lots of maintenance;
- issues related to non-isokinetic sampling + possible plugging of the sampling/heating systems;
- the dust concentration at the sampling point should be representative of the average dust emissions across the complete section;
- the time needed for moving the filter tape for the next sampling, measuring the response without and with dust deposits on the filter can require up to 5 minutes. Some local authorities consider this as non-availability of the monitor and therefore

monitoring dust with a Beta gauge monitor cannot be considered as a continuous monitoring. This might result in the rejection of this type of AMS.

# 6. Compliance with European legal requirements

The most relevant legal requirements for the operators are those related to (1) the choice of a dust AMS with relevant certificates proving its suitability for its monitoring purposes, (2) the choice of an appropriate measurement location and (3) the calibration and the follow-up of the AMS after its installation on site.

## 6.1. Dust monitoring system

### 6.1.1. Dust automated measuring system on the market

All new AMS need to have a QAL1 certificate delivered in accordance with the EN 15267-3. The TÜV clearly identifies the AMS that have been tested in full accordance with the EN 15267-3 on their website www.qal1.de.

Lots of dust AMS available on the market have been tested by official laboratories like the TÜV in Germany or MCERT in the UK, and have a type test approved certificate. Most of these certificates (and especially those delivered by the TÜV prior to the publication of the EN 15267-3) provide a reliable guarantee about the monitoring capability of the dust AMS they refer to, but they are not 100% compliant with the EU requirements.

Table 5 summarizes results from the QAL1 certificates of some AMS chosen at random. *Note: neither Laborelec nor Concawe are recommending the use of those brands.* The table presents the manufacturer, the AMS type, the measuring principle, the certified measuring range and the total expanded uncertainty. The certified measuring range is given in the measuring units (when mentioned on the QAL1 certificate) and in $mg/m^3$. The ELV max is the daily ELV value that could be checked for large combustion plants with the AMS. The total expanded uncertainty in $mg/m^3$ results from all the individual contributors to the measuring uncertainties. The values are low, from 0.35 to 1.38 $mg/m^3$ for an extractive AMS. These values are in fact lower than the uncertainty attached to the manual methods. Based on this uncertainty, the minimum ELV that could be checked with the AMS has been calculated. Note that the relation between the AMS measuring units and the corresponding $mg/m^3$ will depend on the dust characteristics and the expanded uncertainty should be calculated for the site where the AMS is intended to be installed. This latter is almost never done by the operators and the data from the initial lab testing type approval is used instead.

**Table 5: Data from AMS QAL1certificates proposed on the market.**

| Manufacturer | Type | Principle | Certified Measuring Range | | | Tot. Exp. Uncertainty | |
|---|---|---|---|---|---|---|---|
| | | | meas.units | $mg/m^3$ | ELV max (1) $mg/m^3$ | $mg/m^3$ | ELV min (2) $mg/m^3$ |
| SICK | Dusthunter C-200 | transmission + forward scattering | Ext. 0-0.1 | 15 | 38 | **0.82** | 4 |
| | Dusthunter SB100 | backward scattering | SE 0-100 | 15 | 38 | **0.626** | 3 |
| DURAG | D-R 320 | backward scattering | SL 0-500 | 7.5 | 19 | **0.35** | 2 |
| | DR800 | forward scattering | T 0-100 | 15 | 38 | **0.53** | 2 |
| PCME | QAL181 | forward scattering | 100 | 15 | 38 | **0.6** | 3 |
| Dr Födisch | PFM 06 ED | ectractive, forward scattering | | 15 | 38 | **1.23** | 5 |

(1) CMR has to be not larger than 2.5 times the daily ELV
(2) uncertainy from QAL1 may be max. 22.5% of the ELV for dust
(75% of the total max uncertainty from IED, i.e. 75% of 30% of the dust ELV)

The standard EN 13284-1 mentions that with a 30-minute sampling the uncertainties on dust SRM measurement amount to approximately 2 mg/m³. This value is for measurement on saturated flue gases and will be probably lower on dry flue gases.

For low dust concentrations, the uncertainty of the manual dust measurement in accordance with the SRM is higher than the uncertainty of the AMS itself. However the AMS has to be calibrated by the SRM results.

### 6.1.2. Location of the AMS

The general requirements about the AMS location are given in the standard EN 15259, and specificities for dust AMS are developed in the standard EN 13284-2.

It has to be stressed that, at least as important as the brand and type of the AMS, the choice of an appropriate measuring section is crucial to get reliable dust concentration monitoring, but also to get reliable SRM measurements in order to calibrate the AMS.

This includes the accessibility to the measuring platform, an appropriate measuring platform, appropriate measuring points, and as for the SRM measurements, an appropriate measuring section without perturbation of the flue gases flow and of the dust concentration distribution. Chapter 5.2 "Measurement section and measurement site" of EN 15256 states that: *"Plants designed or adapted to enable representative sampling have a section of the waste gas ducting engineered to ensure an ordered flow profile free from vortexing and backflow, where a measurement plane is located that provides a grid of sampling points sufficient to assess the distribution of measurands and reference quantities. The measurement site allows access to the sampling plane for typical sampling equipment via a platform that enables measurement personnel to work safely and efficiently."* Different examples are given for the measuring port and for vertical/horizontal and circular/rectangular ducts in the informative annex A. It is impossible to present all the examples in this report. An illustration of a vertical circular duct and the associated working platform is given in Figure 10.

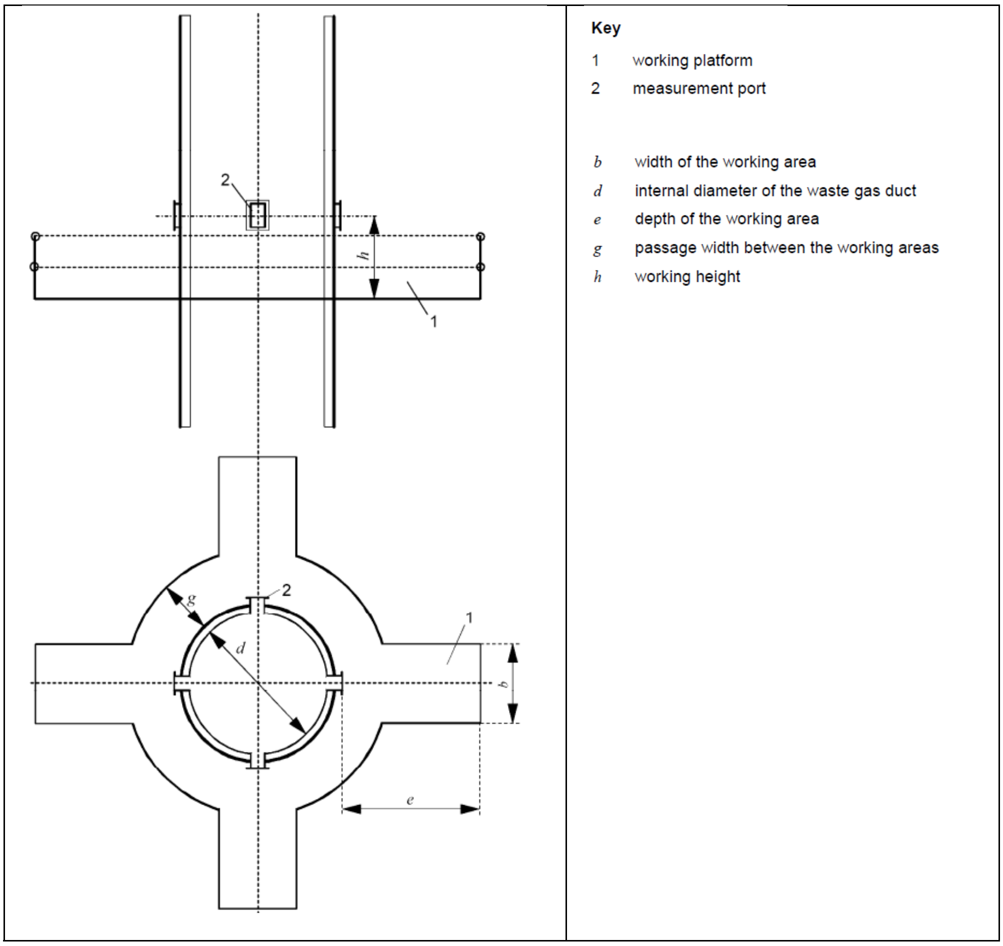



**Figure 10: Example of a working platform and the position of measurement ports in a vertical round waste gas duct (source: EN 15267, figure A.7).**

The standard is not strongly prescriptive on how to reach the requirements. The flow characteristics to fulfil in order to get reliable measurements are summarised in chapter 5.3.2. as well as how the standard recommends to fulfil it, i.e. by choosing a section of the duct with at least five hydraulic diameters of straight duct upstream of the sampling plane and two hydraulic diameters downstream (five hydraulic diameters from the top of a stack). A few other guidelines are given such as working platforms with sufficient load bearing capacity (up to 300kg), sufficient depth of the working area i.e. sum of the internal duct diameter or depth and the wall thickness + 1.5 m for flange-on instruments, and working height from the platform to the measuring flange of approximately 1.2 to 1.5 m.

## 6.2. QAL3 checks

QAL3 tests are described in the standard EN 14181. Between the yearly checks of the AMS by the accredited laboratory performing parallel SRM measurements, the zero and a span point of the AMS have to be checked at a frequency not lower than the time interval referred in the QAL1 as "maintenance interval". These checks have to be performed with test gases or with surrogates.

There are no dust test gases. So, QAL3 has to be performed with optical filters.

In the revised version of the EN 14181 it is stated that if the internal checks referred to as zero and span checks are certified in a QAL1 test report according to EN 15267-3 as QAL3 compliant then they may be used for QAL3.

In the revised version of the EN 14181 it is allowed to use fixed values as limits on the control chart for the QAL3 instead of values calculated from the QAL1 certificate, and ±50 % of the maximum permissible uncertainty may be used to establish the alarm limits of the control chart.

## 6.3. Other parameters to be measured

Emission monitoring values have to be reported in the same units as the ELV, i.e. mg/Nm³ @ reference $O_2$. Most of the time, the oxygen is already measured for the reporting of gaseous pollutants. Gaseous pollutants are often measured by extractive devices in normalised and dry conditions. However, where dust monitoring is required, in addition to the dust AMS, other instruments have to be installed for the environmental reporting:

1. AMS for moisture;
2. Temperature;
3. Pressure;
4. AMS for oxygen.

The uncertainties of these instruments are contributing factors to the uncertainty on the environmental dust reporting at standardised conditions (dry gases; 1013 hPa, 273K and at $O_2$% ref). In addition, moisture, temperature and pressure are needed to calculate the calibration functions when comparing SRM and AMS values, and erroneous values will result in erroneous calibration function.

## 6.4. Revision of the EN 13284

Recently it has been recognised that it can be very difficult to get a monitoring system installed and approved in accordance with the EN 14181 for dust concentrations below 10 mg/m³ and where the maximum allowed uncertainty related to the ELV is about less than a few mg/m³. This is because the uncertainties on the results are significant compared with the measured values for both the AMS (instrument uncertainty and representativeness of the measuring point) and the SRM needed for the AMS calibration/ validation. Part 1 and part 2 of the EN 13284 are currently being revised by the CEN TC/264, WG5 to address cases where low dust concentrations make it impossible to comply with the existing standards. The revised part 1 is expected to be published by the beginning of 2017 and the revised part 2 by the end of 2016.

The main objective of the revision is to reduce the uncertainties of the SRM measurements and to provide an adapted work method when the concentrations are very low. In part one, it is expected that the expanded uncertainty of the SRM should not be higher than a defined proportion of the ELV. Different solutions will be possible to reduce the SRM uncertainties, such as longer sampling times, higher sampling flow, etc. However, it will also be acknowledged, that in some cases, when the dust concentrations are very low compared to the uncertainty of the SRM measurement, neither the check of the ELV nor the calibration of the AMS with the SRM make sense. In this case alternative methods will be proposed for the handling of the results and for the monitoring system.

An accurate calibration of a dust monitor by the instrument manufacturer is impossible because the calibration depends on the dust characteristics such as diameter size distribution, colour, shape and chemical composition. However, for places where it is impossible to get manual measurements accurate enough for a calibration, "manufacture calibration" from the AMS manufacturer, taking into account average calibration factors might be allowed. This would allow for a qualitative measurement of the dust variation but certainly not for an accurate quantification as required in the EU standards.

The requirements for the QAL2 will also be revised. For example, it might be possible to reduce the total 15 results needed for the QAL2 provided that the total SRM sampling time stays the same. Therefore, each individual result will come from a longer sampling time, resulting in a lower uncertainty on the final result. Under certain conditions, it will be allowed to extend the validity of the valid calibration range up to 50% of the ELV.

The revision of the EU standard EN 13248 on low dust concentrations, part one and part two, will allow some flexibility for cases where the uncertainty on the measurement is too high compared with the measured value for drawing any quantitative conclusion. However, the revised standard will not solve all the challenging cases.

## 6.5. Handling of the semi-volatile compounds

EN 13284 defines dust as "*particles, of any shape, structure or density, dispersed in the gas phase at the sampling point conditions which may be collected by filtration under specified conditions after representative sampling of the gas to be analysed, and which remain upstream of the filter and on the filter after drying under specified conditions*".

EN 13284 says that the filters used for the SRM measurements shall be dried for (1) at least 1 h before sampling at a temperature of at least 20 °C above the maximum temperature reached during sampling and post-sampling treatment, and (2) for at least 1 h at 160°C or at the temperature used for sampling. The temperature used while conditioning shall be indicated in the test report.

Indeed, other drying temperatures are allowed when the presence of semi-volatile compounds is suspected. Semi-volatile compounds are defined as compounds being in particulate form at low temperature and ion gaseous form at higher temperature. They can, under specific conditions, be measured/ monitored together with dust. For flue gases where semi-volatiles are present, the SRM conditions for the sampling and for the drying of the filters have to be in line with the operating conditions of the dust AMS in order to get comparable results.

Examples of semi-volatile or unstable compounds mentioned by the standard are:

- hydrates from power plant equipped with desulphurisation processes;

- $SO_3$ and/or organic compounds from heavy fuel oil power plants or diesel engines;
- semi volatile boron compounds from glass furnaces.

Experience shows that the weight of $SO_3$ trapped on the filter together with the dust from coal power plants can amount to up to 50% of the dust.

# 7. Conclusions

The key points from the information provided in the earlier sections of this document are summarized hereafter:

(1) Given the emission limit values set by the IED, the data available in the Refinery BREF and the LCP database, it is reasonable to suggest that, dust emissions are below 5 mg/Nm$^3$ for most gas-fired units in refineries.

(2) Low dust concentration measurement and monitoring are challenging. Some characteristics of dust make continuous monitoring even more complicated than for other pollutants. When the concentrations are very low, it may not be possible to comply with the current standard requirement regarding the quality of the data, (i.e. uncertainty requirement not possible to be met).

a. The existing standards and required measuring uncertainties have been developed in relation with emissions limit values in the range of 10 mg/Nm³ dust. For concentrations below 10 mg/Nm$^3$ it can be very difficult to get an Automated Measuring System (AMS) installed and approved in accordance with the EN 14181.

i. Currently, the two AMS better suited to measure low concentrations of dust are the light scattering method and the beta-gauge methods. The light scattering method does not provide a direct estimation of the dust concentration, so a correlation (calibration) must be established between the instrument output and simultaneous measurement using the Standard Reference Method (SRM).

ii. For low dust concentrations the uncertainty of the manual dust measurement can be higher than the uncertainty of the AMS itself. However the AMS has to be calibrated by the results of the manual measurements.

iii. With low dust concentrations several sources of uncertainty, other than the measurements themselves, may strongly interfere with the dust concentration quantification. These uncertainties are the sources of random variations on both SRM and the AMS values. The uncertainties can make it impossible to fit the calibration criteria stated in the EU regulation.

b. The location where the dust measurements take place has to comply with a number of requirements that might be challenging to comply within an existing stack or duct.

(3) The revision of the EU standard EN 13248 on low dust concentrations, part one and part two, will allow some flexibility for cases where the uncertainty on the measurement is too high compared with the measured value for drawing any quantitative conclusion. However, the revised standard will not solve all the challenging cases.

(4) When the measurements are not possible within a relevant uncertainty, the added value of a continuous monitoring for quantification purposes is questionable. In this case, a monitoring system will only act as a qualitative detection. When the process is expected to be stable in terms of combustion conditions and fuel, discontinuous measurements could be considered as a valid alternative for the quantification of the dust emissions.

# Appendix A. References

[1] Consolidated table for all countries in the NFR14 format v15_GF. United Nations Economic Commission for Europe (Environment and Human Settlements Division, UNECE). Database downloaded from the EEA website. http://www.eea.europa.eu/data-and-maps/data/ds_resolveuid/4e4e71b79c584e45989949d87864a70a

[2] Best Available Techniques (BAT) Reference Document for the Refining of Mineral Oil and Gas Industrial Emissions Directive 2010/75/EU Integrated Pollution Prevention and control. 2015. EUR 27140 EN. Hereafter called REF BREF

[3] Plant-by-plant emissions of SO2, NOx and dust and energy input of large combustion plants covered by Directive 2001/80/EC. Version February 2015. http://www.eea.europa.eu/data-and-maps/data/plant-by-plant-emissions-of-so2-nox-and-dust-and-energy-input-of-large-combustion-plants-covered-by-directive-2001-80-ec-2

[4] COMMISSION IMPLEMENTING DECISION of 9 October 2014 establishing best available techniques (BAT) conclusions, under Directive 2010/75/EU of the European Parliament and of the Council on industrial emissions, for the refining of mineral oil and gas. 214/738/EU).

[5] DIRECTIVE 2010/75/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 24 November 2010 on industrial emissions (integrated pollution prevention and control).

# Appendix B. Distribution list

| Electronic distribution | | | | |
|---|---|---|---|---|
| **Recipient** | | **Outlook** | **Media** | **Remark** |
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| SPRT | | SPRT | SD link | |
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