

Commission of the European Communities



Methods of sampling and analysis for sulphur dioxide, oxides of nitrogen and particulate matter in the exhaust gases of large combustion plant



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### LIST OF TECHNICAL ABBREVIATIONS

	Btu	=	British thermal unit
	°C	=	centigrade
	°F	=	farenheit
	FS	=	full scale
	GJ	=	gigajoule
	g	=	gram
	h	=	hour
	K	=	Kelvin
	kg	=	kilogram
	kPa	=	kiloPascal
	kWh	=	kilowatt-hour
	1	=	litre
	1Ъ	-	pound mass
	mbar	=	millibar
	m <sup>3</sup>	=	cubic metre
	mg	=	milligram
	ml	=	millilitre
	ms	=	millisecond
	mth	=	month
	MW	=	Megawatt, thermal
	μg	=	microgram
	μm	=	micrometer
	nm	=	nanometer
	Nm <sup>3</sup>	=	normal cubic metre
·	ррЪ	=	parts per billion
	рН	=	negative exponent of the concentration of hydrogen ions
	ppm v/v	=	parts per million by volume
	S	=	second
	sđ	=	standard deviation
	th	=	thermie = 10 <sup>3</sup> Kcal
	URL	=	upper range limit
	% v/v	=	percentage by volume
	% w/w	=	percentage by weight

wk = week

yr = year

Remark: All volume data in this report refer to standard conditions (0°C, 101.3 kPa), unless otherwise indicated.

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#### INTRODUCTION

The European Commission has forwarded to the Council a proposal<sup>1</sup> to limit the emissions of  $SO_2$ ,  $NO_x$  and particulates from large combustion plants. The proposed regulation requires the continuous measurement of emissions of these components using approved equipment. All results are required to be calculated to a fixed oxygen content as well as to standard dry conditions using a data treatment procedure and the half-hourly and daily mean values are required to be compared with the limit concentrations.

Within this study, requirements for measuring and controlling the emissions in the different Member States are presented. Portugal and Spain have been included in the study as they will be Member States in future.

Information was requested on the calibration procedures and performance of installed monitoring systems and the organisation (authorised institution, local authority or industry) responsible for the measurements was established.

The measurement methods used in the Member States are listed and described, together with a short comparison and evaluation of instrumentation, for each component to be measured. A survey is given of the number of existing plants including boilers, cement works, ore roasting plants and brick works which would be subject to the Commission's proposals. As far as possible, a breakdown is given as a function of the thermal output of the plants as well as of the fuel type. In some cases the number of plants was determined and in other cases the number of units only. It was always difficult to get exact information on installations in the industrial sector and in order to differentiate between the number of plants and single units, some estimation was necessary. No existing brick works exceeding 50 MW rated heat input (heat release) were identified.

The numbers of installations already equipped with continuous measuring devices are listed. A survey of the expected investment costs for compliance with the Commission's proposals is given, also a figure for the calibration and the maintenance costs.

#### 1. NATIONAL EMISSION REGULATIONS

Requirements for performing emission measurements vary widely between the Member States. These requirements may be encoded in specific legislation or in more general air pollution legislation or may be set down elsewhere in guidance notes. Details of air pollution legislation and of requirements for emission measurements in each Member Country, are given in the following sections.

A summary of the emission measurements at present performed in Member States is given in Table I.

The existing emission limits in Member States<sup>2</sup> are presented in Table II.

	Emission Component							
Member State	Partic disc	ulates cont	SC disc	cont	NO disc	x cont	0 <sub>2</sub> / disc	CO <sub>2</sub> cont
Belgium	X	X	X	<u>9 - 1 - 2 - 10 - 2 - 10 - 20 - 20 - 20 - </u>	X	X		x
Denmark	X	X	X	X	X	X		x
Germany	x	x	x	x	x	X		x
France	X	x	x	x	Х		x	х
Greece	X		х		X			х
Ireland	Х				х			х
Italy	X	X			X			x
Luxembourg	X		X		X			x
Netherlands	Х	X	X	X	X	X		х
UK	Х	X	Х	Х	Х		Х	Х
Portugal	 X	X	x		ويت جند خيد (يين وي خلا وي حله وي		X	
Spain	Х	X	x	Х	Х	х		x

•

TABLE I. - Summary of Emission Measurements Performed in Member States

TABLE	II.	-	Emis	sion	Limit	s in	1 Membe	r States
							the second se	

Membe r	Emission Limits							
State	Particulates	Sulphur Dioxide	Nitrogen Oxides (as NO <sub>2</sub> )					
Belgium	Coal fired power stations <20% ash 500 mg/Nm <sup>3</sup> >20% ash 350 mg/Nm <sup>3</sup> Other solid fired installations special protection areas 150 mg/Nm <sup>3</sup> other areas 300 mg/Nm <sup>3</sup> Liquid fired installations >2000 th/h (~2 MW) 250 mg/Nm <sup>3</sup>	Liquid fired power stations 5000 mg/Nm <sup>3</sup> Other liquid fired installations <1 x 10 <sup>6</sup> k cal/h 850 mg/Nm <sup>3</sup> >1 x 10 <sup>6</sup> k cal/h 4700 mg/Nm <sup>3</sup> More stringent limits in special protection areas.						
Denmark	0il fired power stations and industrial boilers <30 MW new 300 mg/Nm <sup>3</sup> existing 500 mg/Nm <sup>3</sup> 30-100 MW linear variation between limits for extremes 100-300 MW 150 mg/Nm <sup>3</sup> >300 MW 1 g/kg fuel		Coal fired power stations 1000 mg/Nm <sup>3</sup>					
PRG	Solid fuel installations >50 MW new 50 mg/Nm <sup>3</sup> Existing lignite 80 mg/Nm <sup>3</sup> Coal 125 mg/Nm <sup>3</sup> Liquid fuel installations >50 MW 50 mg/Nm <sup>3</sup>	Solid fuel installations new >50 <100 MW 2000 mg/Nm <sup>3</sup> except fluidised 400 mg/Nm <sup>3</sup> * >100 <300 MW 2000 mg/Nm <sup>3</sup> * except fluidised 400 mg/Nm <sup>3</sup> * existing >50 <300 MW 2500 mg/Nm <sup>3</sup> until 1993 then same limits as for new plant. >300 MW 400 mg/Nm <sup>3</sup> * Liquid fuel installations, new >50 MW 1700 mg/Nm <sup>3</sup> >100 MW 1700 mg/Nm <sup>3</sup> >300 MW 400 mg/Nm <sup>3</sup> * Existing as for existing solid fuels Gaseous fuel installations >100 MW 35 mg/Nm <sup>3++</sup>	Solid fuel installations >50 MW new 800 mg/Nm <sup>3</sup> except pulverised coal/wet ash 1800 mg/Nm <sup>3</sup> (>50 MW, 400 mg/Nm <sup>3</sup> ; >300 MW, 200 mg/Nm <sup>3</sup> ) <sup>+</sup> existing 1000 mg/Nm <sup>3</sup> except pulverised coal wet ash 2000 mg/Nm <sup>3</sup> (>50 MW 650 mg/Nm <sup>3</sup> ) <sup>+</sup> pulverised coal, wet ash 1300 mg/m (>50 MW 650 mg/Nm <sup>3</sup> ) <sup>+</sup> ,** Liquid fuel installations >50 MW new 450 mg/Nm <sup>3</sup> ; >300 MW, 150 mg/Nm <sup>3</sup> ) <sup>+</sup> existing 700 mg/Nm <sup>3</sup> (>50 MW, 450 mg/Nm <sup>3</sup> (>50 MW, 50 mg/Nm <sup>3</sup> (>50 MW, 450 mg/Nm <sup>3</sup>					
Greece	All plant, all fuels new 100 mg/Nm <sup>3</sup> existing 150 mg/Nm <sup>3</sup>							

Notes

calculated emission rate for SO<sub>2</sub> shall not exceed 15% for units >300 MW 40% for units 100-300 MW 25% for fluidised bed units >50 MW <300 MW \*

+ figures in brackets required by the Environmental Minister of the Federal States of Germany

++ special standards for some industrial gases

for unlimited lifetime \*\*

Member	Emission Limits						
State	Particulates	Sulphur Dioxide	Nitrogen Oxides (as NO <sub>2</sub> )				
France	Solid/fuel fired post 1976 150 mg/th Overrun authorised: 500 mg/th 200 hrs/yr 250 mg/th 400 hrs/yr Solid fired pre 1976 <1 g/th Overrun authorised: 1 g/th 200 hrs/yr >2 g/th in no case Liquid fired, pre 1976, 250 mg/th Overrun authorised: <1 g/th 200 hrs/yr <0.5 g/th 400 hrs/yr Special prefectoral order for all 'cla	Limits for some special protection areas only assified' installations					
Ireland		÷	-				
Italy	Solid/fuel fired Combustion plant >1000 kg/hr Sliding scale, minimum 250 mg/Nm <sup>3</sup>	Immaission limits only plus S in oil and coal					
Luxembourg	-	-	-				
Netherlands	New coal fired installations 20 g/GJ (approx 50 mg/Nm <sup>3</sup> )	New coal fired power stations 230 g/GJ (approx 550 mg/Nm <sup>3</sup> ) New industrial coal fired installations Pre 1990 600 g/GJ (approx 1430 mg/Nm <sup>3</sup> ) Post 1990 230 g/GJ (approx 550 mg/Nm <sup>3</sup> )	New coal fired installations 270 g/GJ (approx 640 mg/Nm <sup>3</sup> )				
UK	Coal fired power stations pre 1958 460 mg/Nm <sup>3</sup> 1958-1974 115 mg/Nm <sup>3</sup> prorata for ash content >20% post 1974 115 mg/Nm <sup>3</sup> Oil fired power stations post 1974 115 mg/Nm <sup>3</sup> Furnaces for indirect heating Coal fired 543-1362 mg/Nm <sup>3</sup> depending on size Oil fired 139-349 mg/Nm <sup>3</sup> depending on size	- `	-				
Portugal	-	-	-				
Spain	Power stations - solid fuel polluted areas new <50 MW 250 mg/Nm <sup>3</sup> 50-200 MW 150 mg/Nm <sup>3</sup> >200 MW 150 mg/Nm <sup>3</sup> other areas new existing <50 MW 250 mg/Nm <sup>3</sup> 750 mg/Nm <sup>3</sup> 50-200 MW 150 mg/Nm <sup>3</sup> 350 mg/Nm <sup>3</sup> >200 MW 150 mg/Nm <sup>3</sup> 350 mg/Nm <sup>3</sup> power stations - liquid fuel new existing <50 MW 175 mg/Nm <sup>3</sup> 200 mg/Nm <sup>3</sup> 50-200 MW 150 mg/Nm <sup>3</sup> 200 mg/Nm <sup>3</sup> >0 mg/Nm <sup>3</sup> 200 mg/Nm <sup>3</sup> >200 MW 120 mg/Nm <sup>3</sup> 175 mg/Nm <sup>3</sup> >200 MW 120 mg/Nm <sup>3</sup> 175 mg/Nm <sup>3</sup> >200 MW 100 mg/Nm <sup>3</sup> 175 mg/Nm <sup>3</sup> >200 MW 100 mg/Nm <sup>3</sup> 175 mg/Nm <sup>3</sup>	Solid fuel power stations coal/anthracite 2400 mg/Nm <sup>3</sup> lignite 9000 mg/Nm <sup>3</sup> Liquid fuel power stations existing 5500 mg/Nm <sup>3</sup> new 3000 mg/Nm <sup>3</sup>	-				

## 1.1 Belgium<sup>3</sup>

The basic air pollution control law of 1964 has led to several implementation decrees. In 1971 five special protection zones were created. In 1975 sulphur dioxide and particulate emissions standards were established for industrial furnaces and a five year programme for reducing fuel sulphur content was introduced. Maximum Bacharach numbers for smoke from power stations and industrial installations were specified.

At present there is no legal requirement for emission measurements, however, a law which will require continuous measurement of particles, sulphur dioxide and oxides of nitrogen at large combustion plant, from March 1986 onwards, is in preparation. Some emission measurements are already performed by authorised institutions, in some cases at regular intervals, so that the plant operator can avoid dangerous emissions and ensure that emissions do not exceed the limits set down in the plant license. Licenses, issued by the local authorities, may specify the required frequency of emission measurements. Fuel sulphur contents are analysed instead of making  $SO_2$  emission measurements.

## 1.2 Denmark<sup>3,4</sup>

Provisions to enable environmental legislation<sup>3</sup> were introduced in the Environmental Protection Act of 1973. The ensuing Heavily Polluting Enterprises Order passed in 1974 regulates the building and operation of large industrial installations including heat and power generation facilities. The National Agency of Environmental Protection is empowered to issue emission guidelines for  $SO_x$  and  $NO_x$  which are based on the performance of the best available technology and are implemented by municipal and county authorities, subject to appeal to central Government. A permitting system is enforced for major polluters.

Legislation<sup>4</sup> passed in 1984 on the limitation of  $SO_2$  from power plants sets a national  $SO_2$  emission quota derived as a proportion of the 1980 emission. The reduction is to be achieved by restricting the sulphur content of the fuel and by limiting the sulphur dioxide emissions from new power plant. This legislation complements the existing Danish sulphur oxides control policy which regulates fuel sulphur content.

### 1.3 Federal Republic of Germany $5^{-12}$

The general law on air pollution<sup>5</sup> was introduced in 1974.

The Ordinance on Large Combustion Plant (Großfeuerungsanlagen-Verordnung)<sup>6</sup> of 1983 specifies limits for, and requires the measurement of, emissions of particulates, sulphur dioxide, nitrogen oxides, carbon monoxide and oxygen from solid and liquid fuelled installations with a thermal rating of 50 MW and over and gas fuelled installations with a thermal rating of 100 MW and over. The only types of combustion installation to which the Ordinance does not apply, are waste incineration units, coke-ovens, under-grate firing installations, gas turbines, afterburners and furnaces whose combustion products are used to directly heat, dry or otherwise process another material.

All combustion installations covered by the Ordinance which have a residual lifetime in excess of 10,000 hours (from 1 July 1984) must possess continuous measurement devices, in accordance with the specified requirements shown in Table III, these also cover some discontinuous measurements (performed at three yearly intervals). At least three single measurements must be taken at the maximum rating of the plant. When expressed as half-hourly mean values none of the measurements should exceed the emission limit.

The Technical Instructions for Maintaining Air Quality (Technische Anleitung zur Reinhaltung der Luft)<sup>7</sup>, specify emission limits and lay down requirements for continuous emission measurements to regulate the efficiency of abatement measures. Installations with mass emissions of particulates in excess of 15 kg h<sup>-1</sup> or sulphur dioxide emissions in excess of 100 kg h<sup>-1</sup> are subject to these requirements. In addition, cement kilns built after 1974 must provide for the continuous measurement of particulate concentrations, and sulphur dioxide must be measured downstream of sulphuric acid or gypsum plant using waste gases from ore-roasting plant containing >2% SO<sub>2</sub>.

Continuous emission measurement instruments must undergo a suitability test, performed by one of five authorised institutions, to guarantee certain minimum requirements. A list of instruments which have met these requirements is published by the Federal Minister of the Interior (Bundesminister des Innern). Once a continuous emission measurement instrument has been installed, a test of its functions must be carried out annually by an authorised

## TABLE III. - Emission Measurement Requirements under the Large Combustion Plant Ordinance (FRG)

## Continuous Measurements Particulate concentrations All solid and liquid fired plant. All solid and liquid fired plant and Nitrogen monoxide and gas fired plant >400 MW thermal input nitrogen dioxide (if the NO $_2$ emission is <5% of the total $NO_x$ emission only NO need be measured continuously). All solid fired plant, all heavy fuel Sulphur dioxide oil fired plant >100 MW. Carbon monoxide All plant >50 MW. All plant >50 MW. **Oxygen** Discontinuous Measurements Particulate concentrations All gas fired plant, where measurement is considered necessary. Nitrogen oxides All gas fired plant <400 MW thermal input. Sulphur dioxide At gas fired plant where measurement is considered necessary. Heavy metals Only in the case of special solid and liquid fuels.

Gaseous chlorine and fluorineAt solid fired plant and plant usingcomponentsspecial liquid fuels.

institution. An authorised institution must also carry out calibration against a specific manual measurement method, every three years for instruments installed on plant of more than 300 MW thermal input and every five years for those on smaller installations.

Continuous measurements are expressed as half-hourly mean values, adjusted to a specified oxygen content; 97% of these values must not exceed sixfifths of the emission standard and none must exceed twice the standard. In addition, none of the daily mean values may exceed the emission standard.

## <u>1.4 Greece<sup>3</sup></u>

Environmental law is still being developed in Greece and efforts have been focussed on the control of black smoke and SO<sub>2</sub>, especially in urban areas. The authorities can decide on emission values for particular cases more or less at their own discretion.

There is no legal requirement for emission measurements, but the Public Power Corporation makes manual measurements of sulphur dioxide, nitrogen oxides and particulates at irregular intervals at all its power plant. All power plants are equipped with continuous monitoring devices for carbon dioxide and oxygen.

## 1.5 France<sup>13-32</sup>

#### Existing Regulations

Large combustion installations fall, in conformity with French regulations, within the category of "Classified Installations". Their operation is regulated:

- by the application of laws, decrees, administrative orders and circulars<sup>13-32</sup>,
- by way of special regulations relative to "Classified Installations",
   defined by the Act<sup>26</sup> of 19th July 1976 and by the application decree<sup>27</sup> of
   21st September 1977, which stipulate that "Classified Installations" be
   subject to an authorisation expressed in the form of a prefectoral order

specifying the steps to be followed in order to ensure protection of the environment (the inclusion of an impact study in the authorization request files being mandatory). Each installation is therefore in possession of a specific file including special instructions for preventing the emission of pollutants at their source. The Commissioner of the Republic (Prefect) thereby has the authority, within the framework of a prefectoral order, to enforce any provisions judged necessary to protect the environment, including well defined means of monitoring the atmospheric environment and particularly the implementation of continuous measurement devices at the source of emission.

The operation of combustion installations is furthermore regulated by an interministerial order<sup>23</sup> of 20th June 1975 and by the application circular<sup>24</sup> of 18th December 1977 relative to the equipping and operating of thermal installations in view of reducing atmospheric pollution and conserving energy.

These cover the following points:

- the instrumentation of the generator and its correct operation. Thus, each generator with a power:
  - greater than 3000 th/hr should be provided with an automatic analyser of combustion gases showing, at least, the CO<sub>2</sub> content or any equivalent indication,
  - greater than 8000 th/hr should, in addition, be provided with an instrument for continuous measurement, direct or indirect, of the quantity of particulate emitted in the atmosphere.

These instruments (enumerated in the Order of the 20th June 1975) shall be approved (Order<sup>32</sup> of the 29th April 1977) by the Department of Measuring Instruments (SIM) and the tests shall be carried out by bodies designated by the SIM.

- Determining the height of the stack for SO<sub>2</sub> emissions.
- Particulate matter emissions: for installations with a power exceeding 8000 th/hr, provisions concerning particle emission shall be as follows:

- Black smoke index (Bacharach method - Std NF X 43002):

4 for installations set up after January, 1, 1976 5 for previous installations.

The existing particulate emission limits are shown in Table II.

The method to be used for the manual sampling and measurement of particulates in a gaseous flow is defined in the standard NF-X 44052.

The following additional points should also be noted:

- Instructions have been defined for special protection areas (highly urbanised areas, which include the Nord and Rhône departments, Paris and the surrounding region) with effect from 1 January, 1980. The particulate emission from power plant in these areas, whatever the start-up date, is limited to 150 mg/th. Sulphur dioxide emissions<sup>15-17,25,28-31</sup> are restricted by limiting the sulphur content of fuels and by regulating the height of stacks<sup>22</sup>. Specific operating conditions are applied in special protection areas.
- In the case of cement plant (Circular<sup>19</sup> of 25 August, 1971), the gases from the oven should not contain more than 150 mg/Nm<sup>3</sup> of particulates, in normal operation. Authorised overrun is limited to two days and the cumulative duration over one year shall be less than 200 hours.

Under no circumstances shall the particulate emission rate exceed  $1 \text{ g/Nm}^3$ .

The height of the stack is defined in the instructions<sup>23</sup> of 24 November, 1970.

Particulate emissions should be continuously monitored. Moreover, a gravimetric control check shall be carried out at least once a year by an approved body. Sampling shall be carried out over a minimum duration of one hour.

#### Proposed Regulations

The Ministry of the Environment has presented for discussion a set of proposed technical instructions, which would regulate the measurement of emissions of sulphur oxides, nitrogen oxides and particulates from combustion installations, incineration plant and electric fusion furnaces. A minimum list of pollutants to be measured will be drawn up for each category, at national level and the conditions for application will be stipulated by individual prefectoral orders. Oxygen content of the gaseous effluent will also be measured. It is proposed that installations consuming only gaseous fuels of  $\langle 0.2\% \ w/w \$  sulphur will be exempt from measuring emissions of sulphur oxides and particulates. All installations of greater than 50 MW thermal input will be obliged to make continuous measurements.

These provisions are expected to be put into effect for new installations at some date in the first half of 1986. Installations already in existance at this date will be expected to conform within a period of three years, by prefectoral order.

### 1.6 Eire<sup>33-36</sup>

Emissions from scheduled processes are controlled under the Alkali etc. Works Regulations Act<sup>33</sup> 1906, which is identical to the Act of the same name in the United Kingdom. It has not, however, been reinforced or extended by further legislation as in the UK.

The Control of Atmospheric Pollution Regulations<sup>34</sup> 1970 set limits for the length of time during which dark smoke can be emitted from commercial and industrial premises, under the Local Government (Sanitary Services) Act<sup>35</sup> 1962. The 1970 Act also provides for regulations to control sources of atmospheric pollution and regulate their establishment and operation. Maximum atmospheric concentrations of certain pollutants can be specified, as can the measurement and investigation of pollutant emissions. Regulations under the Act could require the licensing of persons engaged in specified works or processes and of premises from which pollutants are discharged to the atmosphere. Particular controls for atmospheric pollution can be specified for particular areas. The Local Government (Planning and Development) Act, 1963 is also relevant in parts. A new set of regulations, the Control of Atmospheric Pollution (Licensing) Regulations<sup>36</sup>, 1985 deals specifically with air pollution caused by the recovery, through burning, of certain metals from scrap, waste material and waste cable. A licensing system for such works is introduced, which requires the operator to use the best practicable means to limit emissions of smoke, dust, grit, gas, fumes and other pollutants.

At present there is no legal requirement for emission measurement but air pollution legislation is currently under review.

## 1.7 Italy<sup>37-41</sup>

The main piece of legislation in the field of air pollution is the Control of Atmospheric Pollution  $Act^{37}$  (1966). Fuel sulphur contents are regulated and there is a permit system for new power plant and other significant pollution sources.

There is no legal requirement for emission measurements but some measurements may be performed at power stations at the request of the local authority. The ENEL (Ente nazionale per l'Energia Elettrica) measures emissions of nitrogen oxides and particulates at some of its power stations. Nitrogen oxides are measured every three months during three day measurement campaigns. Particulates are measured by a manual method on the basis of rules defined by ASME test codes and UNICHIM methods except at post-1973 plant which are generally equipped with opacimeters either for combustion optimisation or for monitoring the efficiency of abatement equipment (Law No 880 of 18 December 1973)<sup>39</sup>. Where oxygen is continuously measured it is for the purpose of combustion control only.

Legal regulations concerning the measurement of <u>immissions</u> of sulphur dioxide, nitrogen oxides and particulates from power stations have been enacted. According to these acts a network has to be installed and equipped with continuous instruments for pollutant measurements and measurement of meteorological parameters. The data are centralised and statistically analysed. ENEL initiates the alert procedure.

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### 1.8 Luxembourg<sup>3</sup>

No specific air pollution control legislation exists but the Minister of Interior, Public Health and Work may authorise the communal administration to enforce certain requirements.

There is no legal requirement for emission measurements and, until now, the need for regular emission measurements by authorised parties has been laid down in the license of individual plant. Measurements of particulate, nitrogen oxides and sulphur dioxide emissions are performed irregularly by authorised institutions in accordance with Governmental decrees or by the operator himself.

### 1.9 Netherlands<sup>3</sup>

The Public Nuisance Act last revised in 1981 covers air pollution and provides for a permit system for the industrial installations governed by it. The Air Pollution Act (1970) applies to stationary sources which contribute significantly to ambient air pollution and which require a permit under this Act. National emission guidelines are decided in the context of a Policy Framework Report by national government. Local authorities have responsibility for the issue of permits and can apply stricter controls than those prescribed nationally.

There is no legal requirement for emission measurements and, until now, the need for regular emission measurements by authorised parties has been laid down in the license of individual plant. Measurements may be performed by the operator, by the local authorities or by a specialised institution, such as KEMA in the case of power plant or TNO for industry. The frequency of measurement varies but would usually be just once, at the start of operation, for small installations and a maximum of twice a year at large plant. Some power stations are required in their license to have continuous emissions measurement instruments which are calibrated at specified intervals. It should be noted that the Dutch emission standards are usually given in g/GJ so reference  $O_2$  or  $CO_2$  contents are not necessary. Consequently continuous measurement of the waste gas flow is necessary but measurement of  $O_2$  or  $CO_2$  concentrations is not. An Ordinance concerning large combustion installations is currently in preparation. The present draft requires that continuous measurement devices be installed on plant greater than 300 MW and that three yearly manual emissions measurements be made at installations in the 50-300 MW range. Smaller installations would only require a single measurement at the start of operation. The Ordinance will specify the conversion to a reference oxygen content, the averaging time and the conditions for the data treatment. It will also list the institutions which are authorised to perform emission measurements.

## 1.10 United Kingdom<sup>42-47</sup>

Legal provisions concerning the measurement of grit and dust emissions from furnaces (or groups of furnaces served by one chimney) burning pulverisd fuel or any other solid fuel at a rate of 100 lb  $h^{-1}$  (45 kg  $h^{-1}$ ) or more, or liquid or gaseous fuel at a rate equivalent to 1.25 x 10<sup>6</sup> Btu  $h^{-1}$  (0.4 MW) or more, are laid down in the Clean Air Act<sup>42</sup> 1956, as amended by the Clean Air Act 1968. The Clean Air (Measurement of Grit and Dust) Regulations<sup>43</sup> 1971, prescribe the administrative process to invoke these provisions in England and Wales and a similar set of Regulations apply in Scotland. A local authority may serve a notice on a plant operator requiring the making of measurements from time to time or at stated intervals of not less than three months, except in exceptional circumstances. The operators of the smaller plant, except for those burning pulverised fuel, may serve a counter notice on the local authority obliging them to carry out measurements from time to time at their own expense.

The British Standard BS 3405:1983<sup>44</sup> lists types of equipment which can be used to measure grit and dust emissions.

The control of major industries, scheduled processes under the Health and Safety at Work etc Act<sup>45</sup> 1974 together with the Health and Safety (Emission into the Atmosphere) Regulations<sup>46</sup> 1983 and the Alkali etc. Works Regulations Act<sup>33</sup> 1906, is centralised under the control of HM Industrial Air Pollution Inspectorate in England and Wales. In Scotland almost identical legislation applies and is administered by HM Industrial Pollution Inspectorate. The IAPI publishes Notes on Best Practicable Means<sup>47</sup> which set out the current requirements arising from the legislation for the main industrial classes of the Schedule, including the sampling and measurement of emissions and the numerical emission limits where applicable. The frequency and time of sampling of emissions is determined by the Industrial Air Pollution Inspector and the results of all tests must be made available to him. The continuous indication of particulate emissions, where practicable, from all power stations, plaster works, aluminium (secondary) works and perlite works and from new PVC polymer plant and cement works is required. Instruments for the continuous monitoring of particulate and other specified emissions are required for new chemical incineration plant and may be required on existing plant.

## 1.11 Portugal<sup>3,93-97</sup>

There are some general acts and decrees concerning air pollution but there is no legal basis for emission measurements. Particulate emission measurements are carried out manually.

## 1.12 Spain<sup>48-50</sup>

Control and measurement of emissions to the atmosphere and immissions are covered by various legislation and application decrees. Legislation for air pollution control is based on the 1972 Law and the Decree for the Protection of the Environment, which establish emission standards for several pollutants, distinguishing between different types of source. More stringent regulations apply in designated "polluted areas".

The 1975 Decree on "Characteristics, Qualities and Use Conditioning of Fuels" establishes maximum allowable sulphur contents for various types of liquid fuel.

Decree<sup>48</sup>, no 833 of 6th February 1975, authorises the competent Ministries to require the installation of instruments for the continuous and periodic evaluation of pollutant emissions from power stations, whenever it is convenient and technically and economically feasible.

Ministerial Order<sup>50</sup> No. 15029 of 25th June 1984 requires all coal fired power plant of greater than 50 MW installed capacity and all oil or natural gas fired power plant of greater than 200 MW installed capacity to possess devices for the continuous measurement and control of emissions of sulphur dioxide, particulates and nitrogen oxides. These devices should preferably be located in the stack and must be connected by data transmission facilities to the central control room. The process unit must be capable of printing the hourly and daily average emissions values and the maximum daily values, which will be reported once a month, together with the total weights of pollutants emitted and the weight of pollutants emitted per kWh of generated power. The monthly reports must be submitted to the competent authorities of the appropriate Autonomous Community, to the General Sub-Directorate of Design, Industrial Quality and Environment, to the General Sub-Directorate of Electrical Energy of the Ministry of Industry and Energy and to the General Secretariat of the Interministerial Commission of the Environment.

This Order will be enacted six months after it has been published in the Official Bulletin of the State, however the General Directorate of Energy is empowered to grant extensions to utilities who are not able to make the required installations within this time.

#### 2. MEASUREMENT METHODS

#### 2.0 Introduction

The Commission's proposed order on large combustion units gives emission concentration standards for particulates, sulphur dioxide and nitric oxide. To control these pollutants continuous measurement methods are required. Nitrogen oxides are emitted predominantly as nitric oxide and only if there is more than 5% NO<sub>2</sub> does the total NO<sub>x</sub> have to be measured.

Nearly all Member States already have some guidelines for the discontinuous measurement of pollutants emitted from combustion installations. There are also some national guidelines for continuous measurement in existance, although these are less widespread. It is not uncommon for a measurement to be in use in a country without a guideline.

There are two ways to carry out emission measurements. First there are the extractive methods which are well established but some sampling and gas handling problems have to be taken into account (see Section 2.1). Secondly there are the in-situ measurement methods, which are at various stages of development. The in-situ instruments are, however, prone to interference problems. Where possible a unified method for emission measurements, which uses either all extractive or all in-situ instruments, is preferred. All measuring devices to be used in connection with the proposed order should be approved by a competent authority.

Certain data treatment conditions are implicit in the standards making it necessary to handle the data on a dedicated computer that will calculate halfhourly and daily mean values, which can then be classified into frequency distributions. The integrity of all data should be checked before it is processed.

The present position in the Member States of EEC, with regard to the use of measurement methods and the development of guidelines for their use, has been assessed for each pollutant or quantity covered by the proposed order. The principal measurement methods, both continuous and discontinuous, which are currently in use have been described in the following sections.

#### 2.1 Sample Conditioning and Handling

Extractive emission measurement instruments require a representative, unmodified sample from which none of the components to be analysed has been lost. The procedures are different for particulate and gas analysis.

#### 2.1.1 Particulate Sampling and Handling

Both manual gravimetric methods and on-line Beta ray attenuation methods for particulate measurement use the extractive principle. Some instruments using the light scattering method are also of the extractive type. The most important principles of sampling and handling are as follows:

- the sampling location must be chosen so that a representative sample is obtained. It should be well clear of bends or other flow disturbances so that the gas flow is homogeneous, and preferably in a vertical duct or flue so that the particle distribution is not affected by gravitational and inertial forces;
- the number of sampling points in the sampling plane should be chosen so as to obtain a statistically representative result. The number of points required can be determined from a preliminary survey of the velocity and temperature profiles across the duct. In general, sampling is only carried out at one point for the on-line Beta ray attenuation and light scattering methods, therefore calibration involving measurements at a number of points in the sampling plane is necessary;
- for discontinuous methods the sampling time selected must be adequate to allow for short term fluctuations in particulate concentration;
- the sampling must be carried out under conditions closely approaching isokinetic, so that the particle size distribution is not modified. This means that the gas velocity in the duct and the sampling velocity must be measured, preferably continuously during sampling. The design of the probe is also important in obtaining an unmodified sample.

The particle separator is the most important component of a particulate sampling system and the selection of a suitable filtration medium and its location in the system are critical. The most commonly used filters are glass fibre or quartz membranes or thimbles. If chemical analysis is to be carried out on the particulate sample an appropriate grade of filter should be used. The particle separator may be located inside the stack or external to it. If it is inside the stack, care should be taken to avoid disturbance of the gas flow by the filter holder and deposition in the probe. Where the separator is outside the stack it is important to maintain the probe at a temperature higher than the relevant dew point.

#### 2.1.2 Gas Sampling and Handling

Extractive systems for flue gas sampling for gas analysis must perform the following functions.

- The sample obtained must be representative of the flue gas.
- The sample flowrate is not as critical as in particulate sampling but most instruments require a sample flow rate within a certain range and it is one of the parameters affecting the instrument response time.
- The integrity of the gas to be analysed must be maintained during the sampling and conditioning procedure.
- The conditioning required is specific to the analytical method and the cooling of the sample and the removal of particles by filtration are usual. Filtration is necessary to prevent plugging of the probe and damage to the analytical system and also because particles may interfere with the measurement results. Cooling to a gas temperature which is within the specification of the instrument is required.

2.1.2.1 Manual Methods. Sample conditioning usually involves:

- removal of particulates by filtration;

- cooling of the sample to ambient temperature with collection of the condensates for analysis if necessary.

<u>2.1.2.2 On-line Methods</u>. Sample conditioning is very important when instrumental methods are used, particularly in maintaining the integrity of the component to be analysed and achieving satisfactory operation of the instrument so that a representative measurement is obtained.

A filtration system which is capable of maintaining a steady, particle free sample flow is required. The sample flowrate should be controlled automatically.

Further conditioning requirements largely depend on whether the maximum sample temperature tolerated by the instrument is above or below the sample dew point. If it is above and there is no interference by water vapour, the sampling probe is simply equipped with a temperature regulation device which keeps the sample line at the optimum temperature for operation of the instrument. If the instrument requires the sample to be at a temperature below its dew point, moisture removal in a permeation dryer or some sort of condenser is required. Measured concentrations of the pollutant are then given under nominally dry conditions.

Additionally, for some instruments, such as flame photometers and some fluorescence and chemiluminescence analysers, dilution of the sample gas may be required to bring the pollutant concentration within the range of the instrument.

#### 2.2 Particulates

A survey of measurement methods for particulates applied in Member States and national guidelines for their use is given in Table IV.

#### 2.2.1 Discontinuous Methods

2.2.1.1 Gravimetric Method<sup>44,51-58</sup>. The gravimetric measurement method is based on the isokinetic extraction of a sample flow from the waste gas stream. The extraction is performed with a sharp-edged nozzle which is positioned in the duct facing into the moving gas stream. To allow for nonuniformity of distribution of particulate concentration in the duct, samples are taken at a pre-selected number of stated positions in the duct cross section. Outside the duct, the sample flows through a filter medium and, if necessary, a

Mamban	Discontinuous	Continuous				
Member State	Gravimetric	Photometric In Situ	Light Scattering	Beta Ray Absorption		
Belgium	NBN X 44-002	X		X		
Denmark	x					
Germany	VDI 2066 Parts 1&2	VDI 2066 Part 4 (draft)	X	X		
Greece	X					
France	AFNOR/NFX 44 052		X	AFNOR/NFX <sup>+</sup> 43017		
Ireland	X					
Italy	x	x				
Luxembourg	X					
Netherlands	NPR 2788	x				
UK	BSI 893 BSI 3405					
Portugal	X	X				
Spain	X	x	X	x		

TABLE	IV.	-	Survey	of	Methods	and	Guidelines	for	Particulate	Measurement	Applied
in Member States											

X - Method is used but there is no national guideline.
 + - Air quality method applicable to source measurement.

pre-separator. The sample flow is measured by means of calibrated gas meters in connection with flow meters or by calibrated throttle devices (eg orifices).

The particulate concentration in the sample flow is determined, based on the filter weight before and after the measurement. The mass of particulates collected in the pre-separator and the possibility of dust deposition in the sampling probe tube also have to be considered.

Heating of the sampling-line may be necessary to avoid vapour condensation.

Instruments also exist in which the filter is located inside the waste gas duct. After a time lapse for the filter to attain the temperature of the waste gas, the instrument is able to measure the particulate concentration in the waste gas at the duct temperature. Heating of the sampling-line to avoid vapour condensation is not necessary. Deposits of particulates in the sampling line upstream of the filter are small and can be neglected in many cases.

The inaccuracy of this method is about  $\pm 10\%$  to  $\pm 20\%$  under reasonably favourable conditions. In some extreme cases these conditions do not always exist and the uncertainty may be higher.

The following factors may affect the representativity of the measurement results:

- fluctuations in the gas flow;
- fluctuations in the particulate concentration;
- location of sampling plane;
- number of sampling points;
- nozzle design;
- nozzle alignment;
- departure from isokinetic sampling.

For the in-stack filter measurements the following performance data are typical:

detection limit:	2 mg								
relative detection limit:	1	5 mg/m <sup>3</sup>							
repeatability:	15	• • •	20% :	in re	espec	t of	f the	classes	3
----------------	----	-------	--------	------------------	-------	------	-------	----------------------	---
	0	•••	100 mg	g/m <sup>3</sup>	to l	00	3	00 mg/m <sup>3</sup>	3

The method is suitable for all particulate carrying waste gases from stationary sources but in some cases there is a limitation on the maximum concentration of approximately  $1 \text{ g/m}^3$ .

## 2.2.2 Continuous Methods

A list of instruments for the continuous measurement of particulates and their manufacturers is given in Annexe 1, together with performance data where available.

<u>2.2.2.1</u> In-situ Photometric Methods<sup>59</sup>. When light is passed through a gas containing suspended particles, the intensity of the transmitted light is reduced due to absorption and scatter by the particles. The ratio of the intensity of the light energy leaving the gas to that of the light energy entering the gas is known as the optical transmission, T, ie

$$T = \frac{I}{I_{o}}$$

where  $I_0$  = intensity of the light energy entering the gas; I = intensity of the light energy leaving the gas.

The logarithm of the reciprocal of the optical transmission is known as the extinction, E:

$$E = \log \left( \frac{I_o}{I} \right)$$

The Lambert law states that the extinction increases with the length of the light path and, for a given dust content, is given by:

$$E = \log \left( \frac{I_o}{I} \right) = \varepsilon 1$$

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where 1 = path length; $\varepsilon = \text{extinction coefficient}.$ 

The extinction coefficient depends on characteristics of the light beam, such as wavelength and spectral distribution and also on dust content and properties of the dust, such as particle size distribution and shape. Within certain limits, however, a linear relationship known as the Beer-Lambert law exists:

$$T = \exp(-\epsilon' c 1)$$

where c = dust content.

Thus, a linear relationship between dust content and extinction can be deduced:

$$\mathbf{E} = \mathbf{\varepsilon}' \mathbf{c} \mathbf{1}$$

This method does not give an absolute value for the solids emission rate, but may be calibrated for quantitative measurement. The relationship determined during calibration is only valid for a particular set of particle parameters and if these alter, due to a change in the mode of operation or a change in the feedstock specification, for instance, the instrument must be recalibrated.

With the exception of the simplest, low price opacimeters, all instruments are of the dual beam type. This means that the light from the source is split into two identical beams, one of which is transmitted across the stack while the other is used as a reference beam and follows a preset path in the transceiver unit. The measurement and reference beams are alternately directed to the detector. The opacity or concentration measurement is made from the balance between transmitted and reference signals. This eliminates most errors due to source intensity variations, temperature variations and long term electronic drift.

Most instruments are of the double pass type; that is a retro-reflector is placed on the opposite side of the stack to the source and detector so that the optical path of the measuring beam is twice the stack diameter. This doubles the measuring sensitivity compared with a single pass instrument. At least one instrument manufacturer uses a six fold optical passage, with a corresponding improvement in sensitivity.

Many instruments use a modulated beam to eliminate background interference and stray light. This means that the measurement (and reference) beam is interrupted at rapid regular intervals and only the alternating current component, synchronised with the interruption frequency, is amplified and displayed.

Calibration of in-situ instruments of this type has some special features compared with that of extractive instruments. Zero calibration using a clear stack or an equivalent dust free path is necessary from time to time. Span calibration can be accomplished by the insertion of neutral density filters into the light beam. Optical alignment calibration, to allow for changes caused by expansion and distortion of the stack which can affect measurements, is usually visual. The design of the optical system can minimise the effects of alignment changes.

Interference by gaseous components is usually negligible except for condensed water vapour and therefore this type of instrument is not suitable for use downstream of a wet scrubbing device unless the waste gases are reheated. Non-uniform dust deposition on the optical surfaces in the measuring and reflector heads can also affect the measurements. Air purging or heating of the window can help to prevent condensation and deposition.

A summary of the performance data for this type of instrument is given in Table V.

2.2.2.2 Light Scattering Measurement. Instruments operating on this principle can be extractive or in-situ. When a parallel aligned beam of light passes through an homogeneously dust laden gas stream, a portion of the light deviates from the axis of the beam. The intensity of the scattered light at a predetermined angle to the original beam direction is dependent on the size of the angle and the size distribution and shape of the dust particles.

Variations in the intensity of the light source and in the sensitivity of the detector are compensated for by directing an unchanged reference beam, in the opposite phase to the measuring beam, on to the photoelectric detector.

	Data from Sui	tability Testing	Data from Manufa	cturer's Literature
Performance Characteristics	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability	90% - 98%	5		<u></u>
Period of unattended operation	2-12 weeks	4		
Influence of maladjustment of light beam	2% FS for 0.1°-0.3° >2% FS for 0.5°	5	±0.5% from optical axis acceptable	1
Temperature dependence of zero [-20 -50°C]	0-2% FS/10°C	5		
Temperature dependence of sensitivity [-20 -50°C]	0-2% FS	2		
Zero drift	Not detectable up to 2% FS/3 months	5	<2% FS/3 months	1
Sensitivity drift	Not detectable up to 2% FS/3 months	4		
Reproducibility	30-100	5		
Repeatability			2% FS	2
Accuracy				

# TABLE V. - Summary of Performance Data for In-Situ Transmissometers

For low dust loadings the light received by the detector is of much lower intensity than in the case of opacity meters, so they must be capable of amplifying a small signal. However, light scatter instruments have the advantage that they do not have to differentiate between small dust content variations and errors in a large signal so they are capable of detecting extremely low concentrations.

In-situ light scatter instruments are generally sensitive to stray light and in small stacks back reflection from the walls can interfere with the readings. To prevent these effects a modulated light beam can be used. Stray light is not significant in extractive instruments.

Periodic recalibration is required; for in-situ instruments zero recalibration must be carried out in a clear stack or in a dark room and span calibration is difficult to achieve. Sophisticated instruments have built-in continuous calibration. In the case of extractive instruments optical filters are used.

Two types of instrument are used:

- One measures the light scatter directly in a flow of gaseous effluent. The emitter and receptor are mutually at an angle of 90°. The light transmitted across the gas flow under test is diffused by particles in suspension (in-situ).
- The other measures the light scatter over an angle of 15° caused by particulate in the waste gas (extractive).

<u>2.2.2.3</u> Beta Ray Absorption<sup>60</sup>. In Beta ray absorption systems a sample is extracted under near isokinetic conditions from the waste gas stream and is drawn through a filter tape. The amount of dust deposited on the filter tape is indicated by a weakening of the Beta radiation on passing through the dust laden filter.

The absorption of Beta radiation can be described by the following exponential law:

 $I = I_0 \cdot \exp(-\mu_m \cdot x);$   $I_0 = Beta radiation after absorption by the dust free filter tape;$  I = Beta radiation after absorption by the dust laden filter tape;  $\mu_m = mass absorption coefficient;$  x = surface density.

A radioactive source of suitable activity and a Geiger-Müller counter for radiation detection are required.

The absorption of each section of the filter tape is measured before and after the dust has been deposited; the differential measurement eliminates the parameters of source activity and heterogenity of the filter tape.

The accuracy of the measurement results may be adversely affected by the following:

- inhomogeneous radiation intensity;
- inhomogeneous dust loading of the filter tape;
- permeability of the filter tape;
- particulate deposits in the sampling line;
- no automatic adjustment of the sample gas flow with regard to changing waste gas velocities in the duct;
- errors in the measurement of the sample flow rate;
- temperature variations;
- pressure variations;
- variations in cell geometry.

Table VI presents a summary of the performance data for Beta ray attenuation analysers.

2.2.2.4 Obscuration Method. Instruments using this method are essentially coarse dust monitors and sensitivity to fine dust is very low. The nozzle of a sampling unit is pointed into the flue gases so that finer particles are carried around it but coarser particles (>10  $\mu$ m) enter as a result of their inertia. These particles settle on a glass window where they gradually build up until they are blown away by a blast of compressed air, at regular intervals. The changing degree of obscuration during a cycle is measured by passing a light

				1
	Data from Sui	tability Testing	Data from Mar	ufacturer's Literature
Performance Characteristic	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability	>90% (over 3 mths)	1	~80%	1
Period of unattended operation	1 wk	1		
Reproducibility	12	1		
Repeatability				
Nominal value of deviation of sample volume	<8%	1		
Zero drift	<2% FS/wk	1		
Sensitivity drift	<4% FS/wk	1		
Precision			±10% for (0.1-22 m deposited dus	g) 1 t

T ABLE VI. - Summary of Performance Data for Beta Ray Absorption Instruments

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beam, from a source outside the duct, through the collected dust and back to a photocell.

Calibration is required for quantitative measurements by means of a dust of similar physical properties and particle size distribution to the one to be measured. Also the sampling nozzle must be positioned so as to obtain a representative sample, which is difficult for large particles in large ducts.

# 2.3 Sulphur Dioxide and Oxides of Nitrogen

These pollutants are discussed in the same section because two of the most widely used continuous measurement methods are employed for both  $SO_2$  and  $NO_x$ .

A survey on methods and guidelines for the measurement of  $SO_2$  is given in Table VII whilst those for  $NO_x$  are given in Table VIII.

# 2.3.1 Discontinuous Methods for Sulphur Dioxide

These methods are suitable for reference determinations of the sulphur dioxide concentrations of exhaust gases from combustion installations. The choice of method depends on the other constituents present in the gases and the degree to which they interfere with the reactions involved in each method. With all these methods the accuracy of the results is highly dependent on the precision of the gas sample volume measurement, so the metering device must be checked regularly.

<u>2.3.1.1</u> Iodometric Thiosulphate Method<sup>61,62</sup>. The sulphur dioxide in the sample is oxidised to sulphuric acid by passing through an iodine solution of known concentration. The unreacted iodine is measured by titration with sodium thiosulphate solution. The concentration of sulphur dioxide is calculated from the amount of iodine used in the oxidation reaction.

Interference may be caused by hydrogen sulphide, nitrogen oxides and halogens. Further errors in the results may occur if the iodine solution has a temperature of more than 30°C or is exposed to strong light.

									Ŭ	ont inuous/	Autometic				
	:	Discontinu	lous/Manual					Extractiv	ve				In-si	tu	
Member State	1odo- metric thio- sulphate	titri- metric hydrogen peroxide	gravi- metric hydrogen peroxide	hydrogen peroxide- thorin <sup>1</sup>	non- disper- sive infra red	non- disper- sive ultra violet	electro- chemical	ultra violet fluor- escence	conducto- metrical	flame photo- metrical	inter- ferential	non- disper- sive infra red	non- disper- sive ultra violet	disper- sive ultra violet	2nd deriva- tive ultra- violet
Belgium			×	NBN T95-202			х			x	-				
Denmark					×							×			
Germany	VDI 2462 Part l	VDI 2462 Part 2	VDI 2462 Part 3	VDI 2462 Part 8	VDI 2462 Part 4	x			VDI 2462 Part 5				×	×	
Greece									x						
France	×				NFX 20351			43019 <sup>+</sup>	NFX 20355	NFX 43020 <sup>+</sup>	×				
Ireland	x	X		x											
Italy															
Luxembourg															
Netherlands	NEN 3104		NEN 3104	x	x	x								X	x
Х	x	X		BS 1747	x			x			•	x			
Portugal				×											
Spain				×	×			×							x

TABLE VII. - Survey on Methods and Guidelines used for SO<sub>2</sub> Measurement in Member States

X Method is used but there is no guideline. ISO method. Air quality method applicable to source measurement.

										Contin	uous/Automa	t I c			
			Disconti	nuous/Manu	al .				Extractive				In-s	ĺtu	
Member State	phenol- disul- phonic acid	acidi- metric titra- tion	sodium sali- cylate	2,6 dimethyl phenol	chromo- tropic acid	Saltzman	non disper- sive ultra violet	disper- sive ultra violet	non- disper- sive infra red	chemi- lumine- scence	inter- ferential	non- disper sive ultra violet	disper- sive ultra violet	non- disper- sive infra red	2nd deriva- tive ultra violet
Belgium			х		NBN T95-301					×					
Denmark										×					
Germany	VDI 2456 Part 1	VDI 2456 Part 2	VDI 2456 Part 8 (Draft)	×			×	×	VDI 2456 Part 3	VDI 2456 Part 7			к		
Greece										×					
France										43018 <sup>+</sup>					
Ireland															
Italy										X					
Luxembourg										×					
Netherlands			NEN 2044 (Draft)						×	×					×
¥			BS 1747			BS 1756 Part 4			X	BS 1747				x	
Portugal															
Spain	x								X						X

TABLE VIII. - Survey on Methods and Guidelines used for  $NO_X$  Measurement in Member States

X Method is used but there is no guideline. + Air quality method applicable for source measurement.

## Performance Characteristics

Uncertainty range (95% probability):	±0.04	g	$SO_2/m^3$
Relative detection limit (99% probability):	0.06	g	$SO_{2}/m^{3}$

2.3.1.2 Titrimetric Hydrogen Peroxide Method<sup>63</sup>. A metered sample of the waste gas is passed through a hydrogen peroxide solution, where any sulphur dioxide present is oxidised to sulphuric acid. The quantity of sulphuric acid is measured by titration with caustic soda.

Interference occurs if any other component is present which can react to form an acid or a base with the absorbing solution. In such cases the sulphur dioxide content should be measured gravimetrically (see chapter 2.3.1.3), as long as the gas sample does not contain sulphur components other than sulphur dioxide and sulphur trioxide.

#### Performance Characteristics

Uncertainty range (95% probability):	±0.05 g SO <sub>2</sub> /m <sup>3</sup>
Relative detection limit (99% probability):	0.03 g $SO_{0}/m^{3}$

<u>2.3.1.3</u> Gravimetric Hydrogen Peroxide Method<sup>64,65</sup>. A metered sample of the waste gas is passed through a hydrogen peroxide solution, where any sulphur dioxide present is oxidised to sulphuric acid. A soluble barium salt is added to precipitate barium sulphate which is measured gravimetrically.

Any sulphur-containing compound will cause interference. If sulphur trioxide is the only interfering component the method can be used once the sulphur trioxide has been eliminated.

#### **Performance Characteristics**

Uncertainty range (95% probability):	$\pm 0.06 \text{ g } \text{SO}_2/\text{m}^3$
Relative detection limit (99% probability):	0.04 g $SO_{0}/m^{3}$

<u>2.3.1.4 Hydrogen Peroxide-Thorin Method</u><sup>66-69</sup>. The sulphur dioxide containing waste gas is filtered and passed through a hydrogen peroxide solution, where sulphur dioxide is oxidised to sulphuric acid. After sampling,

the solution is titrated against a barium perchlorate solution using thorin as the indicator.

Sulphur trioxide and volatile sulphate salts may interfere as they can pass through the quartz wool filter and remain in the absorption solution.

Furthermore, anions forming insoluble salts with barium ions at pH 4.6 may interfere. Volatile salts of multivalent metal cations interfere if the cations react with the thorin indicator.

Generally, with correct application of the procedure, these multivalent metal cations are not detectable in the sample solution. In exceptional cases, eg with extremely high waste gas temperatures or high metal concentrations (salts or metal vapours), the applicability of the method has to be tested.

This method is particularly suitable for measuring low sulphur dioxide concentrations in the absence of other sulphur containing compounds, except for sulphur trioxide. It can also be applied where nitrogen dioxide concentrations are too high to permit the use of the iodometric thiosulphate method (section 2.3.1.1).

#### **Performance Characteristics**

Standard deviation occurs between  $\pm 4.4 \text{ mg/m}^3$  and  $74 \text{ mg/m}^3$  with sulphur dioxide concentrations between 16 mg/m<sup>3</sup> and 2000 mg/m<sup>3</sup>.

Detection limit	0.065	mg	so <sub>2</sub>
Relative detection limit	0.72	mg	SO <sub>2</sub> /m <sup>3</sup>

## 2.3.2 Discontinuous Methods for Oxides of Nitrogen

These methods are suitable for reference determinations of the nitrogen oxide content of waste gases from combustion installations. The choice of method depends on the interfering components present in the gas and on the concentration ranges of nitrogen oxides likely to be encountered.

2.3.2.1 Phenoldisulphonic Acid Method<sup>70</sup>. Nitrogen monoxide and nitrogen dioxide in the gas sample are oxidised quantitatively in a sulphuric acid/

hydrogen peroxide solution to nitric acid. There must be sufficient peroxide to oxidise any SO<sub>2</sub> present as well. The nitric acid is reacted with phenoldisulphonic acid to form a yellow reaction product which is measured photometrically. The concentration of both nitrogen oxides is given as nitrogen dioxide.

The method described measures all nitrogen oxides with the exception of dinitrogen oxide. There are no quantitative data available on interference but nitrates, nitrites and certain nitrogen-containing organic compounds appearing as vapours or suspended matter in the sample can cause considerable positive interference. Sulphur dioxide and other oxidisable substances in higher concentrations affect the quantitative oxidation of nitrogen monoxide. The inference caused by halogens, sulphur and lead compounds is small.

## Performance Characteristics

Standard deviation:	±5% for concentrations around 4.9 g NO <sub>2</sub> /m <sup>3</sup>
Detection limit:	20 μg NO <sub>2</sub>
Relative detection limit (lower limit of measurement range):	0.04 g NO <sub>2</sub> /m <sup>3</sup>

Upper limit of measurement range: About 5.0 g  $NO_2/m^3$ 

<u>2.3.2.2 Acidimetric Titration Method</u><sup>71</sup>. Nitrogen monoxide and nitrogen dioxide are forced out of the gas sample by oxidising them to nitric acid with a solution of hydrogen peroxide containing a foaming agent. The nitric acid is measured by acidimetric titration.

The method can be used for total nitrogen oxides concentrations between 0.05% and 1.0% v/v. The gas sample must have a free oxygen content of at least 1%.

All components of the gas sample which are absorbed by the hydrogen peroxide solution and which can be titrated acidimetrically, will interfere with the measurement result.

### Performance Characteristics

Standard deviation at a total nitrogen oxides content of approximately 3% v/v:

- using test gases: ±1.4%
- measuring in the waste gases of nitric acid plants:  $\pm 2.0\%$

2.3.2.3 Sodium Salicylate Method<sup>67,72,73</sup>. The nitrogen monoxide and nitrogen dioxide components of the gas sample are quantitatively oxidised to nitric acid using a sulphuric acid/hydrogen peroxide solution. The mixture is then treated with sodium salicylate in a medium of concentrated sulphuric acid, so that the nitric acid reacts to form nitro salicylic acid. When a sodium hydroxide - Seignette salt solution is added the yellow anion of nitrosalicylic acid is formed. Its concentration can be measured photometrically and is proportional to the concentration of nitrogen oxides, excluding dinitrogen oxide.

Various side reactions may cause interference problems, depending on the composition of the gas sample. The errors are usually minimal providing the analysis is commenced within twenty minutes of the sample being extracted. Nitrates, nitrites and some nitrogen-containing organic compounds present as vapours or suspended matter can systematically interfere with the measurement results to give spuriously high values. Sulphur dioxide and other oxidisable substances may affect the quantitative oxidation of nitrogen monoxide.

#### Performance Characteristics

Standard deviation:	$\pm 2.0\%$ to $\pm 4.4\%$ in the concentration
	range between 100 and 710 ppm $v/v$
Detection limit:	1.8 $\mu$ g (NO + NO <sub>2</sub> ), given as NO <sub>2</sub>
Relative detection limit:	2 mg/m <sup>3</sup> (NO + NO <sub>2</sub> ), given as NO <sub>2</sub>

<u>2.3.2.4</u> 2,6-dimethylphenol Method<sup>74</sup>. The gas sample is introduced into a gas holder, together with an ozone-oxygen mixture and water. Within five minutes the nitrogen oxides are quantitatively converted to nitric acid. Excess ozone is destroyed by sodium sulphite solution. A sulphuric acid-phosphoric acid-mixture and a 2,6-dimethylphenol solution are added to part of the sample and 4-nitro-2,6 dimethylphenol is formed as a reaction product. It is extracted with dichloromethane and extracted back in sodium hydroxide. The extinction of the yellow coloured solution is measured photometrically at a wave length of 430 nm and is proportional to the nitrogen oxide concentration. No data are available on interference at present.

## Performance Characteristics

Relative detection	1 limit:	<0.03 g	NO <sub>2</sub> /m <sup>3</sup>	3		
Standard deviation	1:	±1.7 to	±2.1%	in	the	measurement
		range 55	50 to 6	570	ррш	

<u>2.3.2.5</u> Chromotropic Acid Method<sup>75</sup>. The gas sample is fed into an oxidising absorbent containing hydrogen peroxide and dilute sulphuric acid. The nitrogen oxides present are converted to nitric acid. The excess hydrogen peroxide is destroyed by the addition of sodium sulphite. The nitrate ions are reacted with chromotropic acid to form a yellow product. The intensity of the yellow colour, which is proportional to the concentration of the nitrogen oxides in the gas sample, is measured photometrically.

This method measures both nitrogen monoxide and nitrogen dioxide. Inorganic nitrates and nitrites and nitrogen containing organic compounds can interfere with the measurement. Certain oxidisable components such as sulphur dioxide can react with part of the hydrogen peroxide, thus affecting the quantitative oxidation of the nitrogen oxides.

No performance data for this method are available.

2.3.2.6 Saltzman Method<sup>76</sup>. The Saltzman method is based on the quantitative diazotization of sulphanilic acid by the nitrate ion, followed by coupling of this derivative with N-(1-naphthyl)-ethylenediamine to form a red coloured azo dye. The sample of flue gas is withdrawn into a syringe of fresh Saltzman solution which contains the aforementioned reagents. The syringe is left for at least three hours, or preferably overnight, for the colour to develop. The concentration of nitrite ions may then be determined by measuring

the absorbance of the solution at 550 nm against a reagent blank. The total oxides of nitrogen concentration, expressed as NO can be obtained from a calibration curve.

Inorganic nitrites can cause positive interference and sulphur dioxide may interfere by acting as a bleach.

#### **Performance** Characteristics

For nitrite ion concentrations  $50-1500 \text{ ml/m}^3 - \text{sd} < 2.5\%$ .

## 2.3.3 Continuous Methods for Sulphur Dioxide and Oxides of Nitrogen

A list of instruments for the continuous measurement of  $SO_2$  and  $NO_x$ , and their manufacturers is given in Annexe 1 together with performance data where available.

<u>2.3.3.1</u> Non-dispersive Spectrometry Methods<sup>77-80</sup>. Many gaseous pollutants absorb light energy in one or more regions of the spectrum. Sulphur dioxide and nitrogen monoxide absorb infra-red radiation and ultraviolet radiation. Each type of pollutant molecule will absorb light at a characteristic wavelength so that it can be distinguished from other absorbing components.

Continuous emission monitors using this method apply the Beer-Lambert law, which states that the transmittance of light (ie the ratio of the intensities of the transmitted and incident light) through a medium which absorbs it, is decreased exponentially by the product  $\alpha$ cl. This is expressed mathematically as

$$T = I/I_0 = e^{-\alpha c I}$$

where T = transmittance;

I = intensity of transmitted light;

I<sub>0</sub> = intensity of incident light;

c = concentration of pollutant;

 $\alpha$  = attenuation coefficient;

1 = path length.

l is a parameter of the instrument and  $\alpha$  is determined during calibration so the concentration of the measured component is proportional to the transmittance.

Non-dispersive instruments obtain the desired selectivity by incorporating a sample of the measured component into the instrument itself, as opposed to selecting the required spectral band by optical techniques. Most NDIR instruments use the detector for the storage of the gas. Many different types of gas filled detector have been developed in order to minimise interference, as shown in Fig. 1. Other NDIR instruments use solid state detectors in conjunction with the gas filter correlation method. In this type, the sample is contained in a gas cell located in a filter wheel. The filter wheel also has an opening or another gas cell filled with a non-absorbing gas which is brought into the path of the light beam alternately with the sample gas Some photometers use a combination of the two methods. NDUV instruments cell. store the sample in the light source itself; a gas filled discharge lamp is used to emit spectral curves characteristic of the measured component. A11 NDUV instruments use a photomultiplier tube as a detector.

Non-dispersive instruments may be extractive or in-situ; the principles are the same but each type has special features which are described below.

#### Extractive

After conditioning the gas sample passes through the sample cell. A beam of light from the source is directed through the sample cell and registered by a detector on the opposite side. Selectivity is obtained using the techniques discussed above. It is necessary to minimise the errors caused by small variations in source intensity and detector sensitivity and by interference. A periodic zero point correction can be used to compensate for the first two but the use of a reference standard is more usual. There are three basic strategies for incorporating such a standard:

- the use of a reference cell containing a non-absorbing gas, so that the intensity of a light beam passing through it will be unchanged, providing a reference signal for comparison with the measurement signal; the use of a dual wavelength system - the beam of the reference wavelength passes through the sample cell unchanged so providing a reference signal;

- if a filter wheel of the type already described is used, the beam can subsequently be split, one part passing through the sample cell and then to a detector and the other passing directly to another detector. As the parent beam is being alternately preabsorbed by the gas cell in the filter wheel and left unmodulated, four signals are obtained. They are related to the pollutant concentration by a logarithmic rule.

Gas conditioning can include the conversion of nitrogen dioxide to nitrogen monoxide, if the total nitrogen oxides concentration is required. A thermal or a catalytic converter is used for this purpose. Carbon monoxide, unburned hydrocarbons and ammonia can all cause interference by undergoing side reactions in the converter. These effects can be minimised by the choice of material of construction and operating temperature.

Summaries of the performance data for extractive NDIR and NDUV instruments for measuring  $SO_2$  and  $NO_{v}$  are given in Table IX and X respectively.

## In Situ

In situ instruments do not require a gas sampling and conditioning system, which is an advantage, however, since nitrogen dioxide cannot be converted to nitrogen monoxide such instruments generally cannot measure nitrogen dioxide concentrations.

The absorption measurement path is actually in the stack or duct; the photometer is located on one side of the stack and the source, or a retroreflector if the instrument is dual pass, is located on the opposite side. Obviously for dual pass instruments the source will be on the same side as the photometer. A reference standard is incorporated by the use of a dual wavelength system in the same way as for extractive instruments.

Sensitivity is usually obtained using the gas filter correlation method, either by the incorporation of a filter wheel, as already described, or by splitting the beam into two and passing one part through a gas cell and the

	Data from Suit	ability Testing	Data from Manuf	acturer's Literature
Performance Characteristics	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability	95-100% (over 3 mths)	4		
Period of unattended operation	1-2 weeks	Q		
Reproducibility	30-257	Q		
Influence of barometric pressure variation	Undetectable -1% FS/10 m bar	Q		
Influence of sample flowrate variation	Undetectable -0.5% FS/10 1 h <sup>-1</sup>	ę		
Lower detection limit	1.3-2% FS	6		
Temperature dependence of zero	Undetectable -2% FS/10°C	5		
Temperature dependence of sensitivity	Undetectable -2% FS/10°C	5		
Zero drift	0.5-2% FS/8 day	6	1-3.5% FS/wk	4
Sensitivity drift	1-4% FS/wk	Q	2% FS/wk -1% FS/da	y 4
Response time	75-200 secs (90%)	Q	Not on comparable basis	
Interference	0.5-7.5% FS	9		
Repeatability			0.5% FS	I

TABLE IX. - Summary of Performance Data for Extractive NDIR Instruments Measuring SO<sub>2</sub> and/or NO<sub>X</sub>

	Data from Sui	tability Testing	Data from Manufac	turer's Literature
Performance Characteristics	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability	90-100%			
Period of unattended operation	l week			
Reproducibility	43-78			
Repeatability			1% FS	1
Influence of barometric pressure variation	1% FS/1% change in pressure		1% FS/1% change in pressure	1
Influence of sample flowrate variation	<1% FS/20 1/h			
Lower detection limit	0.02 g NO <sub>2</sub> /m <sup>3</sup>			
Temperature dependence of zero	1.5% FS/10°C			
Temperature dependence of sensitivity	3% FS/10°C		1.5% FS/10°C	I
Zero drift	<1% FS/week	2	% FS/wk-0.5% FS/day	m
Sensitivity drift	<3% FS/week		2% FS/wk	1
Response time	34-60 secs		0.5 s (90%)	1
Interference	<0.5% FS 2.5% FS 150 mg NO2/m <sup>3</sup>			

TABLE X. - Summary of Performance Data for Extractive NDUV Instruments Measuring SO<sub>2</sub> and/or NO<sub>X</sub>

other through a neutral density filter. The gas filter wheel method can cause signal variability if the frequency of interposition of the filters is low and there are changes in the dust burden. Gas cell leaks can cause span drift but this is overcome by using two gas cells.

There are particular difficulties associated with the design of the optical system which are not encountered with extractive instruments. Most of these stem from the long pathlength involved. The high intensity source requires a temperature of as much as 800-1000°C and this can result in problems with visible light emissions. The high signal attenuation across the long pathlength means that a very sensitive detector is needed. Accurate optical alignment on commissioning and alignment recalibration to compensate for the effects of stack expansion, distortion and vibration are required. The use of a gas cell which totally absorbs the radiation in the absorption band of the analyte (autocollimation) helps to overcome alignment problems.

Cross-sensitivity is difficult to assess over the long pathlengths used. It can be avoided to some extent by measuring in the UV/visible region so that CO,  $CO_2$  and other gases have no influence on the measurements.

Summaries of the performance data for in-situ NDIR and NDUV instruments measuring  $SO_2$  and  $NO_2$  are given in Tables XI and XII respectively.

2.3.3.2 Dispersive Ultra Violet Photometry Methods. The phenomenon of ultra-violet light absorption by heterogeneous molecules is also used for the measurement of nitrogen monoxide and sulphur dioxide concentrations by means of dispersive ultraviolet photometry. There are extractive instruments similar to those using non-dispersive spectrometry and at least one in-situ instrument. In this particular device, the absorption measurement path is in a measurement probe in the waste gas duct. The measurement probe is equipped with wing flaps, which are open during the measurement so that the waste gas passes through the probe. The ultraviolet beam generated by the source passes down the length of the probe and back and then strikes a highly dispersive prism. The prism splits up the UV beam and the specific wave length is directed to a photomultiplier.

For the automatic checking of zero point and reference point the wing flaps on the measurement tube are closed so that it can be filled with calibration gases.

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Performance Characteristics	DALA IION S	Suitability Testing	Data Irom Manu	TACHNICI & PTICIALNIC
	Range	Number of Instruments Considered for Range	Range	Number of Instrument Considered for Range
Availability				
Period of unattended operation				
Reproducibility				
Repeatability			1-2% FS	2
Lower detection limit				
Temperature dependence of zero				
Temperature dependence of sensitivity				
Zero drift				
Sensitivity drift				
Influence of maladjustment of light beam				
Response time		20	<b>30 ms - 3 secs (90</b>	<b>%</b> ) 2
Interference				
Stack temp			200-600°C max	£
Accuracy			±3-5% FS	£

TABLE XI. - Summary of Performance Data for In-situ NDIR Instruments Measuring  $\mathrm{SO}_{2}$  and/or  $\mathrm{NO}_{\mathrm{X}}$ 

	Data from Su	itability Testing	Data from Manu	facturer's Literature
Performance Characteristics	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability	>95% (over 3 mths)	1		
Period of unattended operation	l week	1		
Reproducibility	61-167	1		
Repeatability				
Lower detection limit	<2% FS	1		
Temperature dependence of zero	<1% FS/10°C	I		
Temperature dependence of sensitivity		,		
Zero drift	Not detectable	1	2% FS/day	1
Sensitivity drift	<2% FS/8-12 wks	1	2% FS/day	1
Influence of maladjustment of light beam	>2% FS in angle Range up to 0.5°	1		
Response time	2-12 secs (90%)	1	>5 secs	1
Interference	CO <sub>2</sub> 14.3 vol% CO 1000 mg/Nm <sup>3</sup> NO 1340 mg/Nm <sup>3</sup> NO <sub>2</sub> 923 mg/Nm <sup>3</sup> H <sub>2</sub> O saturated 300 k Dust 250 mg/Nm <sup>3</sup>	) (0.5% 1 () FS		
Stack temp			316°C max	1
Accuracy				

TABLE XII. - Summary of Performance Data for In-situ NDUV Instruments Measuring  $\mathrm{SO}_2$  and/or  $\mathrm{NO}_{\mathbf{X}}$ 

There are no data on interference available at present.

2.3.3.3 Second Derivative Ultra Violet Photometry Method. Instead of using direct absorption spectroscopy as in the methods described in 2.3.3.1 and 2.3.3.2, this method detects and measures the curvature or second derivative of the narrow characteristic absorption band of  $SO_2$  or NO. As in the previous method the absorption measurement path is in a measurement cavity at the end of a probe located in the waste gas duct. Ultra violet light from the source, located in the transceiver, is projected through the measurement cavity and returned to the transceiver by a retro-reflector. A tuned monochromator separates the light into discrete spectral bands and those corresponding to  $SO_2$  and NO can be directed onto the detector.

The wavelength of these two monochromatic beams is modulated with respect to time by a rotating quartz window scanner, before they reach the detector. This allows the second derivative signal to be measured directly. Sequential time separation of the  $SO_2$  and NO signals is accomplished by a small shutter in the monochromator.

There are no quantitative data on interference but in theory UV absorbing interferents should not affect the second derivative.

#### Measurement Devices

Performance data are only available for one instrument and are given below (from manufacturer's literature);

ranges	0- 250 ppm SO <sub>2</sub> , NO (min)
	0-6000 ppm $SO_2$ , NO (max)
zero drift	~0.5% FS/day
calibration drift	<2.5% FS/day
response time	<4 mins
ambient temp range	-30 -140°F
stack temp range	75–800°F

2.3.3.4 Interferential Method. If a beam from a light source is divided into two by an interference filter and the two partial beams are polarised, then

passed through a quartz slice to superimpose them, interference fringes will be obtained. The intensity, I of the fringes in the absence of gas, is given by

$$I = K (A + B \sin wt)$$

where K, A, B = constants

w = angular velocity of a rotating analyser downstream of the interference field

If the beam from the source is first passed through a measuring cell, through which a sample gas containing  $SO_2$ , NO or  $NO_2$  flows, the luminous flux at the output of the cell will have a periodic form, due to the structure of these molecules. The luminous flux registered by a receiver after the interference field will therefore be the product of two periodic functions. The intensity of the flux will now depend on two terms, one which is dependent only on source intensity and absorption in the optical system and is therefore constant, and one which is a function of the absorption of the sample gas and hence of  $SO_2$ , NO or  $NO_2$  concentration.

#### Measurement Devices

The manufacturers give the following performance data for the only instrument on which data are available

ranges 0- 500 ppm min 0-10,000 ppm max

response time 3 secs ambient temp range 10-40°C

#### 2.3.4 Continuous Measurement Methods for SO<sub>2</sub> Only

A list of instruments for the continuous measurement of SO<sub>2</sub> and their manufacturers is given in Annexe 1, together with performance data where available.

2.3.4.1 <u>Conductometry</u><sup>81,82</sup>. In the conductometric measurement method the sample gas is introduced into a suitable liquid reagent and the change of the conductivity after completion of the reaction of the liquid with the gas is measured. In continuous conductometry the sample gas and the reagent liquid are continuously delivered to the reaction chamber. Suitable means must be provided to ensure that the flow of both streams is kept constant. The difference in the conductivity measurements before and after the introduction of the sample gas is proportional to the concentration of the measured component. As the conductivity is highly dependent on temperature, its influence must be compensated for.

To measure the concentration of sulphur dioxide the sample gas is brought into contact with a hydrogen peroxide solution. Thus, the sulphur dioxide contained in the sample gas is absorbed and oxidised to sulphuric acid. The change in conductivity caused by this reaction is a measure of the sulphur dioxide concentration in the sample gas.

Instruments with a measurement range for sulphur dioxide up to 5 g/m<sup>3</sup> suffer from no detectable interference by carbon monoxide, carbon dioxide, water vapour (at saturated temperatures up to 40°C), hydrogen halide and hydrogen sulphide. Interference by sulphur trioxide up to a concentration of 40 mg/m<sup>3</sup> was tested for at combustion installations and was not detectable. There is no information available concerning interference by other components.

#### Measurement Devices

Performance data from suitability testing are available for one instrument of this type, as shown below.

Availability	>95% (over 3 months)
Period of unattended operation	1 week
Reproducibility	No data available
Lower detection limit	<2% FS
Temperature dependence of zero	Not detectable
Temperature dependence of sensitivity	<1% FS/10°C
Zero drift	<0.5% FS/week
Sensitivity drift	<0.5% FS/week

Response time	<3 min
Interference	No data available

<u>2.3.4.2 Flame Photometry</u><sup>83</sup>. Sulphur containing molecules can be excited to luminescence by introduction into a hydrogen flame. A small fraction of them is converted to an  $S_2$  species which reacts with hydrogen and hydroxyl radicals, also formed in the flame, to form an excited  $S_2^*$  species, as shown below.

 $H + H + S_2 \rightarrow S_2^* + H_2$ OH + H + S\_2  $\rightarrow S_2^* + H_2$ O

As the S<sub>2</sub>\* reverts to its ground state it emits radiation in the UV region, a selected band of which can be detected by a photomultiplier tube. The chemiluminescent emission activity is proportional to the square of the flame sulphur atom concentration.

In order to measure only  $SO_2$  and not all sulphur compounds present the other sulphur bearing molecules must be removed before the gas is introduced into the flame. The most common,  $H_2S$ , is usually removed in a scrubber. Interference can also be avoided by the use of a narrow band interference filter and by the geometrical arrangement of the burner which optically shields the photomultiplier tube from the primary flame.

Usually the sample gas must be diluted to around 1 ppm  $SO_2$  if this method is to be used. The maximum range for flame photometers is 0-10 ppm  $SO_2$ .

#### Measurement Devices

The performance characteristics, as reported by the manufacturers, are available for one instrument only and are given below.

Ranges0-1 ppm, 0-0.5 ppm SO2Precision2% FSResponse time<35 s (90%)</td>Ambient temp range10-100°F

From the literature it appears that instruments of this type have a zero and sensitivity drift of about 0.2%/3 days.

<u>2.3.4.3</u> Fluorescence<sup>84</sup>. Fluorescence is a photoluminescent process in which light energy of a given wavelength, usually in the near ultra violet region, is used to excite molecules, which then emit light energy of a longer wavelength as they return to their ground state. The molecules remain in their excited state for 10-100 seconds, during which time they lose some of their energy in vibrational and rotational motion before losing the remainder as light.

Quenching of the radiation by the collision of excited molecules with other molecules causes interference. Each species has a different quenching efficiency so that the degree of quenching depends on the gas composition.

A continuous flow of the sample gas is passed through a temperature controlled reaction chamber designed to minimise stray light. A beam of ultra violet light from a suitable source is passed through a narrow bandpass filter and then through the reaction chamber. A beam of fluoresced light produced by the reaction is passed through a second narrow bandpass filter and then registered by a photomultiplier tube.

Sample dilution is required for source monitoring applications with some instruments.

Table XIII gives a summary of the performance data for fluorescence analysers.

2.3.4.4 Electrochemical Method. A probe with a solid state potassium sulphate electrolyte measuring cell is used in conjunction with a zirconium oxide probe for measuring the oxygen concentration. The EMF across the potassium sulphate cell is generated from the ratio of the potassium atom activities at the electrodes on each side of the electrolyte.

	Data from Suit	ability Testing	Data from Manufa	cturer's Literature
Performance Characteristics	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability				
Reproducibility				
Repeatability				
Lower detection limit			0.001-0.002 ppm	2
Zero drift			0.005 ppm/wk - 0.003 ppm/day	2
Sensitivity drift			2% FS/wk	1
Response time			1.5 - 3 mins (95%)	2
Interference			0.005-0.02 ppm total	2

TABLE XIII. - Summary of Performance Data for Fluorescence Analysers for SO2

The output voltage,  $E_1$ , from the potassium sulphate cell follows the Nernst equation:

$$E_{1} = \frac{RT}{2F} \qquad \frac{P''SO_{2}}{P'SO_{2}} + \ln \frac{P''O_{2}}{P'O_{2}} + C_{1}$$

where

where F = Faraday's constant;

If the partial pressures  $P''_{SO_2}$  and  $P''_{O_2}$  at the reference electrodes are held constant by passing suitable reference gases over the electrodes and the cell temperature is kept constant, the output of the potassium sulphate cell is a function of the partial pressures of oxygen and SO<sub>2</sub> in the flue gas.

The oxygen partial pressure is measured by the zirconium oxide sensor, whose output voltage  $E_2$  is given by:

$$E_2 = \frac{RT}{4F} \quad \ln \frac{P''O_2}{P'O_2} + C_2$$

An electronic package associated with the  $SO_2$  probe accepts  $E_1$  from the potassium sulphate cell and  $E_2$  from the zirconium oxide cell and produces voltage,  $E_3$ , which is a function of the  $SO_2$  partial pressure only.

$$E_{3} = E_{1} - 2 E_{2}$$

$$= \frac{RT}{2F} \ln \frac{P''SO_{2}}{P'SO_{2}} + C_{3}$$

#### Measurement Devices

The performance data specified by one manufacturer of this type of instrument are as follows:

Ranges	10-1000 ppm SO <sub>2</sub> min
	0.25-25% SO <sub>2</sub> max
	0.1-10% 0 <sub>2</sub> min
	0.25-25% 0 <sub>2</sub> max
Precision	±2% of 0 <sub>2</sub> reading
Zero drift	<2% FS/day
Sensitivity drift	<2.5% FS/day
Response time	<3 secs (95%)
Ambient temp range	5–49°C
Stack temp range	20-760°C

## 2.3.5 Continuous Measurement Methods for $NO_x$ Only

A list of instruments for the continuous measurement of  $NO_X$  and their manufacturers is given in Annexe 1, together with performance data where available.

2.3.5.1 Chemiluminescence<sup>67,85,86</sup>. The chemiluminescent method of measuring nitrogen monoxide and nitrogen dioxide is based on the reaction between NO and ozone to give NO<sub>2</sub>. About 10% of the NO<sub>2</sub> produced is in an electronically excited state and a small proportion of these molecules emit light in the infra red region as they revert to their normal state. The intensity of this emission is proportional to the mass flowrate of NO<sub>2</sub>.

The reaction takes place in a thermostatically temperature controlled chamber operating a constant internal pressure, through which the sample and ozone rich air flow continuously. The air has passed through an ozoniser where partial conversion of the oxygen has been accomplished by a high energy UV lamp or a high voltage corona discharge. An ozone filter is usually fitted at the outlet of the chamber to prevent ozone leakage to the surroundings. A narrow band of the chemiluminescence is selected by an optical filter and measured using a photomultiplier tube. If NO and NO<sub>2</sub> are to be measured the NO<sub>2</sub> present must be reduced to NO in a thermal or catalytic converter prior to the reaction chamber. The NO and NO<sub>2</sub> components can be monitored separately by sequential analysis in a single reaction chamber or by using a dual reaction chamber.

As with the fluorescence method quenching of the radiation by other molecules can be a problem. Also, when thermal or catalytic converters are used higher concentrations of carbon monoxide and unburned hydrocarbons can reduce nitrogen oxides to nitrogen, decreasing the measurement results. The presence of ammonia in the waste gas can cause positive interference as it may be converted to NO in the converter. This effect can be minimised by using a relatively low operating temperature, not more than 400°C, and constructing the converter in carbon or molybdenum.

A summary of the performance data for chemiluminescence analysers is given in Table XIV.

# 2.4 Oxygen/Carbon Dioxide

# 2.4.1 Discontinuous Methods

<u>2.4.1.1</u> Orsat Apparatus<sup>87</sup>. A sample of the waste gas is brought into contact with a caustic potash solution which adsorbs the carbon dioxide. The change in volume is a measure of the volume concentration of carbon dioxide in the dry waste gas. It has to be stressed that sulphur dioxide is also adsorbed in the caustic potash solution.

The measurement of the oxygen content is performed in a similar way using pyrogallic acid as the adsorbent. This measurement is usually done with the same gas sample after measuring carbon dioxide in the caustic potash solution as carbon dioxide is also adsorbed in pyrogallic acid.

## 2.4.2 Continuous Methods

A list of instruments for the continuous measurement of oxygen and their manufacturers is given in Annexe 1. NDIR instruments for measuring  $CO_2$  are produced by many of the same manufacturers as NDIR instruments for  $SO_2$  and  $NO_x$ .

	Data from Suit	cability Testing	Data from Manufa	cturer's Literature
Performance Characteristics	Range	Number of Instruments Considered for Range	Range	Number of Instruments Considered for Range
Availability	>95% (over 6 months)	1		
Reproducibility	32-100	1		
Repeatability		1	1% FS	2
Temperature dependence of zero	1.8% FS/10°C	1	1 ppm/5-15°C	1
Temperature dependence of sensitivity	<3% FS/10°C	1		
Zero drift	1.6% FS/wk	. <b>1</b>	0.005 ppm/day - 1% FS/day	£
Sensitivity drift	<12% FS/wk	1	0.02 ppm/day - 1% FS/day	£
Minimum detection limit	5 ppm NO	1	0.01-0.05 ppm	2
Precision		1	0.5% FS	1
Response time	30-40 secs	1	1.5 s - 1 min (95%)	4
Interference	$\begin{array}{cccccc} C0_2 & 10 & vol ~ \% \\ H_2 0 & 0.1 & vol ~ \% \\ S0_2 & 0.5 & vol ~ \% \\ C0^2 & 0.2 & vol ~ \% \\ HCl & 0.28 & vol ~ \% \\ NH_3 & 0.2 & vol ~ \% \end{array}$	FS 1	Interference equivalent 0.04 ppm	I

TABLE XIV. - Summary of Performance Data for Chemiluminescence Analysers

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- 56 -

2.4.2.1 Potentiometric Method. This method works on the principle that electron flow will occur between solutions or gases containing different concentrations of a chemical species, in the presence of an electrolyte and an electrical circuit, because of the difference in chemical potential. The resulting electromotive force is expressed by the Nernst equation:

 $EMF = (RT/4F) \ln (C_1/C_2)$ 

where R = gas constant;

- T = absolute temperature;
- F = Faraday's constant;
- C = concentration

A Zirconium oxide cell, in which the zirconium oxide acts as the electrolyte and as a high temperature catalyst producing oxygen ions, is used for oxygen measurement. The cell can be mounted on a probe which is inserted directly into the stack or duct or used in an extractive instrument. One side of the cell is coated with a thin layer of platinum which forms the flue gas side electrode. The reference electrode, which is in contact with an oxygen containing reference gas, usually air, may be in the form of a wire, a tube or a platinum coating. When the cell is maintained at a temperature in excess of 600°C oxygen molecules on the reference side pick up electrons at the electrodeelectrolyte interface. Zirconia has a high conductivity for oxygen ions because of vacancies in the crystal lattice. At high temperatures the vacancies and oxygen ions move about; the oxygen ions move to the flue gas side of the cell where they release electrons to the electrode and emerge as oxygen molecules.

The generated EMF can be measured and if the cell temperature is stable and the reference oxygen partial pressure is known the flue gas oxygen content can easily be obtained.

Facilities for the introduction of zero and span gases are required for the in-situ calibration of the cell and the results are given under wet conditions.

2.4.2.2 Paramagnetic/Thermomagnetic Methods. Oxygen can be distinguished from the other common gases by the marked paramagnetic properties of its molecules, which have a relatively strong permanent magnetic moment. In an inhomogeneous magnetic field oxygen molecules will be drawn in the direction of increasing field strength.

Various extractive type instruments use this property as a measurement principle in the following ways:

- A properly combined magnetic gradient and thermal gradient can be used to induce and sustain a flow of paramagnetic gas in a closed system. If no other paramagnetic gas is present the increased flow intensity or "magnetic wind" will be proportional to the concentration of oxygen in the sample.
- If two gases of different oxygen content meet in a magnetic field a pressure difference exists between them because of their different magnetic susceptibility. A reference gas  $(CO_2, N_2, O_2 \text{ or air})$  can be drawn into an instrument and split into two equal streams. If the flow restrictions in the two conduits are balanced and one of the streams is contacted with the sample gas within a magnetic field, the resulting pressure difference between the two streams will cause a flow in the channel connecting the two conduits. This flow is proportional to the oxygen content of the sample and can be transduced to an electric signal by a microflow sensor.
- Oxygen molecules in an inhomogeneous magnetic field will build up a partial pressure gradient which will exert a force on a small nonmagnetic hollow dumbbell-shaped body located within the field. The resulting torque can be balanced by a compensating electromagnetic torque so that the body remains in place. The balancing current required is proportional to the oxygen content.

2.4.2.3 Non-dispersive Infra-red Extractive Photometry. Based on the interaction of electromagnetic radiation in the optical spectral range with the molecules of gas (see Chapter 2.3.3.1) the concentration of carbon dioxide can be measured by means of its characteristic absorption spectrum in the infra-red range. Usually, non dispersive infra-red measurement devices with carbon dioxide stored in the receiver chamber are employed.

2.4.2.4 Polarographic Method. There is at least one extractive type instrument available which uses a polarographic sensor to measure oxygen content.

The sensor consists of two electrodes; a reference electrode and a sensing electrode, which is separated from the sample gas by a thin film of electrolyte and a selective semipermeable membrane. By using different electrodes, electrolytes and membranes a wide range of pollutants can be monitored. A Teflon membrane, a silver anode, a gold cathode and an aqueous solution of potassium chloride as the electrolyte have been used for oxygen. The oxygen molecules diffuse through the membrane to the electrolyte solution, where the dissolved gas is reduced or oxidised at the electrode causing a current to flow. The magnitude of the current is proportional to the partial pressure of the oxygen in the sample.

Recharging with new electrolyte is required at reguluar intervals. Calibration is achieved by the use of a reference gas or by exposing the sensor to air.

# 2.5 Volume Flow, Humidity, Temperature, Pressure

# 2.5.1 Volume Flow

<u>2.5.1.1</u> <u>Discontinuous/Manual Methods</u><sup>44,52-58</sup>. The mass flow of an air pollutant emission component is calculated from the volume flow and the concentration of the specific component, measured at the sample conditions. For the determination of the volume flow of flue gases it is necessary to measure their velocity in the stack or duct and multiply it by the stack or duct cross-sectional area.

The gas velocity is calculated from Pitot tube measurements taking the gas density into consideration. At velocities of less than 2 m/s the error of the Pitot measurement is excessive and the measurement should be made with an anemometer.

The density of the waste gas can be calculated, when the following quantitites are known:
- ambient pressure (at the altitude of the measurement plane);

- effective pressure (the difference between pressure in the duct and ambient pressure at the altitude of the measurement plane);
- average gas temperature in the measurement plane;
- composition of the waste gas.

Usually the volume flow is reduced to standard conditions.

2.5.1.2 Continuous Methods. There are various continuous measurement instruments which use the velocity of the waste gas for the determination of the volume flow.

<u>Averaging Pitots</u>. These are multi-impact port pitots which function in much the same way as single point pitots. They consist of two tubes of D-shaped cross-section back to back within a tube of circular cross-section. The probe is inserted into the duct, normal to the flow. There are a number of pressure sensing ports on both tubes, the upstream side senses the total pressure in its sector ie the line static pressure plus the velocity head. These different pressures enter the averaging tube where they mix producing one "high" pressure. The ports on the downstream side sense a suction pressure due to the flow around the probe. The pressures are again averaged producing a single "low" pressure.

The differential pressure ie high minus low, can be measured by a transducer and is proportional to the flow rate. The transducer is obviously an important part of the measurement system and can be electronic or pneumatic.

One variation on the design described above is the side-wall tap version. In this type the true line static pressure is used for the low pressure. This is claimed to improve the accuracy and the repeatability.

<u>Thermal Dispersion Flowmeters</u>. The sensor consists of two thermistors, one of which, the reference thermistor, is preferentially heated by a low power heating element. The sensor is mounted at the end of a probe which is inserted normal to the gas flow. The temperature difference between the reference thermistor and the flow thermistor is greatest when there is no flow and decreases as the gas flows across the flow element, thus cooling the reference thermistor. The temperature differential is electronically converted to a linearised signal output, which provides a reliable indication of flow velocity.

The actual construction varies; the thermistors may be contained in their thermowells directly exposed to the gas flow or located within a sensing tip, with a gas intake located on the side of the probe, in a pocket out of the main flow. Both of these configurations are intended to avoid fouling by particulates. Some models have a bypass loop to allow sample conditioning ie cooling or heating and moisture removal.

<u>Pneumatic Bridge Flowmeters</u>. A probe with two openings at the tip, which are continuously purged, is inserted into the duct normal to the direction of flow. The two channels are connected by an internal tube, the "pneumatic bridge". The purge gas, clean dry air, nitrogen or process gas, flows continuously in the system exhausting out of both openings. A portion of the pneumatic bridge tube is heated and there are thermoelectric sensors to measure the temperature gradients along the tube wall, external to the flow.

At zero velocity the bridge is balanced so that no flow occurs in the sensing portion of the tube and the two purge gas exhaust flows are equal. Where there is a flow in the duct a differential pressure is developed across the probe tip, unbalancing the bridge and causing a small flow of purge gas in the connecting tube. The thermoelectric sensors measure the shift in temperature gradient, which is related to the flowrate in the duct causing the differential pressure.

The purge gas prevents the process gas from entering the probe and thus eliminates problems with plugging, fouling, condensation and corrosion. The quantity of purge gas required is small, usually less than 0.85  $m^3/h$ , which is insignificant compared with the total flow.

Calibration is related to the gas density and the velocity profile. The output signal may be connected to any remote data logging, device, meter, recorder or readout. <u>Vortex Shedding Flowmeters</u>. All vortex shedding flowmeters operate by having a non-streamlined bar or strut running across the flow diameter, creating turbulence downstream. Above a certain minimum Reynolds number, that is, above a certain velocity for a given fluid and duct and at fixed conditions, this turbulence assumes a regular pattern of vortices. The number of vortices shed downstream of the strut per unit time is directly proportional to the fluid flow rate. This is expressed mathematically as vortex frequency,

F = SV/D

where S = Stroudal number (0.207); V = fluid velocity (m/s); D = obstruction (strut) diameter (m).

A pressure sensor can be used to detect the vortex frequency but ultrasonic sensing is superior as it produces a high quality signal over a wide turndown range and a simple rod can be used as the vortex generation strut.

The ultrasonic beam is transmitted through the vortex pattern downstream of the strut. The beam is modulated by the vortices and, if the sizes of the beam and vortices are correctly matched, the modulation index is almost directly proportional to the fluid velocity.

<u>Ultrasonic Flowmeters</u>. Ultrasonic flowmeters sometimes use a method which requires the presence of particles, but another type measures the transit time between two ultrasonic transducers. The transducers are located on opposite sides of the duct, one further downstream than the other.

The gas velocity is given by

$$V = \frac{(L + D)}{2L} (1/t_1 - 1/t_2)$$

where V = measured velocity;

- L = distance along the duct axis between the transducers;
- D = distance perpendicular to the axis between the transducers;
- t<sub>1</sub> = transit time upstream;
- $t_2$  = transit time downstream.

A built in flow computer calculates the volumetric flowrate from the velocity.

Two systems of this type have passed the suitability test in the FRG.

## 2.5.2 Humidity

Measurement methods for humidity are:

- Condensation

The water vapour is condensed in a radiator, using liquid nitrogen or dry ice in a solvent, eg methanol as a cooling agent.

- Adsorption

The adsorption is performed by a suitable adsorbent such as silica gel, calcium chloride, magnesium perchlorate or phosphorus pentoxide.

- Wet and Dry Bulb Temperature

A sample volume is extracted from the waste gas in the duct. The sample is brought into contact with two similar thermometers. One is exposed directly to the gas, while the other, the wet bulb, has a damp wick wrapped around it. The cooling effect due to evaporation of moisture from the wick causes a difference in the temperatures of the two thermometers. The water vapour content in the waste gas can be calculated based on this temperature difference and other parameters. The method can only be applied, if there are no droplets in the waste gas and the waste gas temperature is below 150°C.

The humidity also can be calculated from a mass balance.

Continuous humidity monitors are still at the development stage, however in-situ infra-red instruments for this purpose are promising.

## 2.5.3 Temperature

Temperature measurement is usually made with:

- mercury thermometers
- resistance thermometers
- thermocouples

## 2.5.4 Pressure

For the determination of the ambient pressure mercury manometers or equivalent instruments are suitable.

The determination of the static and dynamic pressure in the waste gas is generally performed with liquid manometers or with inclined liquid manometers, often filled with water. Pressure transducers are also available for this purpose and for continuous recording they facilitate data acquisition.

## 2.6 Calculation of SO<sub>2</sub> Emissions from Fuel Sulphur Content Measurements

The proposed Directive permits the competent authorities to allow discontinuous  $SO_2$  measurement in specific cases where  $SO_2$  emissions are determined by calculation or other appropriate means, which have been verified and approved. Several Member States currently measure fuel sulphur contents, from which  $SO_2$  emissions can be calculated, provided that

- the degree of sulphur retention in the system is known with reasonable accuracy;
- there is no sulphur dioxide abatement equipment;
- the fuel feed rate is known.

# 2.7 Data Treatment Devices<sup>8-12</sup>

The calculation of mean concentrations for an averaging time and the conversion of measured concentrations to standard conditions and to a given reference oxygen or carbon dioxide content make it necessary to use electronic data treatment devices.

A scheme derived by TÜV Rheinland in 1981, for the gradual expansion and improvement of an electronic data treatment system can be used to illustrate the

development of electronic evaluation equipment to the degree of sophistication which will be required to comply with the Commission's proposals. The stages of expansion are represented schematically in Fig. 2.

The most basic system is simply a chart recorder connected to the measuring instrument. An integration device can be added to smooth out the measurement signal and to trigger an alarm if the limit value is exceeded. A further development is the addition of a classification and counter unit which stores the integrated measurement values in the form of a frequency distribution curve over a long period of time. The chart recorder can be retained for parallel indication and recording of the instantaneous values so that peak values can be assessed. The final stage of development is represented by the incorporation of a processor to convert measured values to their reference values before they are fed to the classification unit.

Although such a scheme of staged expansion might be useful in some cases, microprocessors which are capable of performing all of the required data treatment steps are available. Some electronic data treatment devices have been suitability tested in the FRG. The main requirements of the suitability test are listed below.

- The data treatment device must undergo a suitability test for at least three months to guarantee the compliance with certain minimum performance specifications.
- The device must have the facility for adjusting between different integration times in the range 3 to 60 minutes. An integration time of 30 minutes has to be possible. The integration error may be a maximum 1% of the chosen integration time.
- The availability of the devices has to be at least 95%, and preferably 99%.
- The number of classes for the frequency distributions should be at least 40, if the measurement results are directly classified (second expansion stage).

- The number of classes should be at least 20, if the measurement results are related to reference values by a processor before being classified (third expansion stage). The 10th class should be in the range of the emission standard.
- The error caused by the mathematical operations involved in the conversion to the reference values may not exceed 2% of the calculated values.
- If the single measurement values are to be stored the storage capacity shall be sufficient to hold half-hourly mean values relating to a total period of about 40 days.
- All information should be available at any time on a printer or visual display unit.

## 2.8 Installation, Maintenance and Calibration of Continuous Measurement Devices<sup>89</sup>

#### 2.8.1 General Notes

The accuracy of emission measurements is highly dependent on the selection of a suitable measurement position. The measurement positions have to be chosen in such a way that a set of measurement results is obtained which is sufficiently representative of the spatial and time dependent distribution of the mass concentration.

It is preferable to perform the reference measurements for calibrating continuous measurement devices in the same measurement plane. For this purpose suitable ports in the waste gas duct should be provided for the reference sampling probes. Interference effects between the continuous measurements and the reference measurements must be avoided.

The selection of the measurement position should also take account of the need to service the measuring devices at regular intervals. Access to the the installed measurement instruments and the sampling ports must be easily achievable from a safe working platform. The following factors must be considered when installing emission measurement devices:

- sufficient protection against weather influences;
- vibrationless and shock free installation;
- avoidance of external influences due to gases and vapours on the measurement device;
- avoidance of interfering electrical or magnetic fields in the vicinity of the measuring device or measured value transmission system;
- gas lines which could contain condensate should be laid with a slight inclination;
- materials for the sampling equipment should be chosen to prevent corrosion and to avoid interactive reactions between these materials and the sample gas;
- sampling lines should be kept as short as possible to obtain fast response times and to minimise the extent of chemical reactions.

Furthermore, in sampling for gas analyses the following items have to be considered:

- The gas sample should be dust free and in most cases dry.
- The volumetric flow rate, pressure and temperature must lie within the limits dictated by the measurement method.
- The cooling of high temperature sample gas leads to condensation. The possibility of the measured component being soluble in the condensate must be taken into account.

A small positive pressure at the measurement site is sufficient to provide an adequate volumetric flow of sample gas if the analyser outlet discharges directly to the atmosphere. The volumetric flow rate is adjusted to the desired value by needle valves. High pressure conditions require additionally the use of a pressure reducing valve. Under sub-atmospheric pressure conditions gas suction pumps must be employed.

In many cases the best measurement results for continuous instruments can be obtained if the complete measurement device including the sample line is supplied by one manufacturer. Many instrument manufacturers also supply auxillary equipment for sampling, conditioning and metering, which is also included in the suitability testing procedure in the FRG.

#### 2.8.2 Calibration of Particulate Measurement Devices

Before undertaking calibration the measurement device should be tested by

- using reference standards (optical grating filter, glass filter) to check the operation of the instrument;
- inspection all instrument components for any obvious fault;

- checking gas transmission components for leakage;

The functional tests should be carried out at least once a year checking that the sample stream flow is steady.

2.8.2.1 Dust Concentration Measurement Devices. The zero point and reference point for in-situ photometric devices should be checked using a dust free reference path of the same length as the measuring path in the actual waste gas duct. Testing should include checking of devices intended for protecting the optical components from dirt. Additionally, a check should be made on the measured value transmission and recording system to ensure interference free functioning.

Functional tests of light scattering instruments should be made using filter cartridges. Testing should include checking the devices for the protection of the optical components against dirt, including the entire sampling line. The heating system and the volume flow control have to be checked. Functional tests on Beta radiation absorption principle measuring devices comprise of single filter measurements and of checking the sampling system and the time taken for the extraction cycle. Systems which employ isokinetic extraction should also be tested to ensure that this feature is operating correctly.

The calibration determines the relationship between the instrument reading and the actual dust concentration (analytical function). The calibration should be performed by gravimetric reference measurements according to national or international guidelines.

The calibration should be carried out as far as possible under all operation conditions which may occur at the facility which is to be monitored. The number and duration of the individual measurements depends on the operation characteristics and the emission behaviour of the particular plant. If necessary, certain emission conditions which are required for the calibration can be achieved by temporarily adjusting existing dust control equipment.

To make sure that an accurate relationship between the instrument reading and the particulate concentration in the waste gas at the measurement site is obtained, at least 12 measurements under constant plant operation conditions are required. The necessary number of measurements required increases directly with the number of different operation conditions or emission relationships. Calibration measurements should, where possible, be carried out within one single period of unattended operation.

<u>2.8.2.2</u> Smoke Density Meters. Smoke density meters can only provide a qualitative indication of the dust concentration in the waste gas. For this reason the instruments are only adjusted on the basis of empirical values according to the visibility limits of the smoke plume. In a few cases an adjustment in accordance with the Ringelmann chart may be achieved.

The adjustment of the zero point is performed on a smoke free measuring path of the same length as the measuring path in the waste gas duct. The measurement reflector and its associated closing shutter are adjusted to suit the measuring path length and the optical systems focussed to obtain a sharp image. As a reference, the manually operated zero point control reflector is set to suit the brightness of the smoke free measuring path. The instrument sensitivity is adjusted by means of test filters of known opacity.

## 2.8.3 Calibration of Measurement Devices for Gaseous Components

2.8.3.1 Functional Testing. For in-situ measurement devices the performance of the instrument is tested using reference standards (eg filters) over a gas free measuring path. Multicomponent measurement devices should be checked for cross sensitivity between the measurement channels using chart records taken over a period of several days. Optical systems should be checked for dirtiness of the optical surfaces.

For extractive measurement devices the calibration begins with checking the gas tightness of the instrument, including the gas sampling and conditioning equipment, and checking the measured value transmission. The adjustment of the zero point and span is performed using calibration gases or, for conductometric and potentiometric measuring methods, with calibration solutions. The calibration function supplied by the manufacturer is then checked against a number of reference points uniformly spaced over the whole measuring range. If the calibration function is non-linear a greater number of reference values is required.

Finally the interference error must be checked. This can be done by feeding the measured component, together with a suspected interferent which could be present in the waste gas, into the analyser via the sampling system.

During the calibration period the following performance characteristics should be determined at the maintenance intervals

- the zero and span drift;
- the response time (90% time).

2.8.3.2 Measurement Devices for Sulphur Dioxide and Nitrogen Oxides. The calibration of the measurement devices is performed by checking the analytical function with the aid of a reference method according to national or international guidelines. The selection of the reference measurement method is dependent on the concentration of the relevant component in the waste gases. At least 30 pairs of measurements are necessary for calibration purposes to calculate the confidence range of the measurements.

In addition, for optical measurement systems, the waste gas conditions in the measurement plane (temperature, pressure, water vapour content) should be measured in order to allow the measurements taken under operating conditions to be converted to standard conditions.

## 2.8.4 Evaluation of the Calibration Measurements

In the FRG specific guidelines<sup>59,79</sup> exist for the evaluation of the measured values and for calculating the statistical relationship between the instrument readings and the reference measurement values.

If a large portion of the measurement range is covered by measured value pairs the determination of the confidence range is performed by constructing a linear regression without zero point hypothesis.

If, on the other hand, the measured value pairs lie close to one another, the confidence range is calculated from the differences in the individual pairs of measured values.

Non-linear relationships can also be presented by corresponding parabolic functions.

## 2.8.5 Maintenance and Operating Procedure

Maintenance of emission measurement devices can often be carried out by the operator's trained personnel. As a rule it is only necessary to call in the manufacturer's maintenance service for repair work and annual inspections. The scope and frequency of the maintenance work is specific to the instrument and depends on the operating conditions. The maintenance frequency recommended by the manufacturer or derived from suitability tests should be complied with. All maintenance work should be logged.

<u>2.8.5.1</u> In-situ Measurement Devices. The following should be carried out during maintenance or more frequently as part of the normal operating routine:

- cleaning the optical surfaces;
- checking the zero point and reference point measurement signals and sensitivity, if applicable;
- cleaning of filters (purging air, cooling air);
- checking the measurement data recording.

2.8.5.2 Measurement Devices with Extractive Sampling. The following should be carried out by the operator about once a week as part of the normal operating procedure:

- checking the sampling system heating;
- replacement of consumable materials (filters, reagent solutions);
- changing or cleaning of the sampling filters;
- checking that detectors are functioning;
- checking that condensate separation systems are functioning;
- leakage check on gas supply lines and components;
- checking the sample flowrate;
- checking the instrument zero point and sensitivity;
- checking the adsorbent dosing, if applicable.

During any shut-down of the measurement devices all sample gas lines should be purged with inert gas and condensate collection vessels should be drained.

## 2.9 Emission Measurements and Procedures Used by Member States

Table XV summarises the emission measurements and procedures carried out at present in each Member State.

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Methods	
Measurement	
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TABLE	

Country: BELGIUM

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so2	M/C	X	X	0 A AI	gravimetric hydrogen peroxide hydrogen peroxide thorin	electrochemical flame photometrical
NO <sub>X</sub>	×	X	×	0 A AI	sodium salicylate chromotrophic acid	chemiluminescence
PARTI CULATE MATTER	×	X	×	0 A AI	gravimetric method	transmissometers beta gauge
0 <sub>2</sub> only combustion control parameter	¥	×	×	0 A AI	paramagnetic Orsat	paramagnetic
CO <sub>2</sub> combustion con- trol parameter						
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	M M M/G	XXXX		0/A/AI	thermocouple pitot tube manometer adsorption	
DATA TREATMENT	1	1	1	1	1	1

TABLE XV. - cont'd

Country: DENMARK

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	W	х	x			NDIR
NO <sub>X</sub>	W		×			<b>Chemiluminescence</b>
PARTI CULATE MATTER	W	Х	X		Gravimetric	
0 <sub>2</sub> only combustion control parameter	¥		Х			
CO <sub>2</sub> combustion con- trol parameter	Σ		X			
MASS/VOLUME FLOW - Temperature	Σ	×		c	Thermocouple	
- verocity - Pressure - Humidity	ε χ χ	× × ×		D	ritot tube Manometer	
DATA TREATMENT						

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>		×	X		Iodometric thiosulphate Titrimetric peroxide Peroxide-Thorin	
NOX	W	х	Х			
PARTICULATE MATTER	Ж		X		Gravimetric	
0 <sup>2</sup> only combustion control paramete	×		×			
CO <sub>2</sub> combustion con- trol parameter						
MASS/VOLUME FLOW - Temperature	W	X			Thermocouple	
<ul> <li>Velocity</li> <li>Pressure</li> <li>Humidity</li> </ul>	ΣΣΣ	XXX		o	Pitot tube Manometer	
DATA TREATMENT	:	4				

TABLE XV. - cont'd

Country: EIRE

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Country: FEDERAL REPUBLIC OF GERMANY

			Меав	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
S02	T	×	×	AI	mainly hydrogen peroxide-thorin titametric hydrogen peroxide	NDIR NDUV/dispersive UV Conductometric In-situ NDUV dispersive UV
NON	Σ	×	×	IV	Phenoldisulphonic acid Acidimetric Na salicylate 2-6 Dimethylphenol	NDUV dispersive UV NDIR Chemiluminescence
PARTI CULATE MATTER	Σ	Х	x	IV	Gravimetric	Photometric in-situ Some Beta Gauge
0 <sub>2</sub> only combustion control parameter	Σ	×	×	0 <b>1</b>	Orsat Paramagnetic	Paramagnetic Some electrochemical (zirconium oxide)
CO <sub>2</sub> combustion con- trol parameter	Σ		×	o		NDTR
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	M/C M/C	×××	· ×××	0 0/AT 0 0	thermocouple pitot tube manometers two thermometer method	calculation from other parameters pressure transducers
DATA TREATMENT	Σ		×	AI	P	computation

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			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Meth
so2	E I O	×	×	0	<ol> <li>I. Iodometric method</li> <li>Acidimetric method</li> <li>(SO<sub>2</sub> + SO<sub>3</sub>)</li> </ol>	
						<ol> <li>UV Fluoresce</li> <li>NDIR</li> <li>Interferent1</li> <li>method</li> </ol>
NO <sub>X</sub>	X	X		0		Chemiluminescen method
PARTI CULATE MATTER	Σ	×	×	o	- Gravimetric methods Isokinetic sampling - Granulometric measurements	
	:	:		0		l.βraymethod 2.opacimeters
02	F	N N	X at the xit of he cono- iser)			<ol> <li>Electrochemi and paramagn</li> <li>Zirconlum ox</li> </ol>
CO <sub>2</sub> combustion con- trol parameter	æ	X		0	Orsat apparatus: absor- ption in a potassium hydroxide solution	
				0		I.R. absorption
MASS/VOLUME FLOW - Temperature	Σ	X			- thermocouple	
- Velocity - Pressure - Humidity	Σ	×			- platinum resistance Pitot tube Barometers - Manometers Condensation	0
DATA TREATMENT						

TABLE XV. - cont'd

Country: FRANCE

TABLE XV. - cont'd

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Country: GREECE

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	M/C	х		0		conductometrical
NO <sub>X</sub>	М	X		0		chemiluminescence
PARTICULATE MATTER	W	X		0	gravimetric	
0 <sub>2</sub> only combustion control parameter	Σ		X	0		
CO <sub>2</sub> combustion con- trol parameter	¥		×	0		
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	¥	ХХХХ		o	thermocouple pitot tube manometer	
DATA TREATMENT	1	1	1	3	1	1

cont'd	
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XV.	
TABLE	

Country: ITALY

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	υ					
NO <sub>X</sub>	W	x		0		chemiluminescent method
PARTI CULATE MATTER	W	X	X	o	Gravimetric determina- tion (combined probe (ENEL) Isokinetic sampling)	
						opacimeters
0 <sub>2</sub> combustion control parameter	M O <sub>2</sub> measured at the exit of the economiser		X			zirconium oxide method
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	W W W	XXX			thermocouples Pitot tube micromanometers condensation	
DATA TREATMENT						

TABLE XV. - cont'd

Country: LUXEMBOUR(	chi					
			Meas	surements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (O) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	C/M	х		0/AI		
NOX	W	×		0/AI		chemiluminescence
PARTI CULATE MATTER	W	×		0/AI	gravimetric	
02 only combustion control parameter	¥		×	0		
CO <sub>2</sub> combustion con- trol parameter	¥		×	0		
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	ΣΣΣ	XXX			thermocouple pitot tube manometer	
DATA TREATMENT					I	1

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TABLE XV	
TABLE XV	

Country: NETHERLANDS

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	M/C	×	×	0 A AI	Iodometric Thiosulphate Gravimetric peroxide Hydrogen Peroxide- Thorin	NDIR NDUV In-situ Dispersive UV Second Derivative UV
NOX	Ψ	×	×	0 A AI	Na salicylate	NDIR Chemiluminescence Second derivative UV
PARTICULATE MATTER	Ψ	×	×	0 A AI	Gravimetric	Photometric in-situ
02 only combustion control parameter	¥	×	×	0 A AI		Paramagnetic
CO <sub>2</sub> combustion control parameter					NDIR	
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	M/C M/C M/C	XXXX		0/A/AI	thermocouple pitot tube manometer adsorption	
DATA TREATMENT	1	1		1	1	I

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub> *	C - M*	×		¥	(ref. method 6 EPA) Barium perchlorate thorin titration method	
NOX						
PARTI CULATE MATTER	W	X	х	¥	(ref. method 5 EPA) Gravimetric method Isokinetic Sampling	
				0	ō	pacimeter
02	¥			V	(ref. method 3 EPA) Orsat method. Absorp- tion in a pyrogallic acid and potassium hydroxide solution	
co <sub>2</sub>	Ж	×		¥	(ref. method 3 EPA) Orsat method. Absorp- tion in a potassium hydroxide solution	
MASS FLOW - Temperature	W	×			- Thermocouple Iron-	
- Velocity - Pressure - Humidity	W	XX			- Pitot tube - Barometer-Manometers - Condensation	
DATA TREATMENT						

\* The  $SO_2$  measurements will begin next year (1986).

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TABLE XV. - cont'd

Country: PORTUGAL

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			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	¥	x	×	AI	Barium - thorin titra- tion method (ref. method 6 EPA)	
				o		<ol> <li>Second-derivative UV spectrometry</li> <li>NDIR</li> <li>UV fluorescence</li> </ol>
NOX	z	×	×	IV	(ref. method 7 EFA) Absorption in dilute solution of sulphuric acid and hydrogen peroxide	
						<ol> <li>Second-derivative UV spectrometry</li> <li>NDIR</li> </ol>
PARTI CULATE MATTER	×	м	×	• VI-0	(ref. method 5 EPA) gravimetric method isokinetic sampling	
						l.βgauge 2.Opacimeters
0 <sub>2</sub> only combustion control parameter	M measurement located at the exit of economiser		х			Electrochemical cell
CO <sub>2</sub> combustion con- trol parameter	×	×	¥	o	Orsat analysis (ref. method 3 EPA)	
MASS/VOLUME FLOW - Temperature - Velocity	¥	х			<ul> <li>thermocouple</li> <li>thermometer</li> <li>S-type PITOT tube</li> </ul>	
- Pressure - Humidity	Σ	×			(ref. method 2 EFA) - manometer - barometer (Ref. method 4 EPA condensation impingers in series	
DATA TREATMENT	Averaging time:	1 hr 1 day				
NO emission concent Manual measurement Automatic measureme	rations converted : All the resul nt: Transmission On the basis	to a re ts are n by election of the l	ference oted on ric line ast mini	of 02 or CO2 conte a record book (see s to the control r sterial order (198	nt Section 1.12). oom (graphic recorder) 4) see Section 1.12.	

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Country: SPAIN

TABLE XV. - cont'd

cont'd	
XV	
TABLE	

Country: UNITED KINGDOM

			Meas	urements		
	Calculation (C) or/and Measurements (M)	disc	cont	operator (0) authority (A) or authorised institution (AI)	Manual Methods	Automatic Methods
so <sub>2</sub>	C/M	×	×	0	Iodometric Thiosulphate Titrimetric peroxide Peroxide-Thorin	NDIR UV Fluoresence In-situ NDIR
NOX	Ψ	×	×	0	Na salicylate Saltzman	NDIR Chemiluminescence
PARTICULATE MATTER	W	×	×	0/AI	Gravimetric	Photometric in-situ
0 <sub>2</sub> only combustion control parameter	¥	×	×	0	Paramagnetic	Zirconium oxide
CO <sub>2</sub> combustion control parameter	×	×	×	0		
MASS/VOLUME FLOW - Temperature - Velocity - Pressure - Humidity	жжур	<b>X X X X</b>		o	Thermocouple Pitot tube, venturimeter Manometer, aneroid barome	ter
DATA TREATMENT						





3 RD. EXPANSION STAGE

Fig 2 Methods of Data Treatment for Emissions Measurements <sup>89</sup>

#### 3. DISCUSSION OF THE MEASUREMENT METHODS

The criteria used for the selection of measurement methods may be different for existing plant than for new plant. Attention should be given to plant design and to the positioning of measurement devices during the planning stage of new installations. It is expected that the extractive type instruments will be more widely used on existing plant but the in-situ type is likely to be more suitable for new plant, both now and in the future. Also, in-situ instruments are being developed for multi-component analysis applications.

## 3.1 Particulates

#### 3.1.1 Discontinuous/Manual Methods

The existing National Standards are similar and specify important parameters such as sampling position, number of sampling points and the necessity of isokinetic sampling. There is an ISO standard in preparation which will eventually be the primary reference standard. This standard provides an option for sampling with the filter located within the duct or external to it. There are advantages and disadvantages with either method. If the filter is inside the duct the sample is taken at the duct temperature and there is no problem of deposition in the probe but some filtration media may be attacked by flue gas components at duct temperatures. Systems with an external filter may have problems with deposition in the probe.

## 3.1.2 Continuous/Automatic Methods

In-stack transmissometers or opacity meters and Beta ray attenuation instruments are the two most widely used particulate analysers. Transmissometers are used throughout the world and although they do not give an absolute value for the particulate mass emission rate they can be calibrated for quantitative measurement. A procedure for calibration is given in a German guideline. Beta ray attenuation instruments are mainly used in France.

Transmissometers have the advantages that there is no physical disturbance of the gas stream and that the measurement is made over a relatively large gas volume. The best instruments of this type have been demonstrated to be reliable and simple to maintain. All instruments are sensitive to maladjustment of the light beam and careful positioning is required to overcome these problems. Other problems are interference by condensed water vapour and solids deposited on the optical surfaces. The main disadvantage, however, is that the calibration is based on the assumption of constant particle characteristics. It is recommended that transmissometers of the double beam, double pass type are used.

All measurement methods for particulates have the disadvantage that parameters such as attenuation or light scattering rather the actual particle mass are used. Therefore, larger uncertainties have to be expected than are usual in the measurement of gaseous components. The range of uncertainty given by the confidence and the tolerance intervals differs from plant to plant and may be as high as 30 to 50% of the given standard for optical methods.

Nevertheless, this method gives continuous information about the emission behaviour of the individual plant, which is preferable to accurate but infrequent results by a manual gravimetric method.

In the case of opacimeters the facility for calibration to actual particulate concentration is an essential feature. Measurement devices using the obscuration method are unsatisfactory for the purposes of the proposed Directive.

Beta ray attenuation instruments give results which are virtually independent of particulate composition and size but they have the inherent disadvantages of deposition methods, namely a small sampling area, deposition in the sampling lines and the need to mount bulky equipment close to the sampling point. Such instruments operate over a wider selected concentration range than transmissometers, especially at the lower end. In addition, the particulate collected can be used for further analysis.

Light scatter instruments represent an alternative to transmissometers and beta ray attenuation instruments and they may be used to measure a wide range of particulate concentrations.

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## 3.2 Sulphur Dioxide

#### 3.2.1 Discontinuous/Manual Methods

There are several methods in use in Member States and in most cases these are regulated by a National Standard. As can be seen from Table VII, the hydrogen peroxide-thorin method is widely accepted in Europe and is a reference method in the United States. This method will be published as an ISO reference method and will therefore become the primary reference standard for manual measurement of sulphur dioxide.

## 3.2.2 Continuous/Automatic Methods

Many instruments using various principles of operation are available for the continuous measurement of sulphur dioxide, however there are no National Guidelines except in the FRG and France. An indication of the numbers of instruments of each type available is given in Annexe 2. The data is intended only for the purposes of comparison and the survey of manufacturers was by no means exhaustive. It is clear that photometric gas analysers of the non-dispersive type are by far the most common. At present NDIR instruments outnumber the NDUV type, but the UV instruments may be less prone to interference as water vapour does not absorb in the UV region. The second derivative ultra-violet and the interferential type instruments may also have advantages with respect to interference, however the fact that they are produced by only one manufacturer is a severe disadvantage.

Extractive IR/UV photometers are more common than the in-situ type and more reliable performance data are available for them. In general extractive instruments appear to have greater precision and stability than the existing in-situ instruments, some of which have problems with cross-sensitivity and sensitivity to maladjustment of the beam. Cross-stack instruments do, however, have some inherent advantages over extractive instruments. They linearly average concentrations over their measuring path, although in the case of severe flow stratification obtaining a representative measurement can still be a problem. There are fewer separate components since no gas conditioning equipment is required and calibration is simple if the instrument has in-built calibration cells. Also, a single instrument can be used to monitor several gaseous pollutants and even particulates. None of the other types of sulphur dioxide analyser is widely available and performance data is sparse, although some have no other specific disadvantages.

In special cases, the proposed Directive permits the use of discontinuous measurement of  $SO_2$  emissions, in conjunction with determination by calculation or other means, to replace continuous measurement. The method must be verified and approved by an authorised body. Measured fuel sulphur contents might be used to calculate  $SO_2$  emissions with acceptable accuracy in some cases.

## 3.3 Oxides of Nitrogen

#### 3.3.1 Discontinuous/Manual Methods

In general these have been less precise than the automatic optical methods but will continue to be applied for discontinuous emission control and for the verification of guideline requirements. The preferred method would be the sodium salicylate analysis, which is currently in use in a number of Member States, however, the time lapse of more than 24 hours between sampling and obtaining the result is undesirable. The chromotropic acid method has been developed recently to provide for more rapid analyses, but its precision as compared with the sodium salicylate method is not yet known. A further method, the DMP (2,6 dimethyl phenol) method, has recently been developed for use in the FRG for this purpose and a guideline is in preparation.

#### 3.3.2 Continuous/Automatic Methods

The IR/UV photometric methods used for sulphur dioxide can also be applied to the measurement of nitrogen oxides and the comments made in section 3.2.2 still apply. However the chemiluminescence method is in general use in the majority of Member States and several National Standards exist. Instruments using extractive NDIR and NDUV techniques in particular, have been developed more recently but are already as widely available as chemiluminescence analysers as shown in Annexe 2 and some meet suitability test requirements in the FRG. In general, instruments for nitrogen oxide emission monitoring actually measure nitrogen monoxide only, but this is satisfactory since in most cases the amount of  $NO_2$  is less than 5%. (If nitrogen dioxide measurements are required a converter can be incorporated into extractive instruments but in-situ instruments can generally measure nitrogen monoxide only<sup>88-90</sup>.)

#### 3.4 Oxygen and Carbon Dioxide

Oxygen and/or carbon dioxide are measured at all large plant for the purposes of combustion control. The analysers are usually located upstream of the economiser, so if there is abatement equipment downstream it would be necessary to install additional instruments for emissions measurement.

## 3.4.1 Discontinuous/Manual Methods

The Orsat apparatus can be used for the manual intermittent measurement of oxygen and carbon dioxide but continuous analysers, such as zirconium oxide cell and paramagnetic devices for oxygen and infra-red spectrometers for carbon dioxide, are in general use.

## 3.4.2 Continuous/Automatic Methods

A large proportion of the continuous analysers are of the extractive type, in which water vapour is removed during the conditioning process, so that the measurement is on a dry basis. The results can therefore be used directly for further calculations. In-situ instruments measure concentrations on a wet basis so the water content of the flue gas must also be measured. The lifetime of in-situ type sensors is generally shorter because of the more severe operating conditions than that of similar sensors in an extractive application. Also, it is desirable that the calibration procedure can be carried out insitu.

## 3.5 Volume Flow, Temperature and Humidity

## 3.5.1 Discontinuous/Manual Methods

The measurement of the volume flow of flue gases is an essential part of the determination of particulate emissions from industrial processes. The manual methods for volume flow measurement used in Member Countries are all similar, as are those for measuring temperature and humidity.

#### 3.5.2 Continuous/Automatic Methods

At most large combustion plant it is possible to calculate the volume flow of flue gas from other measured parameters, such as fuel feed rate or composition and steam load. The calculated volume can be corrected to specified reference conditions. It may be necessary to measure volume flow continuously at certain types of installation, cement kilns, for instance. In addition to the well established methods, such as pitot tubes for velocity measurement and turbine flowmeters, various new techniques have been developed. These include vortex shedding, ultrasonic, pneumatic bridge, thermal dispersion and laser instruments. At least some of these methods are under evaluation for suitability tests in the FRG.

It may be necessary to know the humidity of the flue gas to determine the concentrations and mass emission rates of pollutants. Instruments for the continuous measurement of the water content of flue gases are just becoming available; in-situ infra-red instruments for this purpose are promising.

## 3.6 Data Treatment

It is expected that the large quantities of data given by the measuring instruments installed as a result of the proposed Directive will be best handled by special data treatment devices. Emissions should be calculated to standard conditions and to a given reference oxygen content so that they can be compared directly with the limit concentrations.

It will be possible to calculate trends and forewarn the operator using a trend alarm, picture screens etc. This information will be helpful in reducing emission concentrations.

The results of these analyses should be in a form to provide reliable emissions inventory data.

In the Federal Republic of Germany all large combustion plants have been equipped with a data treatment device.

#### 4. SURVEY OF LARGE COMBUSTION INSTALLATIONS

#### 4.1 Numbers of Installations Affected by the Commission's Proposals

The number of installations greater than 50 MW thermal input which fall into the categories covered by the draft CEC Directive, are shown in Tables XVI-XXVII for individual Member States. No brick works greater than 50 MW thermal input have been identified in any Member State, so this category has not been included.

Several factors may have led to discrepancies in the figures:

- Some sources have determined numbers of plant while others have determined the number of units, however, as far as possible, the figures have been given in accordance with the draft Directive. This states that "where two or more separate plants are installed in such a way that, taking technical and economic factors into account, their waste gases can be discharged through a common stack, the combination formed by such plants is to be regarded as a single unit".
- In general there has been some difficulty in obtaining precise information on installations in the industrial sector, making some degree of estimation necessary.
- Figures from some sources may include some installations smaller than 50 MW thermal input.
- The European cement manufacturers object to the proposed Directive, therefore data on this industry are sparse.

The data sources and their reliability are discussed below for each Member State.

## Belgium (Table XVI)

The number of industrial boilers was determined by the comparison of figures from various sources. The information on industrial boilers is very

poor. The total number of ore-roasting plant may include plant of less than 50 MW thermal input.

#### Denmark (Table XVII)

Figures are available only for power stations. Many Danish power stations have facilities for coal or oil firing, although coal is usually the preferred fuel, which makes it difficult to categorise them according to fuel type. Such installations have been included in the category of the principal fuel.

#### Federal Republic of Germany (Table XVIII)

The data on boilers are based on an investigation performed by the Umweltbundesamt. Data on plant are available for power plant, but the information on industrial installations is insufficient.

The total number of ore-roasting plant of all sizes is given but this figure could include plant of less than 50 MW thermal input.

The figures for cement work installations are based on data from the Forschungsinstitut der Zementindustrie (Research Institute for the Cement Industry).

#### Greece (Table XIX)

There are no boilers of greater than 50 MW thermal input except for those in power plant. The data for power plant were obtained from the Public Power Corporation, which operates all power plant in Greece.

#### France (Table XX)

The number of installations has been determined according to the definitions given in the draft Directive on large combustion installations. The numbers of installations of greater than 300 MW thermal input were supplied by CITEPA. The numbers of installations of less than 300 MW thermal input were estimated from fuel consumption and load data.

The statistics for the French cement industry were not available.

#### Eire (Table XXI)

Data were obtained by circulating a questionnaire to the relevant departments of Local Authorities. The fuel type and exact size of installations other than power stations is not known, except for the one industrial installation in the greater than 300 MW thermal input category.

## Italy (Table XXII)

The number of installations given in the table applies only to electricity generator boilers. These statistics were supplied by ENEL. It was not possible to obtain data for heating, process industry or cement works.

## Luxembourg (Table XXIII)

The authorities were available to supply precise data on the small number of installations greater than 50 MW thermal input. There are no cement work installations or ore-roasting plant in this size range.

## The Netherlands (Table XXIV)

The figures for electric power plant were obtained by KEMA and those for industrial plant are taken from an emissions inventory compiled by TNO. In some cases single boilers may have been counted as plant.

The figures for ore-roasting plant may include plant of less than 50 MW thermal input.

#### United Kingdom (Table XXV)

Data were obtained by circulating a questionnaire to the CEGB, the Institution of Environmental Health Officers and Local Authorities and by contacting various trade associations The exact size and the fuel type of industrial boilers is not known.
### Portugal (Table XXVI)

The numbers of installations presented in the table were given by the Ministry of the Environment. They include paper pulp works, power stations and cement works.

### Spain (Table XXVII)

The data presented in the table refer only to electricity installations and were supplied by UNESA. It was not possible to obtain numbers for heating, process industries and cement works.

### 4.2 Numbers of Installations at which Emission Measurements are already made

The number of installations, in each category, at which emissions of particulate,  $SO_2$  and  $NO_x$  are already measured continuously, is shown for each Member State in Tables XXVIII-XXXIX. Oxygen and/or  $CO_2$  are measured on all boilers of greater than 50 MW thermal input.

Capa Insi	acity of callation MW	Total Number	Coal Fired	Oil Fired	Gas Fired
1.	Installation	s for electric	ity, heating and proce	88	
50 101 201	··· 100) ··· 200) ··· 300)	119	90	13	16
	>300	27	10	11	6
Sub	otal	146	100	24	22
2.	Installation	is in cement wo	rks*)		
50 101	··· 100 ··· 200	2 (7)	2 (7)		-
201	300 >300			-	-
Sub	cotal	2 (7)	2 (7)	_	
3.	Installation	s in ore-roast:	ing plants	<u></u>	
50 101 201	100 200 300 >300	4   -			
Sub	otal	4	_	_	_
Tota	al number of	installations			
50 101 201	100) 200) 300)	125	92	13	16
	>300	27	10	11	6
Tota	1	152	102	24	22

### Member State: BELGIUM

\*) figures without brackets: number of installations

figures in brackets: number of boilers/kilns

# TABLE XVII. - Number of Installations

Capa	city of	Total	Coal	011	Gas
Inst	allation MW	Number	Fired	Fired	Fired
				·	
1.	Installation	is for electricity	, heating and proce		
50	100				
101	200	1 (1)	1		
201	300	2 (4)	2	,	
	>300	13 (59)	9	4	
Subt	otal	16 (64)	12	4	
2.	Installation	ns in cement works			
50	100				
101	200				
201	300				
	>300				
Subt	otal				
3.	Installation	ns in ore-roasting	plant		
50	100				
101	200				
201	300				
	>300				
Subt	otal				
Tota	al number of	installations			
50	100				
101	200	1 (1)	1		
201	300	2 (4)	2		
	>300	13 (59)	9	4	
Tota	 1	16 (64)	12	4	_

Member State: DENMARK

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\* utility installations only

figures without brackets: numbers of installations figures in brackets: number of boilers

# TABLE XVIII. - Number of Installations

Capa Ins	acity of tallation MW	Tota Numl	al ber	(	Coal Fired		0il Fired	1	Gas fired
1.	Installations	for e	lectricity,	heatin	ng and	process*)		<u>,</u>	
50	100	21 (3	339)	15	(165)		(146)	6	(28)
101	••• 200)	36 (3	354)	22	(232)	8	(103)	6	(19)
201	>300	171 (	185)	93	(120)	24	(48)	54	(17)
Sub	total	228 (8	878)	130	(517)	32	(297)	66	(64)
2.	Installations	in cer	ment works‡				<u></u>		
50	100	20	(32)	20	(32)		-		-
101	••• 200	_			-		-		-
201	>300	-			_		-		-
Sub	total	20	(32)	20	(32)	<del></del>	-	<u> </u>	
3.	Installations	in ore	e-roasting p	plants				. <u></u>	<u> </u>
50	100	5			* <u>-</u>				
101	••• 200	-							
201	··· 300	-							
Subi	total	5			-		-		-
Tota	al number of i	nstalla	ations						
50	100	46 (3	371)	35	(197)		(146)	6	(28)
101	··· 200)	36 (3	354)	22	(232)	8	(103)	6	(19)
201	>300	171 (1	185)	93	(120)	24	(48)	54	(17)
Tota	al	253 (9	910)	150	(549)	32	(297)	66	(64)

### Member State: FEDERAL REPUBLIC OF GERMANY

\*) figures without brackets: utility installations only.

figures in brackets: total number of utility and industrial boilers.

# figures in brackets: number of kilns.

## TABLE XIX. - Number of Installations

Capa Inst	acity of tallation MW	To Nu	tal mber	Cc F1	al red	C Fi	)il Ired	Gas Fired
1.	Installations	for	electricity	, heating	, and proc	ess*)		
50	100	_	(1)	-	(1)		(-)	-
101	200)	1	(3)	1	(1)	-	(2)	-
201	>300	6	(16)	4	(12)	2	(4)	-
Subi	total	7	(20)	5	(14)	2	(6)	
2.	Installations	in c	ement works	*)				
50	100	1	(3)	1	(3)		-	
101 201	••• 200		-	-	-		-	-
201	>300		-	-	-		-	-
Subt	total	1	(3)	1	(3)		_	_
3.	Installations	in o	re-roasting	plants	<u> </u>			<u></u>
50	100		-	-	-		-	-
101 201	••• 200		- -	-	-		-	-
201	>300		-	-	-		-	-
Subt	total		-	-	-		-	-
Tota	al number of in	nstal	lations					
50	100	1	(4)	1	(4)	-	(-)	
101	··· 200)	1	(3)	1	(1)	-	(2)	-
201	>300	6	(16)	4	(12)	2	(4)	-
Tota	al	8	(23)	6	(17)	2	(6)	

Member State: GREECE

\*) figures without brackets: number of installations

figures in brackets: number of boilers/kilns

## TABLE XX. - Number of Installations

Member State: FRAN	CE
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Capa Inst	acity of tallation MW	Total Number	Coal Fired	0il Fired	Gas Fired
1.	Installations	for electricity,	heating and proces	38	
50	100	409	31	288	90
101	··· 200)	182	15	124	43
201	>300	46	26	15	5
Subi	total	637	72	427	138
2.	Installations	in cement works			
50 101 201	100 200 300 >300		No Dat	a	
Subt	total				
3.	Installations	in ore-roasting	plants		
50 101 201	··· 100 ··· 200 ··· 300 ≻300				
Subt	total	-		_	_
Tota	al number of in	nstallations	91-91-91-91-91-91-91-91-91-91-91-91-91-9		
50	100	409	31	288	90
101 201	200)	182	15	124	43
	>300	46	26	15	5
Tota	al	637	72	427	138

# TABLE XXI. - Number of Installations

Capa Inst	city of allation MW	Total Number	Coal Fired	Oil Fired	'Gas Fired
1.	Installation	ns for electric	ity, heating and process		
50 101 201	100) 200) 300)	10			
	>300	6	1 (+1 peat fired)	4	-
Subt	otal	16	2	4	-
2.	Installation	ns in cement wo	rks		
50 101 201	100 200 300 >300				
Subt	otal				
3.	Installation	ns in ore-roast	ing plants		
50 101 201	100 200 300 >300				
Subt	otal				
Tota	al number of	installations			
50 101 201	··· 100) ··· 200) ··· 300)	10			
	>300	6	1 (+1 peat fired)	4	
Tota	.1	16	2	4	

Member State: EIRE

## Member State: ITALY

Capa Inst	acity of callation MW	Total Number	Coal Fired	Oil Fired	Gas Fired
1.	Installation	ns for electricity	7*		
50 101 201	100) 200) 300) >300	(34) (31) (5) (58)	(6) (7) (2) (12)	(28) (24) (3) (38)	(8)
Subt	total	(128)	(27)	(93)	(8)
2.	Installation	ns in cement works	3		
50 101 201	100 200 300 >300				
Subt	total				
3.	Installatio	ns in ore-roasting	g plants		
50 101 201	100 200 300 >300				
Subt	total			*******	
Tota	al number of	installations			
50 101 201	100 200 300 >300	(34) (31) (5) (58)	(6) (7) (2) (12)	(28) (24) (3) (38)	(8)
Tota	al	(128)	(27)	(93)	(8)

\* NB Numbers of boilers not numbers of plant

## TABLE XXIII. - Number of Installations

Capa Inst	acity of tallation MW	Total Number	Coal Fired	0il Fired	Gas Fired
1.	Installations	for electricity, h	eating and proce	288	
50 101 201	100 200 300 >300	2 1 - -	- 1 - -	2 - - -	- - -
Subi	total	3	1	2	
2.	Installations	in cement works			
50 101 201	100 200 300 >300				
Sub	total	_		_	-
3.	Installations	in ore-roasting pl	ants		
50 101 201	100 200 300 >300				
Subi	total				-
Tota	al number of i	nstallations			
50 101 201	100 200 300 >300	2 1 -	- 1 - -	2 - - -	- - - -
Tota	al	3	1	2	-

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# Member State: LUXEMBOURG

# TABLE XXIV. - Number of Installations

Member	State:	THE	NETHERLANDS

Capa Inst	acity of tallation MW	Total Number	Coal Fired	0il Fired	Gas Fired
1.	Installations	for electricity	y, heating and process		
50	100	105	1	25	79
101	200)	56	0	12	44
201	>300	55	6	4	45
Subt	total	216	7	41	168
2.	Installations	in cement works	5		
50 101 201	100 200 300 >300				
Subi	total	-	_		-
3.	Installations	in ore-roasting	g plants	<u></u>	
50 101 201	100 200 300 >300	2 - - -			
Subt	total	2		_	_
Tota	al number of i	nstallations		· · · · · · · · · · · · · · · · · · ·	
50	100	107	1	25	79
101 201	··· 200)	56	0	12	44
271	>300	55	6	4	45
Tota	al	218	7	41	168

## TABLE XXV. - Number of Installations

Capacity of Installation MW		Total Number	Coal Fired	Oil Fired	Gas Fired	
1.	Installation	ns for electricity	, heating and proce	288*		
50	100	(51)	(23)	(27)	(1)	
101	200	(37)	(36)	(1)	-	
201	300	(7)	(4)	(3)	-	
	>300	(68)	(52)	(16)	-	
Subt	otal	(163)	(115)	(47)	(1)	
2.	Installation	ns in cement works				
50 101 201	100 200 300 >300					
Subi	otal	21	21	_	-	
3.	Installation	ns in ore-roasting	; plant			
50 101 201	100 200 300 >300					
Subi	otal			<u> </u>		
Tota	al number of	installations	······································	<u> </u>		
50 101	··· 100 ··· 200					
201	··· 300 >300					
		·····				

## Member State: UNITED KINGDOM

figures without brackets: number of installations.

figures in brackets: number of individual flues.

# TABLE XXVI. - Number of Installations

# Member State: PORTUGAL

Capa Inst	acity of tallation MW	Total Number	Coal Fired	0il Fired	Gas Fired
1.	Installation	s for electricity	, heating and proce	:55	
50	100	2		1	1
101	··· 200)	3		2	1
201	>300	4	1	3	
Subi	total	9	1	6	2
2.	Installation	s in cement works	5		
50	100	2		2	
101	200	1		1	
201	>300	2		2	
Subt	otal	5		5	
3.	Installation	s in ore-roasting	g plant		
50	100				
101	200				
201	>300				
Subt	cotal				
Tota	al number of :	installations			
50	100	4		3	1
101	200)	6		5	1
201	>300	4	1	3	
Tota	al	14	1	11	2

# TABLE XXVII. - Number of Installations

Capa Inst	acity of tallation MW	Total Number	Coal Fired	0il Fired	Gas Fired
1.	Installation	ns for electricity	**		
50 101 201	100 200 300 >300	(14) (16) (8) (33)	(4) (8) (3) (20)	(10) (7) (4) (12)	(0) (1) (1) (1)
Subt	total	(71)	(35)	(33)	(3)
2.	Installation	ns in cement works	, <u>, , , , , , , , , , , , , , , , , , </u>		
50 101 201	100 200 300 >300				
Subt	total				
3.	Installation	ns in ore-roasting	; plant		
50 101 201	100 200 300 >300				
Subt	total				· · · · · <u>· · · · · · · · · · · · · · </u>
Tota	al number of	installations			
50 101 201	100 200 300 >300	(14) (16) (8) (33)	(4) (8) (3) (20)	(10) (7) (4) (12)	(0) (1) (1) (1)
Tota	 1	(71)	(35)	(33)	(3)

Member State: SPAIN

\* NB Numbers of boilers not numbers of plant

# $\frac{\text{TABLE XXVIII. - Number of Installations where Particulate,}}{\frac{\text{SO}_2 \text{ and NO}_x \text{ Emissions are Measured Continuously}}}$

Member State: BELGIUM

Type of	Emission				
Installation	Particulate		so2		NOx
1. Installations for	electricity,	heating	and	process	
Coal fired					1
Oil fired					
Gas fired					
Subtotal					1
2. Installations in	cement works				
Coal fired	2				
Oil fired					
Gas fired					
Subtotal	2				
3. Installations in	ore-roasting	plant			
Coal fired					
Oil fired					
Gas fired					
Subtotal				- <u> </u>	
4. Total number of i	nstallations				
Coal fired	2				1
Oil fired					
Gas fired					
Total	2				1

### TABLE XXIX. - Number of Installations where Particulate, SO<sub>2</sub> and NO<sub>2</sub> Emissions are Measured Continuously

# Emission Type of NOx Installation SO2 Particulate 1. Installations for electricity, heating and process Coal fired Oil fired Gas fired 9 Subtotal 22 8 2. Installations in cement works Coal fired Oil fired Gas fired Subtotal 3. Installations in ore-roasting plant Coal fired **Oil fired** Gas fired Subtotal 4. Total number of installations Coal fired Oil fired Gas fired 22 Total 9 8

Member State: DENMARK

# TABLE XXX. - Number of Installations (Boilers\*) whereParticulate, S02and NOContinuously

NO<sub>x</sub>

~470

~238

~17

~725

2

2

~472

~238

~17

~727

### Emission Type of Installation so<sub>2</sub> Particulate 1. Installations for electricity, heating and process Coal fired ~480 ~470 0il fired ~255 ~238 Gas fired ~735 ~708 Subtotal 2. Installations in cement works Coal fired 32 3 0il fired Gas fired Subtotal 32 3 3. Installations in ore-roasting plant Coal fired 0il fired

#### Member State: FEDERAL REPUBLIC OF GERMANY

Gas fired

Subtota1

Coal fired

0il fired

Gas fired

Total

\* In this case the figures refer to numbers of boilers not numbers of installations.

~473

~238

~711

5

~512

~255

~772

4. Total number of installations

# $\frac{\text{TABLE XXXI.} - \text{Number of Installations where Particulate,}}{\frac{\text{SO}_2 \text{ and NO}_x \text{ Emissions are Measured Continuously}}$

Member State: GREECE

				Emissi	lon		
Inst	ype of tallation		Particulate		50		NO
					502		x
1.	Installations	for	electricity,	heating	and	process	
Coa	l fired						
0i1	fired						
Gas	fired						
Sub	total					· · · · · · · · · · · · · · · · · · ·	
2.	Installations	in	cement works	······			
Coa	l fired		<u></u>			<u></u>	
0i1	fired						
Gas	fired						
Subi	total						
3.	Installations	in o	ore-roasting	plant			
Coal	l fired						
0i1	fired						
Gas	fired						
Subt	total		·······				
4.	Total number of	of in	nstallations				
Coal	fired						
011	fired						
Gas	fired						
Tota	1		0		0		0

# TABLE XXXII. - Number of Installations where Particulate, $SO_2$ and $NO_x$ Emissions are Measured Continuously

Member State: FRANCE

Ty	vpe of			Emiss	ion		
Inst	allation		Particulate*		so <sub>2</sub>		NOx
1.	Installations	for	electricity,	heating	and	process	
Coal	fired	<u></u>	72		1		
<b>0i</b> 1	fired		427				
Gas	fired		138				
Subt	otal		637				
2.	Installations	in d	cement works				
Coal	fired						
0i1	fired			No Da	ata		
Gas	fired						
Subt	otal						
3.	Installations	in d	ore-roasting	plant			
Coal	fired						
0i1	fired						
Gas	fired						
Subt	otal		<u></u>		<u>-</u>		
4.	Total number of	of in	nstallations				
Coal	fired						
0i1	fired						
Gas	fired						
Tota	1		≥693		1		

\* Installations are generally equipped with optical instruments and 40 beta gauges are also installed.

# $\frac{\text{TABLE XXXIII. - Number of Installations where Particulate,}}{\frac{\text{SO}_2 \text{ and NO}_x \text{ Emissions are Measured Continuously}}$

				- <u> </u>			
T	ype of			Emissi	Lon		
Inst	tallation		Particulate		so <sub>2</sub>		NOx
1.	Installations	for	electricity,	heating	and	process	
Coa	l fired						
<b>0i</b> 1	fired						
Gas	fired						
Subi	total						
2.	Installations	in	cement works				
Coa	l fired						
011	fired						
Gas	fired						
Subi	total					· · · · · · · · · · · · · · · · · · ·	- <u>lava</u> r <u>a.</u> - <u>i</u> <u>i</u> .
3.	Installations	in	ore-roasting	plant			
Coa	l fired		<u>, , , , , , , , , , , , , , , , , , , </u>				
<b>0i</b> 1	fired						
Gas	fired						
Subt	total						
4.	Total number of	of in	nstallations				
Coal	l fired						
011	fired						
Gas	fired						
Tota	al	- <u>+</u>	0	···	0		0

Member State: EIRE

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# $\frac{\text{TABLE XXXIV.} - \text{Number of Installations where Particulate,}}{\frac{\text{SO}_2 \text{ and } \text{NO}_x \text{ Emissions are Measured Continuously}}$

Member State: ITALY

Т	ype of	]	Emission					
Ins	tallation	Particulate*	so <sub>2</sub>	NO				
1.	Installations	for electricity, he	ating and process	3				
Coa	l fired							
011	fired							
Gas	fired							
Sub	total							
2.	Installations	in cement works						
Coa	1 fired							
0i1	fired		、					
Gas	fired							
Sub	total							
3.	Installations	in ore-roasting play	nt					
Coa	1 fired							
011	fired							
Gas	fired							
Sub	total	A - E - Marine - B - E - E - E - E - E - E - E - E - E		<u> </u>				
4.	Total number o	of installations						
Coa	l fired							
011	fired							
Gas	fired							
Tot	al	0	0	0				

\* New power plant are equipped with opacimeters for optimising combustion conditions.

# $\frac{\text{TABLE XXXV.} - \text{Number of Installations where Particulate,}}{\underline{\text{SO}_2 \text{ and NO}_x \text{ Emissions are Measured Continuously}}}$

Member State: LUXEMBOURG

Type of	Emission				
Installation	Particulate		so <sub>2</sub>		NOx
1. Installations for	electricity,	heating	and	process	
Coal fired			<u> </u>		
Oil fired					
Gas fired					
Subtotal	,				
2. Installations in	cement works				
Coal fired					
Oil fired					
Gas fired					
Subtotal					
3. Installations in	ore-roasting	plant			
Coal fired					
Oil fired					
Gas fired					
Subtotal					
4. Total number of i	nstallations				
Coal fired	······································				
Oil fired					
Gas fired					
Total	0		0		0

# $\frac{\text{TABLE XXXVI. - Number of Installations where Particulate,}}{\frac{\text{SO}_2 \text{ and NO}_x \text{ Emissions are Measured Continuously}}}$

Member State: THE NETHERLANDS

T	vpe of			Emissi	Lon		
Inst	allation		Particulate		so <sub>2</sub>		NOx
1.	Installations	for	electricity,	heating	and	process	
Coa	l fired		8		3		
011	fired						
Gas	fired						
Subt	total		8		3		10
2.	Installations	in	cement works				
Coal	fired						
0i1	fired						
Gas	fired						
Subt	total		2,4°,₩,-,,,,,,,,_,_,_,,_,,,,,,,				
3.	Installations	in	ore-roasting	plant			
Coal	fired						
<b>0i</b> 1	fired						
Gas	fired						
Subt	otal						
4.	Total number of	of i	nstallations			<u></u>	
Coal	fired		8		3		
011	fired						
Gas	fired						
Tota	1		8		3		10

# $\frac{\text{TABLE XXXVII. - Number of Installations/Flues where}}{\frac{\text{Particulate, SO}_2 \text{ and NO}_{\text{x}} \text{ Emissions are}}{\text{Measured Continuously}}}$

Member State: UNITED KINGDOM

Emission					
Partico	ulate		so <sub>2</sub>	NOx	
ricity,	heating	and	process*		
115					
20					
1					
136			. <u> </u>	<u></u>	
cement	works				
21					
21					
ore-roa	sting pla	ant		<u> </u>	
			- <u></u>		
nstalla	tions				
				<u></u>	
	Particular ricity, 115 20 1 136 cement 21 21 ore-roa	Particulate ricity, heating 115 20 1 136 cement works 21 21 ore-roasting pla	Emis Particulate ricity, heating and 115 20 1 136 cement works 21 21 ore-roasting plant nstallations	Emission         Particulate       SO2         ricity, heating and process*         115         20         1         136         cement works         21         21         ore-roasting plant	

\* In this case the figures refer to numbers of individual flues not numbers of installations.

# TABLE XXXVIII. - Number of Installations where Particulate, $SO_2$ and $NO_x$ Emissions are Measured Continuously

Member State: PORTUGAL

Type of	Emission				
Installation	Particulate	so <sub>2</sub>	N		
1. Installations for	r electricity,	heating and	process		
Coal fired		<u> </u>			
Oil fired					
Gas fired					
Subtotal		·····			
2. Installations in	cement works				
Coal fired					
Oil fired					
Gas fired					
Subtotal	1				
3. Installations in	ore-roasting p	lant			
Coal fired					
Oil fired					
Gas fired					
Subtotal					
4. Total number of i	installations				
Coal fired					
Oil fired					
Gas fired					
Total	1	0			

# $\frac{\text{TABLE XXXIX. - Number of Installations where Particulate,}}{\frac{\text{SO}_2 \text{ and NO}_x \text{ Emissions are Measured Continuously}}$

Member State: SPAIN

Type of		Emission				
Installation	Particulate		so <sub>2</sub>		NOx	
1. Installations fo	r electricity,	heating	and	process		
Coal fired					3	
Oil fired						
Gas fired						
Subtotal	58*		22		3	
2. Installations in	cement works					
Coal fired						
Oil fired						
Gas fired						
Subtotal	· · · · · · · · · · · · · · · · · · ·					
3. Installations in	ore-roasting	plant				
Coal fired						
Oil fired						
Gas fired						
Subtotal						
4. Total number of	installations	· · · · · · · · · · · · · · ·				
Coal fired				·······		
Oil fired						
Gas fired						
 Total	58*		22		3	

\* including 1 Beta gauge

### 5. SURVEY OF MEASUREMENT COSTS

The total cost to individual installations for compliance with the Commission's proposals can be broken down into investment, installation and operating costs. The relative contribution of each depends to some extent on whether an extractive or in-situ system is selected but also on the measurement principle and on site specific details.

### 5.1 Investment Costs

Investment costs are to some extent dependent on the measurement principle of the instrument, but prices are likely to reflect existing market forces as well as manufacturing costs. Investment costs should not be considered in isolation; for instance a sophisticated instrument with in-built maintenance functions might represent a higher investment cost than a simpler instrument but have reduced maintenance costs. If a multicomponent analyser can carry out the measurement functions of several single component analysers the investment costs are lower, however, if the multicomponent analyser were at an early stage of development there might be a maintenance cost penalty. Calibration costs may be higher for in-situ instrumentation than extractive equipment.

The investment costs listed in Table XL are for equipment only and do not include the cost of installing the instrument. Installation costs are very dependent on site specific details; for example, they can be extremely high if special access platforms must be provided for a high stack. The same access platform can be used for several instruments so installation costs should be considered in connection with a monitoring package rather than a single instrument. It should also be noted that these considerations apply mainly to existing plant and that the incremental cost for installing emission monitoring equipment on a new installation would be relatively low.

### 5.2 Maintenance Costs

Regular maintenance, by trained personnel is necessary to ensure that an emission measuring device functions correctly and produces results of acceptable accuracy. Maintenance costs are best expressed in terms of manpower requirements. FRG experience has shown that manpower requirements can vary from 20-60 instrument/man/yr. On average, these are about 25 h yr<sup>-1</sup> for transmissometers and about 100 h yr<sup>-1</sup> for a single component extractive device, including sampling and conditioning, with an extra 50 h yr<sup>-1</sup> for each additional instrument using a shared extraction system. The total manpower requirement for a monitoring package for particulates,  $SO_2$ , NO and  $O_2$  can therefore be estimated at 225-250 h yr<sup>-1</sup>. These figures make the assumption that suitably qualified personnel carry out the maintenance and that adequate laboratory facilities and equipment are available. These requirements should be taken into account at the planning stage in order to minimise operating costs.

Calibration against an appropriate manual reference method by an authorised body is required at installation and thereafter at intervals of no more than 5 years. The estimated costs of these calibration measurements, including evaluation and programming of calibration factors, are:

- particulates 3500 11500 ECU depending on plant size (on a basis of approximately 15 comparison measurements)
- gaseous components 6000 ECU to calibrate  $SO_2$ , NO,  $O_2$  by ca 30 comparison measurements

Once a year a functional test of the instrument has to be performed. In this test the sampling line and the response time of the whole measuring device have to be checked. Possible sources of interference must be identified and corrected. In-situ instrument optics have to be cleaned and the calibration tested at a clean reference path.

The costs for an annual check will be about 230 ECU's for an extractive instrument and about 320 ECU's for an in-situ instrument. The latter price is higher as very often the instrument has to be demounted for test purposes.

### 5.3 Case Study

As shown in Table XL, the total investment cost for equipment to measure particulates,  $SO_2$ , NO,  $O_2$  and carry out data treatment would be in the range ECU 44000-65000 based on the use of a transmissometer for particulate measurement and an extractive system for the gaseous components. Alternatively if a Beta attenuation analyser were used for particulate measurement, in connection with in-situ instruments for the gaseous components the total investment cost would be in the range ECU 60000-75000.

There would be additional costs for calibration of approximately ECU 10000 for smaller boilers and ECU 18000 for larger boilers.

It is interesting to relate the total investment cost to other costs connected with boiler operation; a figure of 1% of the annual fuel cost has been suggested as an initial estimate and a basis for further discussion. The following examples<sup>92</sup> give the order of magnitude of this figure for several plant sizes, on a given basis:

- (a) for a combustion plant with 50 MW thermal input and a yearly operation equivalent of 2000 h full load, 1% of the annual fuel cost would be up to ECU 14000-28000. Such a plant would be one of the smallest affected by the proposed Directive;
- (b) for a larger combustion plant with 300 MW thermal input and a yearly operation equivalent of 4000 h full load, 1% of the fuel costs would be up to ECU 160000-320000 a year.
- (c) for a rather large combustion plant with 1000 MW thermal input and a yearly operation equivalent of 6000 h full load, 1% of the annual fuel costs would be up to ECU 800000-1600000.

The estimated total investment and calibration costs are expressed below in % annual fuel costs, for the three plant sizes:

(a) 2-6% of the annual fuel cost;
(b) 0,2-0,6% of the annual fuel cost;
(c) 0,04-0,1% of the annual fuel cost.

The investment and calibration costs for large boilers are less than 1% of the annual fuel cost.

The investment and calibration costs for the smallest boilers to be affected by the proposed Directive are more significant, therefore the possibility of multiplexing 2 or 3 boilers with one set of extractive instruments to measure the gaseous components should be considered.

	Measurement Principle	Investment Cost of One Instrument (ECU)
Particulates	Opacimeters	2700 - 4000
	Transmissometer	6000 - 12000
	Beta attenuation analyser	18000 - 25000
	Multicomponent - dust/SO <sub>2</sub>	26000 - 32000
Gaseous components	Extractive (NO, SO <sub>2</sub> and O <sub>2</sub> )	32000 - 46000
	In-situ	
	- photometers (NO and/or SO <sub>2</sub> )	17000 - 20000
	- electrochemical (SC	0 <sub>2</sub> ) 14000 - 35000
	- zirconia cell/probe	1700 - 4300
Data Treatment		5500 - 18000

# TABLE XL. - Investment Costs

### 6. CONCLUSIONS AND RECOMMENDATIONS

To meet the requirements of CEC Contract No. 84-B-6642-11-005-11-N, the contractors, IRCHA, TÜV Rheinland and WSL, studied methods of sampling and analysis for  $SO_2$ ,  $NO_x$  and particulate matter in the exhaust gases of large combustion installations and, where possible, the objectives have been met. The group have come to the following conclusions:

- Existing and proposed emission regulations and limits have been documented for the Member States, including Spain and Portugal. It has been found that there are considerable differences in the national regulations and in the way which emission limits are specified and controlled. Harmonisation of the national legislation, as outlined in the Commission's proposals, is necessary. A survey of the applied measurement methods has been undertaken as part of this objective. There is a considerable number of manual and continuous measurement methods already in use. In some Member States these are already incorporated in national guidelines.

In general one manual method will become the primary reference method for continuous measuring devices for each component. The calibration of continuous particulate measuring devices, such as transmissometers will use existing guidelines until a special ISO standard for manual measurement and calibration purposes becomes available. The proposed method is expected to be circulated in preparation for an ISO/DP within the next few months. A second ISO/DP for the manual measurement of SO<sub>2</sub> has been prepared as is at the ballot stage. It is highly desirable that a further ISO standard is developed as a primary reference method for the measurement of  $NO_x$ . Calibrations should be carried out by suitably trained personnel.

Continuous measurement methods are preferable, although the error in the data from some continuous measurement systems is greater than that from the most accurate manual methods. It is highly desirable to have continuous data even if there is some loss in precision. The measurement methods for gases and particulates can be of the extractive on-line or in-situ type. At present, the former are in general use for gases and the latter for particulates.

The extractive on-line devices may be more cost-effective for existing plant than in-situ instruments, however, for new plant specifically designed to incorporate emission measuring equipment, in-situ devices may be preferred. In general in-situ instruments for gas measurements are at an earlier stage of development than extractive instruments, but they have considerable advantages providing that an acceptable quality of measurement can be achieved.

It is essential that standardised suitability tests of the type already in use in the FRG, which include laboratory testing and testing in the field, are defined and applied in all Member States. National approval laboratories using common suitability test procedures should be established as soon as possible.

Continuous measurement devices for volume flow and water content are still under development, but two systems for volume flow measurement have already met the required standards in the FRG. However, these data can often be obtained by calculation from process parameters.

- At present, emissions of sulphur dioxide are determined by calculation in many Member States, but such calculations do not take into account variations in the retention of sulphur in the system. Where sulphur dioxide abatement systems are in operation determination of sulphur dioxide emissions by calculation is unsatisfactory.
- An essential function of the measurement system is to handle the incoming data in a systematic way and to present the output in a way which facilitates evaluation of plant operating performance and compliance with emission limits. The data handling system should be subjected to suitability testing in accordance with a common certification procedure for all Member States.
- The number of installations of greater than 50 MW thermal input has been determined, as far as possible, for each Member State. No brickworks in this category were identified. Reliable data have been obtained for

power plant greater than 300 MW thermal input in all Member States and adequate information has been collected for power plant in the 50-300 MW The data for industrial plant is less reliable because of range. difficulties in identifying the large number of plant and differentiating between single boilers and installations. The latter point is extremely important because it would usually be desirable to equip individual boilers rather than stacks with measurement systems, even where several boilers share a common flue. In some cases it may be possible to reduce costs by multiplexing extractive measurement devices for gaseous The data available for installations in cement works is components. sparse because the European cement manufacturers object to the proposed Directive and were not always willing to supply information. The numbers of cement works and also ore-roasting plant are, however, small compared with the numbers of industrial boilers and these installations are in the smaller size range.

The number of boilers equipped with continuous measurement devices varies widely from Member State to Member State and from pollutant to pollutant. The absence of comprehensive statistics on industrial boiler plant in many Member States makes detailed analysis of the data impossible.

In connection with the continuous measurement of emissions, cost data have been presented for investment, installation, calibration and Investment costs are to some extent dependent on the maintenance. measurement principle of the instrument, but prices are likely to reflect existing market forces as well as manufacturing costs. In some situations the installation costs can be higher than the investment In general, the cost of the instruments required for compliance costs. with the proposed Directive is similar for large plant and small plant. It should be noted that the investment costs for different installations are related to the maintenance and calibration costs. The total costs for smaller plant are relatively significant and the possibility of multiplexing instruments should be taken into account.

If the Commission's proposals for the measurement of  $SO_2$ ,  $NO_x$  and particles were implemented it would produce comparable data for Member States, which would make it possible to assess progress in reducing total emissions.

An efficient emission monitoring system must fulfil the following conditions.

- The individual instruments will be calibrated using the appropriate reference methods, to ensure comparability of data with other continuous measurement methods.
- The instruments and data treatment devices should have undergone suitability tests which guarantee that they fulfil minimum requirements accepted in all Member States. The institutions authorised to perform these tests will be listed by national or EC authorities.
- Manual emission measurements and the calibration of continuous measurement devices will be performed only by authorised institutions.

Some of these recommendations exceed the scope of the Commission's proposals and indicate areas where further investigation may be required.

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## ANNEXE 1. - INSTRUMENT MANUFACTURERS AND PERFORMANCE DATA

## Measurement Devices for Particulates

## In-situ Photometric Devices

for concentration measurement:

DURAG	D-R 110) no longer in production
DURAG	D-R 280)
DURAG	D-R 280-10
SICK	RM 3g) no longer in production
SICK	RM 4 )
SICK	RM 41/RM 41-3
SICK	GM 21

for opacity measurement:

DURAG	D-R 116	no	longer	in	production
DURAG	D-R 216				
SICK	RM 61				
THERMOELECTRON					

Table Al shows the performance characteristics of the instruments which are still in production.

## Light Scatter Devices

WESGLAGLE ELECTRONICS	FORDUST
SIGRIST	Photometer KTN
PILLARD	OPASTOP GP, 1000 H

## Beta Attenuation Devices

VEREWA,	Betastaubmeters F50 and F60
	(no longer in production
	F904 (currently undergoing
	suitability testing in the FRG)
ENVIRONNEMENT	MPSI 41V

Performance characteristics for the Verewa F50 and the Environnement MPSI 41V are given in Table A2.

Obscuration Devices

KENT AIRFLOW DEVELOPMENTS Dust Monitor 2010 (CERL II) SEROP

## Measurement Devices for SO<sub>2</sub> and/or NO<sub>x</sub>

## Non-dispersive Infra red

Extractive

BENDIX	MODEL 501	so <sub>2</sub>
KENT	INFRAGAS 400	so <sub>2</sub>
HORIBA INSTRUMENTS	ESDA 813	so <sub>2</sub>
HORIBA INSTRUMENTŚ	ENOA 812	NOx
HARTMANN & BRAUN	URAS 3G	so <sub>2</sub>
HARTMANN & BRAUN	URAS 3E	so <sub>2</sub>
MAIHAK	UNOR 6N-R	so <sub>2</sub>
MAIHAK	UNOR 6N-F	so <sub>2</sub>
MAIHAK	unor 4n	NO <sub>x</sub>
MAIHAK	UNOR 6N	NO <sub>x</sub>
SIEMENS	ULTRAMAT 2	so <sub>2</sub>
SIEMENS	ULTRAMAT 3	so <sub>2</sub>
SIEMENS	ULTRAMAT 32	SO <sub>2</sub> , NO <sub>x</sub>
BECKMAN	MODEL 864	so <sub>2</sub>
BECKMAN	MODEL 865	$so_2^{-}$ , NO
LEYBOLD HERAEUS	IR BINOS	S0 <sub>2</sub> , NO

DIAMANT 6000	so <sub>2</sub> , no <sub>x</sub>
SAPHIR 816	so <sub>2</sub>
MODEL 450	so <sub>2</sub> , no
MODEL 330	so <sub>2</sub> , no
1000 SERIES	so <sub>2</sub> , no
	DIAMANT 6000 SAPHIR 816 MODEL 450 MODEL 330 1000 SERIES

## Non-dispersive Ultra Violet

Extractive

LEYBOLD-HERAEUS	UV BINOS	so <sub>2</sub>	
HARTMANN & BRAUN	RADAS IG	NO <sub>x</sub> ,	NO
HARTMANN & BRAUN	RADAS IE	NO <sub>x</sub> ,	NO

In-situ

SICK	GM 21	so <sub>2</sub>	
THERMOELECTRON		so <sub>2</sub> ,	NO

Performance data for these instruments, where available, are given in Tables A3 and A4.

## Second Derivative Ultra-violet - In-situ

LEAR-SIEGLER	SM 810	so <sub>2</sub> no

Dispersive Ultra-violet

Extractive

ANACON	M207	so <sub>2</sub> ,	NO <sub>x</sub>
TELSEC PROCESS ANALYSIS	OKOMETER	so <sub>2</sub> ,	NO2

	In-situ		
	MEASUREX	MODEL 2225	so <sub>2</sub> , no
	Performance data are given in Tab	le A5.	
Inter	ferometry		
	SERES	SIMS 5600	so <sub>2</sub> , no <sub>x</sub>
	Measurement Devi	ces for SO <sub>2</sub> Only	
Fluore	escence		
	MELOY	SA 700	
	THERMOELECTRON	MODEL 43	
	COSMA	ONYX 520	
	Performance data are given in Tab	le A6.	
Electi	cochemical		
	THETA SENSOR	MODEL 1050	
	WESTINGHOUSE	MODEL 260	
Flame	Photometry		
	MELOY	SA 185-2 FPD	
	MELOY	SA 165	
Conduc	tometry		
	Wösthoff	MIKROGAS-MSK-SO <sub>2</sub> -E1	

## Measurement Devices for NO<sub>x</sub> Only

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## Chemiluminescence

BECKMAN	MODELS 951 and 955
MELOY	NA 520-2B
THERMOELECTRON	MODEL 10
SIGNAL	MODEL 4000
BENDIX	MODEL 8101-C
COSMA	TOPAZE 810/820
ENVIRONNEMENT	AC 20 M

The performance data for these devices where available are given in Table A7.

## Measurement Devices for Oxygen

## Zirconium Oxide Cell

In-situ:

DRAYTON MACDHUI	LT, ST, HT
HARTMAN & BRAUN	CGRGMI
INSTRUMATIC	401
KENT	Z-FG
LAND COMBUSTION	
SYBRON	700 SERIES
WESTINGHOUSE	218/450/132
TELSEC	TPA 422

Extractive:

ANALYSIS AUTOMATION	450
ANACON	M78
BECKMAN	7010
HITECH	HITOX
TELSEC	TPA 424
COSMA	QUARTZ 650

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Polarographic:

## BECKMAN MODELS 7003/741

Paramagnetic

HARTMAN & BRAUN	MAGNOS 3
MAIHAK	OXYGOR 6N
HORIBA	EOXA-816
SIEMENS	OXYMAT 2 & 3
COSMA	DIAMANT 6002
ENVIRONNEMENT	SEMAXOX
HERMANN MORITZ	MU10
SERVOMEX	750A/760A

Thermomagnetic

HARTMAN & BRAUN	MAGNOS 5T
LEEDS & NORTHRUP	7863
SIEMENS	OXYMAT 1

## Flow Measurement Devices

Vortex Shedding

BESTOBELL	MODEL 3167
SCHEME ENGINEERING	VF 588
TECHNITRON	

Insertion Turbine

DELTA CONTROLS	
MAURER	MAUFLO SR and FR SERIES
QUADRINA	PROBEFLO and Q-FLO

Pneumatic Bridge

TELEDYNE HASTINGS - RAYDIST AFI and AFI-L SERIES

Thermal Dispersion

KURTZ	SER	IES !	515/5	55	
FCI	LT8	1/LT	70		
SARASOTA	FM	711,	713,	771,	773

Averaging Pitot

INDUSTRIAL MEASUREMENT DEVICES	BETA PROBE
TAYLOR INSTRUMENTS	FLOBAR ELEMENT
LOBA	DELTA TUBE
AGAR	
STANDARD CORPORATION	ANNUBAR ANR75, ANF86

Ultrasonic

PANAMETRICS	
SICK	

MODEL 7000/7100 VMA

## Data Treatment Devices

- integration devices with simple classification

DURAG	D-IG	260
SICK	MR-2	

- integration devices with classification and reference value calculation

DURAG	D-MS	385		
MAIHAK	MEAC	EDASK	and	EDASR
NIS				
SICK	MR-3			
SIEMENS	SAE			
RWE	ZEUS			

TABLE Al Performance	Characteristics of In-s	itu Photometric Particuls	ate Measurement Devices
	DURAG D-R 280-10	DURAG D-R 216	SICK RM 41
Source of performance data	German suitability test	German suitability test	German suitability test
Measurement quantity	Particulate concentration	Smoke density	Particulate concentration
Period of unattended operation	4-6 weeks	2-4 weeks	2 weeks
Influence of maladjustment of light beam	<2% FS up to 0.5°	<2% FS up to 0.5°	<2% FS up to 0.5° or up to 0.1° depending on measuring range
Temperature dependence of zero point [-20 -50°C]	<2% FS	<±1% FS	<0.5% FS
Temperature dependence of the sensitivity	<0.23% FS/10°C		<0.5% FS/10°C
Zero drift	<1% FS/3 months	<1% FS/3 mths	Not detectable
Sensitivity drift	<2% FS/3 months	Not detectable	<2% FS/3 months
Ambient Temperature Range	-30 -55°C	-30 -55°C	-30 -55°C
Reproducibility	54-57	101	30–78
Availability	>95%	>95%	286

**A8** 

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	SICK GM 21	SICK RM 61
Source of performance data	German suitability test	German suitability test
Measurement quantity	Particulate concentration	Smoke density (opacity)
Period of unattended operation	8-12 weeks	12 weeks
Influence of maladjustment of light beam	<2% FS up to 0.5° depending on measuring range	2% FS up to 1°
Temperature dependence of zero point [-20 -50°C]	<±2.8% FS	1.0% FS
Temperature dependence of the sensitivity [-20 -50°C]	<2% FS	I
Zero drift	Not detectable	2% FS/3 months
Sensitivity drift	<2% FS/period of unattended operation	
Ambient Temperature Range	-20 -50°C	-30 -50°C
Reproducibility	91-106	96
Availability	266	38%

TABLE A1. - cont'd

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	VEREWA F50	ENVIRONNEMENT MPSI 41V
Source of performance data	German suitability test	<ol> <li>Manufacturer's literature.</li> <li>French operating experience.</li> <li>French suitability test.</li> </ol>
Measurement ranges	0-20 up to 0-350 mg/Nm <sup>3</sup>	2-2000 mg/Nm <sup>3</sup>
Availability	x06<	80%2
Period of unattended operation	1 week	1 month <sup>3</sup>
Reproducibility	12	In the range 40-1100 mg m <sup>-3</sup> max relative sd: 25%
Nominal value deviation of sample volume	<8%	
Ambient temperature range	-10 -40°C	$-20 - 50^{\circ}C^{3}$
Zero drift	<2% FS/week	
Sensitivity drift	<4% FS/week	2%/10 days @ 40 mg m <sup>-3 3</sup> 1%/10 days @ 150 mg m <sup>-3</sup>
Precision		$\pm 10\%$ (for 0.1-22 mg of deposited dust) <sup>1</sup>

TABLE A2. - Performance Characteristics of Beta Ray Absorption Instruments

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	HARTMANN & BRAU	N URAS 3G/URAS 3E	MAIHAK UNO	R 6N-R	MAIHAK UNOR 6N	MALHAK UNOR 4N
Source of data	German suf	tability test	German suitab	ility test	German suitability test	German suitability test
Type	Extr	active	Extract	1ve	Extractive	Extractive
Tested ranges	0- 500 0-4000	mg/Nm <sup>3</sup> SO <sub>2</sub> mg/Nm <sup>3</sup> SO <sub>2</sub>	0-1000 mg/	m <sup>3</sup> SO <sub>2</sub>	0-400 mg/m <sup>3</sup> NO <sub>X</sub> 0-4000 mg/m <sup>3</sup> NO <sub>X</sub>	0 400 mg/m <sup>3</sup> NO <sub>X</sub> 0-2000 mg/m <sup>3</sup> NO <sub>X</sub>
Availability	· 100% (ov	er 3 mths)	>96% (over 3	months)	>95% (over 3 mths)	>95% (over 3 mths)
Period of unattended operation		2 wks	8 day	Ø	8 days	8 days
Reproducibility	OE	-106	33-5	2	51-116	64-103
Influence of barometric pressure variation	1% FS/	10 m bar	<0.5% FS/1	5 m bar	Not detectable	Not detectable
Influence of sample flowrate variation	Not de	tectable	<0.5% FS/	20 1/h	Not detectable	Not detectable
Lower detection limit	2	% FS	1.37	FS	1.5% FS	1.5% FS
Ambient temp range	5	45°C	0-40	U	0-45°C	0.45°C
Temp dependence of zero	2% F	s/10°C	Not detec	table	Not detectable	Not detectable
Temp dependence of sensitivity	2% F	s/10°C	Not detec	table	Not detectable	<3% FS
Zero drift	<2% FS/wk		1% FS/8 days		<2% FS/8 days	<2% FS/8 days
Sensitivity drift	<4% FS/wk		1.5% FS/8 days		<3.5% PS/8 days	<3.5% FS/8 days
Response time	(%06) s 06		<200 s (90%)		<80 s (90%)	<100 s (90%)
Interference	460 mg/m <sup>3</sup> CO 0 470 mg/m <sup>3</sup> NO 0 190 mg/m <sup>3</sup> NO <sub>2</sub> 2 15 vol% CO <sub>2</sub> 0 H <sub>2</sub> O saturated f 276 K a	.5% FS .5% FS % FS .5% FS 4% FS if inter- erence taken into ccount during alibration	<pre>1 vol % C0 5800 mg/m<sup>3</sup> N0 860 mg/m<sup>3</sup> N0 15 vol % C0 H<sub>2</sub>0 saturated 303 K</pre>	Not detectable <0.5% FS <0.5% FS Not detectable Not detectable	0.9 vol % CO ) 14.9 vol % CO <sub>2</sub> ) 2 g/m <sup>3</sup> SO <sub>2</sub> )1-2.5% FS H <sub>2</sub> O saturated ) 300 K )	0.9 vol X C0 ) 14.9 vol X C0 ) 2 g/m <sup>3</sup> So <sub>2</sub> )0.5-2X FS H <sub>2</sub> 0 satrated ) 300 K )

TABLE A3. - Performance Characteristics of NDIR Instruments for  $\mathrm{SO}_2$  and/or  $\mathrm{NO}_{\mathrm{X}}$ 

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	SIEMENS ULTRAMAT 2	SIEMENS ULTRAMAT 32	BENDIX MODEL 501	HORIBA ESDA 813	HORIBA ENOA 812
Source of data	German suitability test	German suitability test	Manufacturer's literature	Manufacturer's literature	Manufacturer's literature
Type	Extractive	Extractive	Extractive	Extractive	Extractive
Tested ranges or available ranges	0-5000 mg/Nm <sup>3</sup> SO <sub>2</sub>	0-100, 0-500, 0-2000 mg/Nm <sup>3</sup> SO <sub>2</sub>	0-2000 ppm	0-100/200 ppm (min) S0 <sub>2</sub> 0-1000/2000 ppm (max) S0 <sub>2</sub>	0-100/200 ppm NO <sub>X</sub> (min) 0-1000/2000 ppm NO <sub>X</sub> (max)
Availability	>95% (over 3 months)	>95% (over 3 months)			
Period of unattended operation	8 days	8 days			
Reproducibility	82-187	52-257			
Influence of barometric pressure variation	<0.5% FS/10 m bar	Not detectable			
Influence of sample flowrate variation	<0.5% FS/10 1/h	Not detectable			
Lower detection limit	37. FS	1% FS			
Ambient temp range	5-45°C	5-45°C			
Temp dependence of zero	Not detectable	Not detectable			
Temp dependence of sensitivity	Not detectable	Not detectable	1% FS/10°C		
Zero drift	<0.5% FS/8 days	1-2% FS/8 days	3.5% FS/wk	2% FS/wk	2% FS/wk
Sensitivity drift	<1.5% FS/8 days	1-3.5% PS/8 days		2% FS/wk	2% FS/wk
Response time	<130 s (90%)	75-90 s (90%)		4 mins (90%)	1 min (90%)
Interference	1.5 vol % C0 ) 15 vol % C0 ) 5800 mg/m <sup>3</sup> NO) <0.5% FS 880 mg/m <sup>3</sup> NO <sub>2</sub> ) . H <sub>2</sub> 0 saturated) 303 K )	1 vol Z CO ) 14 vol Z CO ) 248 mg/m <sup>3</sup> NO ) 0-3.5Z FS 32 mg/m <sup>3</sup> NO <sub>2</sub> ) H <sub>2</sub> O saturated) 300 K )			

TABLE A3. - cont'd

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		BECKMAN MODEL 864	BECKMAN MODEL 865	KENT INFRA GAS 400	ANALYSIS AUTOMATION MODEL 450	TELSEC PROCESS ANALYSIS, MODEL 330	CODEL 1000 SERIES
	Source of data	Manufacturer's 11terature	Manufacturer's literature	Manufacturer's literature	Manufacturer's literature	Manufacturer's literature	Manufacturer's literature
	Type	Extractive	Extractive	Extractive	In-situ	In-situ	In-situ
	Tested ranges or available ranges	0-10%, 0-30% SO <sub>2</sub>	0-10% NO 0-30% SO <sub>2</sub> plus many others	0-300 ppm S0 <sub>2</sub> 0-300 ppm S0 <sub>2</sub> (min)	0-1000, 0-10000 ppm So <sub>2</sub> , NO	200 ppm NO <sub>2</sub> , SO <sub>2</sub> (min) max depends on process	0-10000 ppm SO <sub>2</sub> (max) 0-1000 ppm NO <sub>X</sub> (max)
	Availability						
	Period of unattended operation						
	Reproducibility			Repeatability 0.5% FS	Repeatability 1% FS	Repeatability 2% FS	
	Influence of barometric pressure variation						
413	Influence of sample flowrate variation						
	Lower detection limit						
	Ambient temp range	-1-49°C	-1-49°C	0-40°C	0-40°C (stack temp up to 400°C)	-5-60°C . (stack temp up to 600°C)	-20 -70°C . (stack temp <200°C)
	Temp dependence of zero						
	Temp dependence of sensitivity						
	Zero drift	1% FS/day	1% FS/day	17 PS/wk			
	Sensitivity drift	1% FS/day	1% FS/day				
	Response time	0.5-26 s (90%)	0.5-26 s (90%)		200 ms - 3s (90%)	2 s (90%)	
	Interference						
	Accuracy				±3 <b>7</b>	±3%	±5%

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TABLE A3. - cont'd

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	THERMOELECTRON D12000	HARTMANN & BRAUN RADAS 1G	SICK CM 21
Source of data	Manufacturers' literature	German suitability test	German suitability test.
Туре	In-situ	Extractive	In-situ
Tested ranges or available ranges	NO SO <sub>2</sub>	SO <sub>2</sub> 0-0.02% min NO <sub>x</sub> 0-0.01% min	0-3000 mg/m <sup>3</sup>
Availability		90-100% (over 3 mths)	95% (over 3 mths)
Period of unattended operation		1 week	8-12 weeks
Reproducibility		43-78	77
Influence of barometric pressure variation		17 FS/17 change in pressure	
Influence of sample flowrate variation	N/A	<17 FS	N/A
Lower detection limit		$0.02 \text{ g } \text{NO}_2/\text{m}^3$	
Ambient temp range	-18 -50°C (stack temp 316°C max)	15-45°C	-20 -55°℃
Temp dependence of zero		37 FS/10°C	<17 FS/10°C
Temp dependence of sensitivity		27 FS/wk 27 FS/wk	Not detectable <27 FS/period of unattended operation
Zero drift	2% FS/day	<17 FS/wk	12 s (90%)
Sensitivity drift	2% FS/day	<37 FS/wk	
Response time	>5 secs	34-60 secs	
Interference		CO <sub>2</sub> 10 vol Z ) CO 590 mg/m <sup>3</sup> )0.5Z SO <sub>2</sub> 2.7 g/m <sup>3</sup> )FS H <sub>2</sub> O saturated 300 K) NO <sub>2</sub> 150 mg/m <sup>3</sup> 2.5Z FS	$\begin{array}{cccc} \text{CO}_2 & 14.3 \text{ vol}\% & ) \\ \text{CO} & 1000 \text{ mg/Nm}^3 & ) \\ \text{NO} & 1340 \text{ mg/Nm}^3 & ) < 0.5\% \text{ FS} \\ \text{NO}_2 & 923 \text{ mg/Nm}^3 & ) \\ \text{H}_2^{\text{O}} & \text{saturated} & 300 \text{ K} ) \\ \text{dust} & 250 \text{ mg/Nm}^3 & ) \end{array}$
Influence of madjust- ment of beam			>2% FS in angle range up to 0.5°C

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## TABLE A4. - Performance Characteristics for NDUV Instruments for Measurement of SO2 and/or NOx

	ANACON MODEL 207	TEL CEC DDACECC ANALVETS AVAMETED
	Manifostimonal 14 tonstino	
Source of data	mainut accurers attrerature	Mailutaccurers allerature
Type	Extractive	Extractive
Tested ranges or available ranges	SO <sub>2</sub> NO <sub>2</sub>	$0-0.2$ , $0-100$ vol % $SO_2$ $0-250$ ppm, $0-1$ vol % $NO_2$ (visible not UV)
Availability		
Period of unattended operation		
Reproducibility	Repeatability 1% FS	
Influence of barometric pressure variation		
Influence of sample flowrate variation		
Lower detection limit		
Ambient temp range	-10 -45°C	10 -50°C
Temp dependence of zero		
Temp dependence of sensitivity		
Zero drift	<0.5%/day	2% FS/wk
Sensitivity drift		
Response time	0.55 (90%)	
Interference		
Influence of maladjustment of beam		

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TABLE A5. - Performance Characteristics of Dispersive UV Instruments for  $\mathrm{SO}_2$  and/or NO

	MELOY SA700	THERMOELECTRON MODEL 43
Source of data	Manufacturer's literature	Manufacturer's literature
Ranges	0-250 ppb min 0-10000 ppb max	0-0.5 ppm (min) 0-5 ppm (max)
Lower detectable limit	1.0 ppb	0.002 ppm
Zero drift	3 ppb/day	0.005 ppm/wk
Sensitivity drift	3% of reading at 80% URL	2% FS/wk
Response time	1.5-3 mins (95%)	2 mins (95%)
Ambient temp range	0-45°C	5-45°C
Interference	20 ppb total	Interference equivalents 0.000 ppm 0 <sub>3</sub> , H <sub>2</sub> S, NO <sub>2</sub> , CO <sub>2</sub> , CO, C <sub>2</sub> H <sub>4</sub> , H <sub>2</sub> O 0.005 ppm NO

TABLE A6. - Performance Data for Fluorescence SO2 Analysers

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	BECKMAN 955 (951 has been suitability tested in Germany)	BENDIX 8101-C	THERMOELECTRON MODEL 10	SIGNAL MODEL 4000
Source of data	Manufacturer's literature	Manufacturer's literature	Manufacturer's literature	Manufacturer's literature
Ranges	10 P/10 <sup>6</sup> NO min 10000 P/10 <sup>6</sup> NO max	0-0.2 ppm NO min 0-2 ppm NO max	0-2.5 ppm NO min 0-10000 ppm NO max	0-4 ppm NO min 0-10000 ppm NO max
Minimum detection limit	0.1 P/10 <sup>6</sup>	0.01 ppm	0.05 ppm	
Repeatability			1% FS	1% FS
Precision	0.5% FS	0.01 ppm at 80% URL		
Temp dependence of zero				1 ppm/5-15°C
Zero drift	1% FS/day	0.05 ppm/day	l ppm/day	
Sensitivity drift	1% FS/day	0.02 ppm/day	1% FS/day	
Ambient temp range	4-38°C	5-40°C	0-40°C	
Response time	5 secs (95%)	1 min (95%)	1.7 secs (90%)	1.5 secs (95%)
Interference		Interference equivalent 0.04 ppm		

TABLE A7. - Performance Data for Chemiluminescence NO<sub>X</sub> Analysers

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## ANNEXE 2. - MARKET AVAILABILITY OF DIFFERENT TYPES OF CONTINUOUS ANALYSER FOR SO<sub>2</sub> AND NO<sub>x</sub>

# TABLE A8. - Market Availability of Different Types of Continuous Sulphur Dioxide Analyser

Type of Instrument	Number of manufacturers producing this type of instrument for sulphur dioxide measurement
IR/UV Photometers	
Non-dispersive infra-red (extractive) Non-dispersive infra-red (in-situ) Non-dispersive ultra-violet (extractive) Non-dispersive ultra-violet (in-situ)	9 3 1 2
Total number of non-dispersive IR/UV photometers	15
Dispersive ultra-violet (extractive) Dispersive ultra-violet (in-situ) Second derivative ultra-violet (in situ) Interferential (extractive)	2 1 1 1
Total number of IR/UV photometers	20
Conductometric instruments Flame photometers Fluorescence analysers Electrochemical analysers	1 1 3 2

# TABLE A9. - Market Availability of Different Types of ContinuousNitrogen Oxide Analyser

Type of Instrument	Number of manufacturers producing this type of instrument for oxides of nitrogen measurement
IR/UV Photometers	
Non-dispersive infra-red (extractive)	5
Non-dispersive infra-red (in-situ)	3
Non-dispersive ultra-violet (extractive)	1
Non-dispersive ultra-violet (in-situ)	1
Dispersive ultra-violet (extractive)	2
Second derivative ultra-violet (in situ)	1
Interferential	1

Chemiluminescence analysers

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European Communities — Commission

#### EUR 10645 — Methods of sampling and analysis for sulphur dioxide, oxides of nitrogen and particulate matter in the exhaust gases of large combustion plant

F. Brieda, K. Bull, K.W. Bühne, M. Callais, T. Menard, St. C. Wallin, M.J. Woodfield

Luxembourg: Office for Official Publications of the European Communities

1986 — VIII, 156 pp. —  $21.0 \times 29.7$  cm

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In connection with the continuous emission monitoring requirements of the proposed CEC Directive, COM(80)704 Final, 15 December 1983, IRCHA, TÜV-Rheinland and WSL have studied methods of sampling and analysis for SO<sub>2</sub>, NO<sub>x</sub> and particulate matter in the exhaust gas of large combustion installations. Relevant existing and planned legislation in Member States has been outlined. Continuous measurement methods for SO<sub>2</sub>, NO<sub>x</sub> and particulates and discontinuous methods, some of which will be used as primary reference standards, have been described and discussed.

In addition, continuous and discontinuous methods for the measurement of oxygen, carbon dioxide, volume flow, temperature, humidity and pressure have been considered. The data treatment requirements of the proposed Directive and the type of equipment necessary for their implementation are discussed. The methods used in each Member State and the relevant national guidelines are tabulated. The number of installations and/or boilers, in each Member State, which will be affected by the monitoring requirements of the proposed Directive and the number of installations which is already in conformity have been estimated. These estimations are subject to certain constraints because of the quality of the data available.

The investment, installation, maintenance and calibration costs of continuous measurement and data treatment devices have been estimated.

A need for the harmonization, within the Community, of national legislation and national guidelines for measurement methods and for the establishment of national instrument approval laboratories, using standardized suitability testing procedures, has been identified.

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