

cost of control of sulphur dioxide, nitrogen oxides and particulates emissions from large combustion plants in oil refineries

Prepared by CONCAWE's Ad hoc Group on the Cost of Control
of SO₂, NO_x and Particulates in Large Combustion Plants

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ABSTRACT

This report presents the results of a study on the cost of controlling emissions of SO₂, NO_x and particulates from oil refineries by flue gas treatment. Reductions of emissions have been chosen in accordance with those proposed by the European Commission's Directive "Limitation of the emission of pollutants into the air from large combustion plants". The report calculates the retrofitting cost for the "average" CONCAWE refinery and the total cost for all EEC refineries. A short review of available emission reduction techniques is also included.

Dit rapport geeft de resultaten weer van een studie betreffende de kosten van het verminderen van de uitstoot van SO₂, NO_x en vaste deeltjes door olieraffinaderijen met behulp van rookgasbehandeling. De gekozen uitstootverminderingen zijn overeenkomstig de voorstellen vervat in de Richtlijnen van de Europese Commissie betreffende de vermindering van de uitstoot voor luchtverontreinigende stoffen door grote vuurhaarden. Het rapport berekent de korten van voorzieningen van bestaande installaties van de "gemiddelde" CONCAWE raffinaderij en de totale kosten van alle raffinaderijen in de Europese Gemeenschap. Een beknopt overzicht van beschikbare technieken ter beperking van de uitstoot is eveneens inbegrepen.

Der Bericht enthält das Ergebnis einer Studie über die Kosten der Einhaltung der Grenzwerte für SO₂, NO_x - und Feststoffemissionen aus Ölraffinerien durch Rauchgasbehandlung. Die Herabsetzung^x der Emissionen entspricht den Vorschlägen in der Vorschrift der Europäischen Kommission "Begrenzung der Schadstoffemission in die Luft aus Grossfeuerungsanlagen". In dem Bericht werden die Nachrüstkosten für die "durchschnittliche" CONCAWE-Raffinerie und die Gesamtkosten für alle EG-Raffinerien berechnet. Eine kurze Übersicht über verfügbare Techniken zur Emissionsverminderung ist eingeschlossen.

Ce rapport présente les résultats d'une étude sur le coût du contrôle des émissions de SO₂, NO_x et de particules dans les raffineries de pétrole par traitement des fumées de combustion. Les taux de réduction des émissions qui ont été considérés sont en accord avec ceux proposés par la Directive de la Commission Européenne sur "la limitation de la pollution de l'air par les grandes installations de combustion". Le rapport donne les coûts pour la raffinerie "CONCAWE" moyenne et pour l'ensemble des raffineries de la CEE. Ces coûts comprennent les coûts d'adaptation des installations existantes. Une présentation rapide des techniques actuellement disponibles pour la réduction des émissions est également jointe au rapport.

Este informe presenta los resultados de un estudio sobre el costo del control de emisiones de anhídrido sulfuroso (SO₂), óxidos de nitrógeno (NO_x) y partículas sólidas procedentes de la combustión en refineries mediante el tratamiento de los humos. Las reducciones de emisiones han sido escogidas de acuerdo con las propuestas en la Instrucción de la Comisión Europea titulada "Limitación de la emisión de contaminantes del aire procedentes de grandes plantas de combustión". En el informe se calcula el costo de amortización para la refinería "media" de CONCAWE y el costo global para todas las refineries de la CEE. Se incluye también, una breve revisión de las técnicas de reducción actualmente existentes.

Questo documento descrive i risultati di uno studio relativo alla valutazione del costo per ridurre mediante trattamento dei fumi le emissioni di SO₂, NO_x e polveri da impianti di grande combustione nelle raffineries di petrolio. I lavori di riduzione degli inquinanti sono fissati in accordo alla Direttiva proposta dalla Commissione Europea "Limitazione delle emissioni di inquinanti nei fumi da impianti di grande combustione". Il documento valuta il costo necessario per la raffineria "media" CONCAWE ed il costo totale per tutte le raffineries della CEE. Si fornisce anche una breve descrizione delle tecnologie di trattamento disponibili.

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SUMMARY

In response to a proposal from the Commission of the European Communities for a Council Directive (COM(83) 704 final) on the limitation of atmospheric pollutant emissions from large combustion plant of over 50 MW rated heat output, CONCAWE set up a study group to examine potential control methods and estimate anticipated capital and operating costs to achieve the proposed reductions for existing plant in oil refineries.

The study is based on applying control measures to the "average" CONCAWE refinery. This was derived from combustion and refinery fuel information, supplied by about 85% of the participating oil refineries. According to the Directive, member countries would have full flexibility in how the overall reductions are achieved. However, for the purpose of this study it is assumed that the proposed percentage reductions will apply to all oil refineries, although this may not be the case in individual member countries.

Preliminary screening studies were carried out to indicate the most cost-effective means of controlling sulphur dioxide (SO_2), nitrogen oxides (NO_x) and particulates emissions from large combustion plant. Three options were considered for SO_2 control: flue gas desulphurization (FGD), fuel oil desulphurization and fuel oil gasification. FGD, despite very high associated capital and operating costs, is the least costly. A selection of five commercial FGD processes (one throwaway, two gypsum and two regenerable) were chosen to determine their economics in a refinery situation. De NO_x processes and electrostatic precipitators (ESPs) were studied as being probably the most suitable means to achieve the proposed reductions in NO_x and particulates emissions respectively from refineries.

The study assumed only single train units and a minimum additional retrofitting cost of 25% on capital for all control facilities. The specific conditions in oil refineries require special consideration. This is because of the need to use the by-products of refinery processes as a fuel which may make it difficult to segregate fuel oil for burning. It was assumed that flue gas from fuel oil-firing can be largely segregated from that from fuel gas firing for FGD and ESP treatment. If this is not possible for safety, reliability or flexibility reasons the cost given in the report will be substantially increased.

To reduce SO₂ emissions by the proposed 60% using FGD, the minimum retrofiting costs are estimated to be (1 \$US = 3 Hfl.):

| | Costs (\$US million) | |
|-----------------------|----------------------------|--------------------|
| | "Average" CONCAWE refinery | All EEC refineries |
| Capital investment | 22 - 50 | 2,200 - 5,000 |
| Annual operating cost | 10 - 16 | 1,000 - 1,600 |

This equates to an operating cost of around 2,600 - 4,500 \$US/metric ton of sulphur removed.

It should be noted that waste disposal on a significant scale is required for many of the FGD processes and the disposal cost and space requirements can be considerable. Also, experience with FGD within oil refineries is limited.

For the proposed 40% reduction in NO_x emissions using DeNO_x processes, minimum retrofiting costs^x for the assumed number^x of DeNO_x units are estimated to be:

| | Costs (\$US million) | |
|-----------------------|----------------------------|--------------------|
| | "Average" CONCAWE refinery | All EEC refineries |
| Capital investment | 3.6 - 10 | 360 - 1,000 |
| Annual operating cost | 1.4 - 4 | 140 - 400 |

And similarly for the proposed 40% reduction in particulates emissions using ESPs the minimum retrofiting costs, including waste disposal, are estimated to be:

| | Costs (\$US million) | |
|-----------------------|----------------------------|--------------------|
| | "Average" CONCAWE refinery | All EEC refineries |
| Capital investment | 2.5 | 250 |
| Annual operating cost | 0.9 | 90 |

There is limited experience with ESPs on oil-fired plant.

The control of sulphur dioxide emissions incurs by far the greatest control costs of the three pollutants considered. Retrofitting of controls to existing refineries would be difficult because of space limitations close to combustion plant and multiplicity of stacks in some refineries. In those cases where space limitations are more severe than in the average CONCAWE refinery costs can be considerably higher.

It is considered that the above costs are accurate to an estimated -10/+40%.

1. INTRODUCTION

A proposal from the Commission of the European Communities for a Council Directive on the limitation of pollutant emissions into the air from large combustion plants (COM(83) 704 final, 19 December 1983) was submitted to the Council of the European Communities .

The Directive proposes a reduction in pollutant emissions from fired combustion plants (solid, liquid or gaseous fuels) whose rated thermal output, including that of ancillary plants, is equal to or greater than 50 MW (thermal). Emissions from a common stack (regardless of number of individual furnaces/boilers discharging to it) are understood to be defined as a single combustion plant. Specified emission limits are proposed for new plant, but for existing plants, overall reductions to be achieved by end of 1995 (basis 1980 emissions) are proposed as follows:

- 60% for sulphur dioxide (SO₂)
- 40% for oxides of nitrogen (NO_x)
- 40% for particulate matter (solids)

Member countries have full flexibility on how this is to be achieved. Programmes are to be drawn up not later than 31st December, 1986, for the progressive reduction of total annual emissions from large combustion plant.

In reaction to the above Directive, CONCAWE has obtained information regarding large combustion plants (by stacks) from about 85% of oil refineries who participate in CONCAWE.

A CONCAWE Ad hoc Group was set up to:

- i) examine potential control methods for pollutant emissions from existing refinery large combustion plant;
- ii) estimate anticipated capital and operating costs to limit emissions from existing refinery plant by the percentages proposed in the Directive.

The study group concentrated on the "average" CONCAWE refinery as a basis for costing and assumed that the proposed reductions would apply to oil refineries, although it is up to Member States to decide how to achieve these reductions.

2. BASIS OF STUDY

Table 1 shows the characteristics of the "average" CONCAWE refinery as regards the number of stacks in each of the three size categories, i.e. > 100, 50-100 and 10-50 MW (thermal). It also gives the actual amounts of oil and gas fired. The data relates to 1982 (when the CONCAWE questionnaire was sent out to all refineries the Draft Directive did not yet refer to 1980 as the base year).

The data show that the "average" refinery has 1.4 stacks of 50-100 MW (thermal) and 1.9 stacks of over 100 MW (thermal) capacity (i.e. those covered by the Directive). In 1982, these stacks handled flue gas from the combustion of 133 MW (thermal) of oil of average 2.7% wt sulphur and 178 MW (thermal) of refinery fuel gas.

Sulphur in refinery fuel gas can be reduced by methods already practised widely in refineries. However, there will be some refineries which require additional investment for fuel gas sulphur removal. For SO₂ reduction this study considered only the sulphur in fuel oil fired in combustion plants.

Nitrogen oxide emissions, although normally greatest for oil firing, are emitted from both oil- and gas-fired combustion plants. It is necessary to consider both when reducing NO_x emissions. This report uses as typical an NO_x discharge concentration of 500 mg/Nm³ in flue gas for the "average" CONCAWE refinery. This is based on correlations supported by some measured data.

Particulate emissions relate almost entirely to fuel oil combustion. Therefore, only oil-fired units were considered in this respect. As no measured CONCAWE data are available, this report uses a particulate discharge concentration of 200 mg/Nm³ in flue gas of oil-fired plant for the "average" CONCAWE refinery. This is based on a correlation with fuel oil quality.

Table 1 Characteristics of stack size distribution in the "average" CONCAWE refinery - 1982

| Category of stack size MW thermal | Number of stacks | Design firing MW thermal | Actual firing MW thermal | Oil-fired MW thermal | Gas-fired MW thermal | Oil sulphur % wt. |
|-----------------------------------|------------------|--------------------------|--------------------------|----------------------|----------------------|-------------------|
| > 100 | 1.93 | 445 | 254 | 111 | 143 | 2.6 |
| 50-100 | 1.38 | 98 | 57 | 22 | 35 | 2.8 |
| 10- 50 | 3.15 | 89 | 54 | 24 | 30 | 2.7 |
| Sub Total for stacks > 50 MW | 3.31 | 543 | 311 | 133 | 178 | 2.7 |
| TOTAL | 6.47 | 632 | 365 | 158 | 207 | 2.7 |

Average fuel oil sulphur content taken as 2.7% wt for stacks > 50 MW (thermal).

3. OPTIONS TO LIMIT POLLUTANT EMISSIONS

The main options for the control of pollutant emissions from refineries are set out in Figure 1. Sulphur dioxide emissions necessitate the greatest reduction under the proposed Directive and also involve the highest cost control measures. They were, therefore, the main area of investigation.

Fig. 1 Main options for control of pollutant emissions

FLUE GAS DESULPHURIZATION TECHNIQUES

- | | | | |
|-----|-------------|-----|----------------|
| (1) | Throwaway | | |
| | - | dry | spray dryer |
| (2) | Gypsum | | |
| | - | wet | two processes |
| (3) | Regenerable | | |
| | - | dry | copper oxide |
| | - | wet | sodium sulfite |

DeNO_x PROCESSES

- (1) Low NO_x burners
- (2) Thermal DeNO_x
- (3) Catalytic DeNO_x

PARTICULATES CONTROL

- (1) Cyclone separators
- (2) Baghouses
- (3) Wet gas scrubbers
- (4) Electrostatic precipitators

3.1 CONTROL OF SO₂ EMISSIONS

Costing studies have shown that fuel oil gasification is a totally uneconomic proposition for refinery fuel oil desulphurization purposes alone. Fuel oil desulphurization is also considered uneconomic for refinery fuel oil alone. Although involving less retrofitting problems than flue gas desulphurization (FGD), both fuel oil gasification and fuel oil desulphurization for refinery fuel are commercially less attractive than FGD if no incentives other than pollution control factors exist.

Of the three options studied, FGD, despite its high capital outlay, is the lowest cost means of controlling sulphur dioxide emissions from fuel oil-fired combustion plant. Many of the commercially available processes can be adapted to refinery size combustion plant. Several of the systems are largely proven technology in commercial use, mainly within coal-fired power stations. The restricted space in refineries, particularly around existing stacks, however, creates difficulties in retrofitting, and in some situations this may be physically impossible.

3.2 CONTROL OF NO_x EMISSIONS

One option to reduce nitrogen oxide is to retrofit "Low NO_x" burners to existing combustion plant. NO_x reductions typically range from 20-50% and in general will be less than the 40% proposed in the Directive. These burners, however, cannot be used in many existing furnaces/boilers, e.g. those with high intensity burners, difficult geometry, or using highly viscous fuel oil. For this reason thermal and catalytic "DeNO_x" processes were examined as proven means of achieving a 40% reduction. In practice, a portion of the proposed NO_x reduction might be achieved using low NO_x burners.

3.3 PARTICULATES EMISSIONS

Particulates control by fuel oil gasification is grossly uneconomic. Particulates control is achieved to some extent in most FGD processes. For those processes which do not provide a 40% reduction, electrostatic precipitators (ESPs) were considered. Experience on oil-fired heaters is, however, very limited.

Modifications to combustion equipment can assist in the overall particulates emissions reduction. A reduction of 10-50% may be possible, but is strongly dependant on burner/atomizer combination and quality of fuel fired.

4. COMMERCIAL PROCESSES INVESTIGATED

4.1 SO₂ CONTROL

Five of the commercially available FGD processes were chosen for investigation to determine their economics in a refinery situation. Those selected cover the range of commercially proven processes.

- | | | |
|---|-------------------------|---------------------------------|
| - | dry throwaway process | spray dryer |
| - | wet gypsum processes | gypsum vendor 1 |
| | | gypsum vendor 2 |
| - | dry regenerable process | copper oxide/sulphur product |
| - | wet regenerable process | sodium sulphite/sulphur product |

Note:

An undefinable percentage of the gypsum produced may find outlets, mostly at no realization. If there exists no commercial outlet the gypsum has to be dumped at a cost.

Appendix I gives a brief process description of each type.

Two gypsum processes were selected because this type of wet process has most full scale experience, although nearly all of it is in coal-fired power stations.

4.2 CONTROL OF NO_x EMISSIONS

Three options to control NO_x emissions were examined, i.e.

- low NO_x burners
- thermal DeNO_x
- catalytic DeNO_x

Appendix II gives some process details.

4.3 CONTROL OF PARTICULATES EMISSIONS

ESPs are frequently used in coal-fired power plants, but their use on oil-fired plant is very limited. Four of the five FGD systems studied incorporate some particulates reduction (ESP, wet scrubber or baghouse).

Brief process descriptions are given in Appendix III.

5. WASTE PRODUCTS FROM FLUE GAS TREATMENT

5.1 FLUE GAS DESULPHURIZATION

The waste sludge produced in flue gas desulphurization is a mixture of calcium compounds, together with a small amount of ash containing trace impurities.

For the treatment of waste from an FGD plant several options exist:

- (1) Ponding
- (2) Landfill of dewatered sludge
- (3) Stabilization or fixation to produce aggregate
- (4) Oxidation of calcium sulphite to gypsum

In the past the most widely used method has been the disposal of slurry waste to a pond. Although originally considered a cheap method in the USA, it is no longer considered desirable and it is not practical in Europe.

The second option is a dewatering system for the waste slurry. Use is made of a thickener followed by a vacuum filter or centrifuge. The dewatered solids are then transported to a landfill. However, often the dewatered solids are not suitable for landfill directly and require further processing, e.g. oxidation.

The third technique is stabilization or fixation, which has been developed using fly-ash or chemical additives to produce an aggregate. This, however, may present additional problems if it becomes classified as a chemical waste.

The fourth option is gypsum production. The specifications for gypsum require that the product be of a high purity. In order to achieve this, particulates removal equipment needs to be installed upstream of the FGD unit resulting in a requirement for the disposal of fly-ash.

Although the gypsum technique is currently the favoured FGD process in Europe, it is forecast that the production of gypsum will soon exceed the consumption. The operators of gypsum FGD systems may in future have to pay for the transport or eventually the disposal of gypsum waste.

The disposal of waste will cause environmental problems, e.g. leaching and space requirements.

5.2 PARTICULATES REMOVAL BY ESPs

Electrostatic precipitators for particulates removal also produce waste which needs disposal. The cost for this disposal is included in the cost estimate for particulates removal.

6. ASSUMPTIONS MADE IN THE STUDY

6.1 PROCESS RELIABILITY

Only single train units were considered in this study despite some reservations about their capacity to achieve 335 days operation per year (as proposed by the Directive). To assume parallel trains for reliability reasons would increase capital costs by approximately two thirds.

6.2 RETROFITTING

The retrofitting of FGD, catalytic DeNO_x and ESP facilities to existing refinery stacks is very site-specific and also varies from one process to another. A major problem will be the availability of adequate space in the proximity of existing combustion plants. As a minimum retrofitting cost, an additional 25% is included in the capital cost estimates, but this does not include ducting to distant sites. The alternative of locating FGD, catalytic DeNO_x or ESP plant at an available, but distant, site involves routing large ducting across existing facilities which is practically difficult and very expensive.

6.3 APPLICATION TO "AVERAGE" CONCAWE REFINERY

In 1982, the "average" CONCAWE refinery was operating at about 60% of design firing capacity. In practice individual plants at times were operated at varying loads up to full design firing. In order to avoid operating restrictions, any equipment modifications are normally designed for full capacity. This practice is applied to all pollution control processes considered in this report.

The calculated required FGD design capacity for the "average" CONCAWE refinery was 213 MW (thermal) at a 90% removal efficiency to give a 60% overall SO₂ reduction (Appendix IV). The average CONCAWE refinery (Table I) has 1.9 stacks greater than 100 MW (thermal). This study assumes that the FGD requirement will be split between two 110 MW (thermal) units (rounded up from two times 106.5).

The DeNO_x capacity is based on the full design firing of 543 MW (thermal) since both oil and gas firing produce NO_x.

A 40% reduction in NO_x by catalytic or thermal DeNO_x requires treatment of 362 MW (thermal) equivalent of flue gas (Appendix IV).

For cost estimating purposes the number of DeNO_x installations was assumed to be two of 100 MW (thermal) and three^x of 50 MW (thermal). If a NO_x reduction of over 40% is required, the capital cost may increase^x considerably.

The particulates removal design capacity is based on the total oil-fired capacity of 319 MW (thermal) (Appendix IV). 170 MW (thermal) equivalent treatment is required. As the nearest size cost estimates available applied to 100 MW (thermal), two units of this size were assumed for the cost estimate.

6.4

POTENTIAL SIGNIFICANT ADDITIONAL FGD COSTS

It should be noted that sulphur trioxide will pass as an aerosol through wet scrubber type FGD systems to some extent (about 30% of feed concentration). Depending on the levels in untreated flue gas, it may be necessary to increase flue gas reheat temperatures above levels assumed in this report to avoid acid dew-point corrosion problems. A possible alternative is to install a dry precipitator with ammonia injection, if excessive reheat is to be avoided.

It was assumed that desulphurization and particulates removal capacity would be only required for the fuel oil fraction of refinery fuel fired. Therefore, the cost obtained is minimum cost. Main factors that will increase the cost given are:

- higher flue gas volumes than assumed as a consequence of mixed fuel firing;
- corrosion problems with existing stack if this cannot handle treated gases from FGD systems (particularly of the wet types). In this situation a new stack or flue duct will be required.

7. CAPITAL AND OPERATING COSTS

7.1 INVESTMENT COSTS

Capital cost data were obtained from contractors and vendors for the processes studied. Wherever possible direct investment costs were obtained and collected data adjusted to take account of any significant differences between different contractors' costs for similar pieces of equipment. The direct costs were then worked up to obtain total erected costs. This was based on a standard calculation agreed by the CONCAWE study group. An allowance was made for the investment necessary for the associated utilities requirements. No allowance was made for other off-site requirements such as roads and offices. Costs are based on a Netherlands location, with investment in January 1984.

7.2 ANNUAL OPERATING COSTS

A method to determine operating costs was agreed by the study group, and contractors data worked-up accordingly. A standard CONCAWE annual capital charge of 25% has been included in the annual operating costs. Operating period is assumed to be 335 day/annum for all processes, although this may not be achievable for FGD plant.

8. COSTS FOR CONTROL MEASURES

8.1 FLUE GAS DESULPHURIZATION

Figures 2 and 3 show the estimated minimum capital and anticipated annual operating costs for refinery sized 50-300 MW (thermal) FGD units. As might be expected, the regenerable FGD systems producing a finished sulphur product attract the highest capital costs. The throwaway systems attract the least capital cost, but produce large quantities of sludge for disposal. The operating cost calculations include the assumption that authorized dumps are available at a cost, including transport, of 60 \$US/metric ton of sludge.

To achieve a 60% SO₂ reduction in the "average" CONCAWE refinery and in all EEC refineries the retrofitting costs would be:

| | Costs (\$US million) | |
|-----------------------|----------------------------|--------------------|
| | "Average" CONCAWE refinery | All EEC refineries |
| Capital investment | 22 - 50*) | 2,200 - 5,000 |
| Annual operating cost | 10 - 16 | 1,000 - 1,600 |

*) 60% SO₂ reduction

The costs of sulphur removed are:

Capital cost 5,700 - 12,900 \$US/annual metric ton of sulphur removed

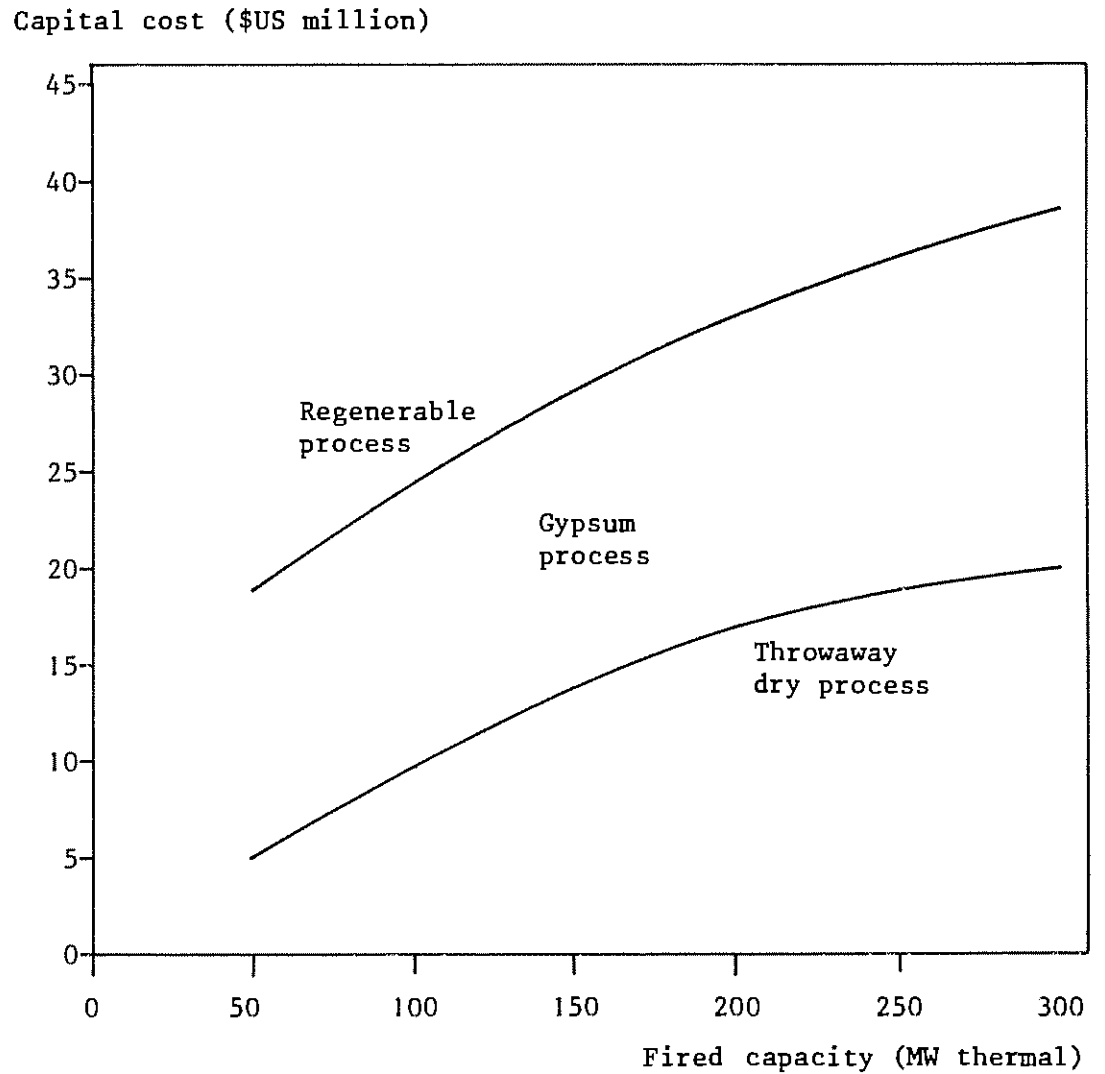
Operating cost 2,600 - 4,500 \$US/metric ton of sulphur removed

8.2 CONTROL OF NITROGEN OXIDES

To ensure a reduction of 40% in emissions, it is necessary to use either a thermal or catalytic DeNO_x process. Minimum retrofitting costs for these processes are estimated as follows:

| | Costs (\$US million) | |
|-----------------------|----------------------------|--------------------|
| | "Average" CONCAWE refinery | All EEC refineries |
| Capital investment | 3.6 - 10 | 360 - 1000 |
| Annual operating cost | 1.4 - 4 | 140 - 400 |

Fig. 2 Flue gas desulphurization: minimum capital cost for retrofitting to existing refinery stack (50-300 MW thermal)



(Basis: location in the Netherlands, 1984)

Generally the lower cost will apply to thermal DeNO_x and the higher to catalytic DeNO_x. Choice of process and cost will be site-specific.

8.3 CONTROL OF PARTICULATES EMISSIONS

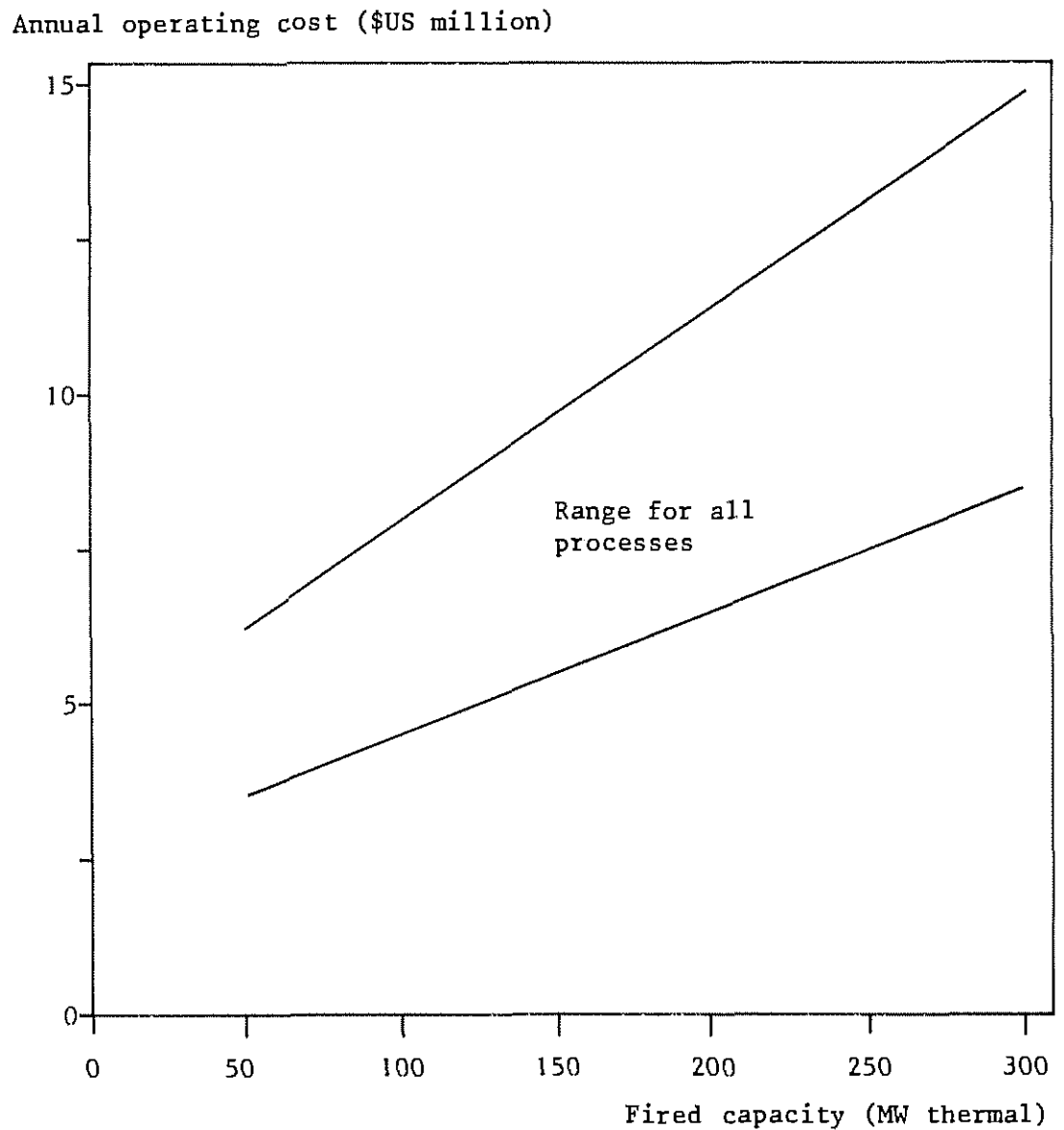
ESPs are probably the most practical means of controlling particulate emissions, although experience is very limited on oil-fired combustion plant. Assuming a 75% removal efficiency, the retrofitting costs to achieve a 40% reduction in emissions using ESPs are as follows:

| | Costs (\$US million) | |
|-----------------------|----------------------------|--------------------|
| | "Average" CONCAWE refinery | All EEC refineries |
| Capital investment | 2.5 | 250 |
| Annual operating cost | 0.9 | 90 |

8.4 SENSITIVITY OF COST DATA

The data received from process vendors and contractors' data have been of varying quality depending on the state of development of the process. Every effort has been made to standardize direct costs for similar pieces of equipment and it is considered that the above minimum costs are accurate to approximately -10%/+40%.

Fig. 3 Flue gas desulphurization: estimated annual operating costs for refinery-size unit



(Basis: location in the Netherlands, 1984)

9. CONCLUSIONS

For all EEC refineries the minimum capital and annual operating costs to achieve the proposed emission reductions (basis: location in the Netherlands, 1984) are:

| | Costs (\$US million) | |
|-----------------|----------------------|-----------------------|
| | Capital cost | Annual operating cost |
| SO ₂ | 2,200 - 5,000 | 1,000 - 1,600 |
| NO _x | 360 - 1,000 | 140 - 400 |
| Particulates | 250 | 90 |

This equates to an operating cost of around 2,600 - 4,500 \$US/metric ton of sulphur removed.

Control of sulphur dioxide emissions from existing refineries would incur by far the greatest control costs of the three pollutants considered in the proposed Directive.

Flue gas desulphurization (FGD) despite its high capital and operating costs is a much lower cost technique for SO₂ reduction than either desulphurization or gasification of refinery fuel oil alone. However, there is limited commercial experience with FGD processes in oil refineries.

Low NO_x burners will not necessarily achieve the proposed 40% NO_x reduction, and retrofitting would be impossible in many, if not most existing refinery combustion plant. Therefore, thermal and catalytic DeNO_x processes were studied to develop costs. However, there will be situations where low NO_x burners may assist in the overall NO_x reduction.

Particulates removal from oil-fired combustion plant is probably best achieved with electrostatic precipitators (ESPs). However, there is limited experience with ESPs on oil-fired combustion plant.

Retrofitting of emission controls to existing refineries would be difficult because of plot limitations and multiplicity of stacks in some refineries.

The impact of retrofitting emission control processes, particularly FGD systems, may necessitate further significant investments in some refineries, e.g. new stacks and ducting.

Waste disposal on a significant scale is required for ESP and for many of the FGD processes considered.

Combustion equipment modifications can contribute to an overall particulates emissions reduction, but probably not sufficient to achieve 40%.

1. FLUE GAS DESULPHURIZATION PROCESSES STUDIED - BRIEF PROCESS DESCRIPTIONS

1.1 DRY THROWAWAY (SPRAY DRYER)

The key component of the spray dryer FGD process is a chamber in which hot flue gas is contacted with a spray of extremely fine droplets of an absorbent slurry. The SO_2 (and SO_3) is absorbed in the droplets forming a mixture of calcium compounds (sulphites, sulphate).

The main steps to this process are:

- Absorbent preparation.
Lime is slaked to give 20-25% $\text{Ca}(\text{OH})_2$. A portion of the dry calcium compounds removed is slurried and recycled. The combined slaked lime/recycle is pumped to the spray dryer.
- Absorption.
The slurry is atomized and mixed with hot flue gas. SO_2 rapidly dissolves in the liquid phase and reacts to form the above salts. The flue gases simultaneously evaporate the water, leaving a free-flowing dry powder. The percentage SO_2 removal can be greater than 95%, and 99% of the SO_3 is also removed.
- Solids removal.
Solids collect in the conical base of the spray dryer chamber and in a baghouse downstream.
- Reheat.
The stack gas is reheated prior to discharge via the stack.
- Preparation of solids for disposal.
For landfill purposes a mixture of solid and water gives material of limited stability. The stability can be increased, if necessary, by the addition of coal-derived fly-ash.

1.2 GYPSUM (WET)

This process is essentially a wet lime scrubbing process. The chief steps are:

- Cooling of flue gases, absorption, neutralization and oxidation.
The alkaline scrubbing solution (moving counter-current to ascending flue gases in a scrubber) absorbs SO_2 and some SO_3 . The rich solution is oxidized by injection of air to form calcium sulphate, which is then largely removed for gypsum production. The suspension removed is neutralized with calcium hydroxide or calcium carbonate to form more gypsum.

- Stack gas reheat.
This can be accomplished either by regenerative heat exchange with hot flue gases, by partial by-pass of flue gases or direct reheat.
- Particulates removal.
Particulates in flue gas are usually removed using impingement baffles (mist eliminator) on top of the scrubber.
- Gypsum preparation.
The discharge slurry is pumped to a thickener where precipitated calcium sulphate settles under gravity. The thicker underflow is then pumped to a dewatering system and gypsum to specification is produced.

1.3 DRY REGENERABLE (COPPER OXIDE/SULPHUR PRODUCT)

This is based on the reaction of elemental copper with sulphur dioxide and oxygen in a fixed bed reactor. Copper-containing extrudates are placed between layers of wire gauze spaced between acceptor passages for flue gas flow. Copper sulphate is formed as flue gases pass along the surface of the acceptor material.

The spent acceptor is regenerated with a steam diluted hydrogen-containing regeneration gas produced by steam reforming naphtha. Several reactors are used in a swing operation to maintain continuous flue gas treatment.

The regenerator off-gas contains SO_2 , water vapour and unreacted fuel gas. The water vapour is condensed in a quench tower and the condensed water is steam-stripped to remove dissolved gases such as SO_2 , CO and CO_2 . The SO_2 is then stored under pressure and released to a modified Claus plant where elemental sulphur is produced.

1.4 WET REGENERABLE (SODIUM SULPHITE/SULPHUR PRODUCT)

In this process particulates are removed from the flue gas in a pre-scrubber which also cools the gas. The gas is then routed to a scrubber where fuel gases flow upwards and counter to a solution of sodium sulphite, absorbing SO_2 to form sodium bisulphite. The saturated flue gas then passes through a mist eliminator (similar to 1.2 above) and the flue gases are reheated by methods similar to the wet throwaway method, prior to discharge to atmosphere.

The rich absorbent is pumped to a regenerator where an evaporator is used to strip SO_2 . The clean absorbent is then returned to the absorber section.

To remove inert sulphate and thiosulphate, the absorber is purged. Overhead vapour from the evaporator is then routed to a condenser and stripper to give SO_2 suitable as feed to a Claus plant where elemental sulphur is produced.

1. NO_x CONTROL PROCESSES - BRIEF PROCESS DESCRIPTION

1.1 LOW NO_x BURNERS

Low NO_x burners stage either air or fuel addition with the aim of reducing peak temperature and residence time and thereby decreasing NO_x formation.

The decreases obtained vary from 20 to 50% with an average of about 35%.

Low NO_x burners cannot be retrofitted to many of the furnaces and boilers in existing refineries, since some have very large, high-intensity burners (8-10 MW each) for which proven low NO_x replacements are not yet available. Others burn very viscous fuel oils which may not be suitable for low NO_x burners.

1.2 THERMAL DeNO_x

Thermal DeNO_x is a non-catalytic process for removing oxides of nitrogen from flue gas by gas phase reaction with ammonia at high temperature (900-1200°C). Ammonia is injected through multiple nozzles into the radiant or convection section of process furnaces and boilers. To achieve good mixing, the small amount of ammonia is injected along with a carrier gas, usually air or steam. NO_x reductions from 40% and up to greater than 70% have been demonstrated in full-size combustion plant.

1.3 CATALYTIC DeNO_x

The catalytic DeNO_x process converts nitrogen oxides by mixing ammonia vapour with the flue gas. The mixture is passed through a catalyst bed where NO_x is reduced to nitrogen and water vapour with ammonia at a temperature of 250-400°C.

The quantity of ammonia applied is approximately 1.1 mole of NH₃ per mole of NO_x.

The catalytic DeNO_x process can tolerate small quantities of SO₂ and can achieve an efficiency of over 90% NO_x removal. A catalyst life of more than four years has been proven in several commercial units.

1. CONTROL OF PARTICULATES EMISSIONS - BRIEF DESCRIPTION

1.1 CYCLONES

A cyclone is a particulates collector in which the major collecting force is centrifugal. The gas with particulates enters the cyclone tangentially and particles are separated from the gas.

For particulates removal from oil-fired combustion equipment flue gases cyclones are less suited in view of the sticking properties of these particles which create problems in removal from the cyclone. Also, cyclones are relatively ineffective for smaller particles (less than 10 microns) and at lower than design throughputs.

The use of cyclones has, therefore, not been considered as an option for particulates removal in this study.

1.2 BAGHOUSES

Baghouses remove particulates by filtering flue gas through multiple parallel fabric bags suspended in large structures. They are unsuitable for removal of particulates from the flue gas of oil-firing because the somewhat "sticky" carbonaceous particles bind the fabric. They can, however, be used with the spray dryer FGD process because calcium salts make up most of the particulates.

1.3 WET GAS SCRUBBERS

Wet gas scrubbers rely on inertial impacting of water droplets and particulates. The wet particulates are allowed to build up to an extent in the liquid which is recirculated through the scrubber. This can be a tower or a venturi. Wet gas scrubbers have been used for particulate removal, but their use is restricted to special cases, e.g. incinerators, catalytic crackers and some flue gas desulphurization processes. They have two disadvantages:

- (1) the outlet flue gas is necessarily cold and may require reheat before discharge.
- (2) The solids in the circulating scrubbing liquid are held at a concentration of 5% wt or less. This necessitates a thickening process on the purge stream to make a waste product suitable for disposal.

In the absence of other requirements, e.g. FGD, wet gas scrubbers are unlikely to be an appropriate choice for particulates removal from oil-fired plant.

1.4 ELECTROSTATIC PRECIPITATORS (ESPs)

In an ESP, the flue gas is passed through an intense electrical field set up between electrodes of opposite polarity. This imparts a negative charge to the particles which are then attracted to the positively charged collector electrodes. The collected particulates are removed by intermittent rapping of the electrodes which dislodges and breaks-up the particulate layer into large agglomerates, which are heavy enough to fall out of the gas stream without being re-entrained.

The design of an ESP is related to the gas volume, gas temperature, particulate concentration, particulate size and particulate composition. For effective precipitation, the gas must remain in the electrical field for a sufficient length of time for the particles to take up their charge and be precipitated onto the collector electrodes. Other parameters being constant, the smaller the particle, the longer the necessary contact time.

ESPs are normally used on coal-fired boilers, where particulate removal efficiencies of 99.5% are achieved in some modern installations. ESPs are also used on oil-fired boilers, but to a much more limited extent since fuel oil has only ppm ash compared to several % ash for coal. The composition of ash from oil-firing differs markedly from that of coal fly-ash. Whereas the latter is almost all inorganic, the former contains high percentages of unburned carbonaceous material. This significantly affects ESP performance and hence design. The oil ash has a low resistivity and thus a lower migration velocity than coal ash. The quantity of ash per unit volume is low, making high precipitation efficiency more difficult. However, for a constant outlet concentration, ESPs on oil-fired units do not need the high efficiency of ESPs on coal-fired units because inlet particulate concentration is low.

The small quantities of particulates removed in electrostatic precipitators contain heavy metals in varying concentrations. This may present difficulties of disposal. There is limited experience in Europe in coping with such waste.

The particulates from an oil-fired unit are sticky and this would tend to plug hoppers and conveyors and foul the high voltage insulators. The plugging problem is overcome by having electric heaters or steam coils under the hopper insulation and by using a wet dust collection system. In the latter, ash drops into a shallow water-filled tank in which a paddle keeps the ash in suspension. A water flushing system for the internal surfaces of the hopper may also be necessary. Fouling of the high voltage insulators is overcome by blowing clean heated air across each insulator.

1. CALCULATION OF REQUIRED DESIGN CLEAN-UP CAPACITY FOR THE AVERAGE CONCAWE REFINERY

For FGD

Table 1 shows that the "average" CONCAWE refinery rated firing capacity in 1982 was 543 MW (thermal) of which 133 MW was utilized for oil-firing and 178 MW for gas-firing.

It is assumed that the non-utilized capacity of 232 MW (thermal) would be provided by fuel oil to the extent of 80%, on the basis that most refinery gas producers (e.g. cracking units) were already close to full capacity and little additional fuel gas would be made available. This results in a "design" oil-firing rate for cost estimates of:

$$133 + 0.8 \times 232 = 319 \text{ MW (thermal)}$$

Since 60% overall SO₂ reduction is proposed, but the FGD processes considered remove 90%, only:

$$\frac{60}{90} \times 319 = 213 \text{ MW (thermal)}$$

of oil-firing capacity will require flue gas desulphurization.

For DeNO_xing

A 40% reduction of NO_x emissions with a thermal or catalytic DeNO_x process having an efficiency of 60% requires, for the average CONCAWE refinery, treatment of:

$$\frac{40}{60} \times 543 = 362 \text{ MW (thermal)}$$

equivalent of flue gas.

For particulates removal

Electrostatic precipitators of 75% efficiency are assumed available.

To achieve 40% reduction of 319 MW (thermal) oil-firing design the capacity required will be:

$$\frac{40}{75} \times 319 = 170 \text{ MW (thermal)}$$