measured data on NO_x emissions in european refineries

Prepared by the CONCAWE Air Quality Management Group's Special Task Force on NO_x emissions from european oil refineries (AQ/STF 24).

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ABSTRACT

Recent NO_x concentration measurements provided by CONCAWE member companies, confirm earlier estimates of oil refinery NO_x emissions. For 1985, emissions from refineries are calculated to contribute 1.4% and 1.2% of total man-made NO_x emissions in OECD Europe and EEC-10 respectively.

Recentelijke meetingen van concentraties NO_x door de in CONCAWE samenwerkende oliemaatschappijen bevestigen vroegere gedane schattingen van emissies van NO_x. Berekeningen geven aan dat in 1985 de bijdrage door olieraffinaderijen 1.4% bedraagt van de totale uitstoot in de OECD landen in Europa en 1.2% van het totaal in de tien landen van de E.E.G.

Von CONCAWE-Mitgliedsfirmen durchgeführte neue NO_x-Konzentrationsmessungen bestätigen frühere Abschätzungen über die NO_x-Emissionen aus Erdölraffinerien. Im Jahre 1985 betrug der berechnete Anteil der Raffinerien an den gesamten anthropogenen NO_x-Emissionen 1.4% in den OECD-Ländern bzw. 1.2% in den EG-10-Ländern.

Des mesures récentes de la concentration de NO_x fournies par des Sociétés membre de CONCAWE confirment les estimations faites dans le passé concernant les émissions de NO_x par les raffineries . Pour l'année 1985 l'on a calculé que les émissions des raffineries contribuent de 1,4% et de 1,2% du total des émissions de NO_x par l'homme dans les pays de l'OECD Europe et de l'EEC-10 respectivement.

A lo largo de 1985 las emisiones de NO_x imputables a refinerías serían del orden del 1,5 por ciento (uno y medio por ciento) del total de las emisiones de NO_x de origen humano en Europa Occidental (O.C.D.E. y C.E.E.).Esta estimación se basa en mediciones recientes, que han confirmado los resultados obtenidos en anteriores estudios de CONCAWE.

Dati relativi a recenti misure di concentrazione di NO_x sono stati messi a disposizione dalle società partecipanti nel CONCAWE. Essi confermano precedenti stime sulle emissioni di NO_x da raffinerie petrolifere. Sulla base di questi dati è stato calcolato che le emissioni dalle raffinerie nel 1985 hanno contribuito per l'1.4% e l'1.2% alle emissioni totali antropogeniche nell' Europa Occidentale (OCSE) e nella CEE-10 rispettivamente.

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SUMMARY

Recent NO_x concentration measurements, provided by CONCAWE member companies, confirm earlier estimates of total oil refinery NO_x emissions. Refinery heaters averaged 325 mg/Nm^3 and refinery boilers 500 mg/Nm^3, both expressed as NO_2. The average NO_x concentration in refinery flue gas is 380 mg/Nm^3. The total 1985 emissions from refineries are calculated to contribute 1.4% and 1.2% of total man-made NO_x emissions for OECD Europe and EEC-10 respectively.

1. INTRODUCTION

Measured data on NO_x emissions were provided by CONCAWE member companies in the form of individual data sets for separate heaters and boilers. These data were analysed to indicate both the range of NO_x in refinery flue gas and the average concentration. The average concentration was used to estimate the total mass NO_x emission from West European refineries, using 1985 throughputs and estimated fuel consumptions.

2. NO_x CONCENTRATION IN HEATER AND BOILER FLUE GAS

Figs. 1 and 2 show the distribution of NO_x concentration measurements in refinery heaters and boilers, respectively. The data (in total 187 points) have been compiled from information provided by CONCAWE member companies and cover units with oil, gas and mixed firing.

Fig. 1 (refinery heaters), based on 76 separate data points, shows that NO_x concentration in flue gas, expressed as NO_2, varies mainly from about 100 to 550 mg/Nm^3 (a span of 450 mg/Nm^3). Fig. 2 (refinery boilers), shows that emissions vary in the main from about 350 to 700 mg/Nm^3 (a span of 350 mg/Nm^3). For the data submitted, the mean NO_x emission from boilers, mainly oil fired, is 500 mg/Nm^3 and from heaters, with a greater proportion of gas firing, is 325 mg/Nm^3 - Figs. 1 and 2 clearly indicate the wide range of NO_x concentration in flue gas from both heaters and boilers. Standard deviations are 100-120 mg/Nm^3 from the mean values.

Assuming total refinery fuel fired typically splits 2:1 between heaters and boilers respectively, the average NO_x concentration in refinery flue gas is 380 mg/Nm^3.

3. CALCULATION OF NO_x MASS EMISSIONS

The mass NO_x emission from the petroleum industry may be calculated from a knowledge of the ratio of flue gas/heat fired, the total heat fired and the average NO_x concentration in flue gas. The dry flue gas rate (corrected to 3% O_2) for typical refinery fuel oil is 12 Nm^3/kg oil. Refinery fuel gas produces more flue gas per kg of fuel, typically 13.4 Nm^3/kg, but a lower mass of fuel gas is required since gas has a higher calorific value. Hence the oil/gas ratio is not a significant factor. The fuel fired in the typical refinery in 1985 is 5.3% of refinery throughput as shown by the survey just completed by CONCAWE (1).The OECD Europe refinery throughput is 560 Mt/yr (2) and

EEC-10 throughput is 400 Mt/yr (3). Thus annual NO_x emissions from refinery fuel firing are:

OECD Europe:

$$12 \times 10^3 \times 0.053 \times 560 \times 10^6 \times 380 \times 10^{-9} = 135\ 300 \text{ t}$$

EEC-10:

$$12 \times 10^3 \times 0.053 \times 400 \times 10^6 \times 380 \times 10^{-9} = 96\ 700 \text{ t}$$

To the above must be added the NO_x emissions from fluid catalytic crackers, which are estimated to add 11% to the fuel fired emissions. This results in total NO_x emissions from the petroleum industry of 150 200 t/yr in OECD Europe and 107 300 t/y in the EEC-10.

Comprehensive statistical data on total man-made emissions in OECD Europe and in the EEC-10 in 1985 are not available, but from the published information (4,5) they should be in the region of at least 11 Mt/yr and 9 Mt/yr respectively. Thus the petroleum industry contribution in the two cases is calculated to be 1.4% and 1.2% of the total man-made emissions.

4. CONCLUSIONS

a) The many factors influencing NO_x formation (e.g. heater/boiler design, combustion intensity, fuel quality, combustion air preheat) lead to a wide variation in NO_x concentration in flue gas.

b) The mean NO_x concentration from 76 refinery heater data points is 325 mg/Nm^3 and from 111 boiler data points is 500 mg/Nm^3.

c) Assuming total refinery fuel fired typically splits 2:1 between heaters and boilers respectively, the average NO_x concentration in refinery flue gas is 380 mg/Nm^3.

d) For 1985, the calculated petroleum industry NO_x emissions are 1.4% and 1.2% of total man-made NO_x emissions in OECD Europe and the EEC-10 respectively.

5. REFERENCES

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Fig. 1 NO_x emissions from refinery heaters

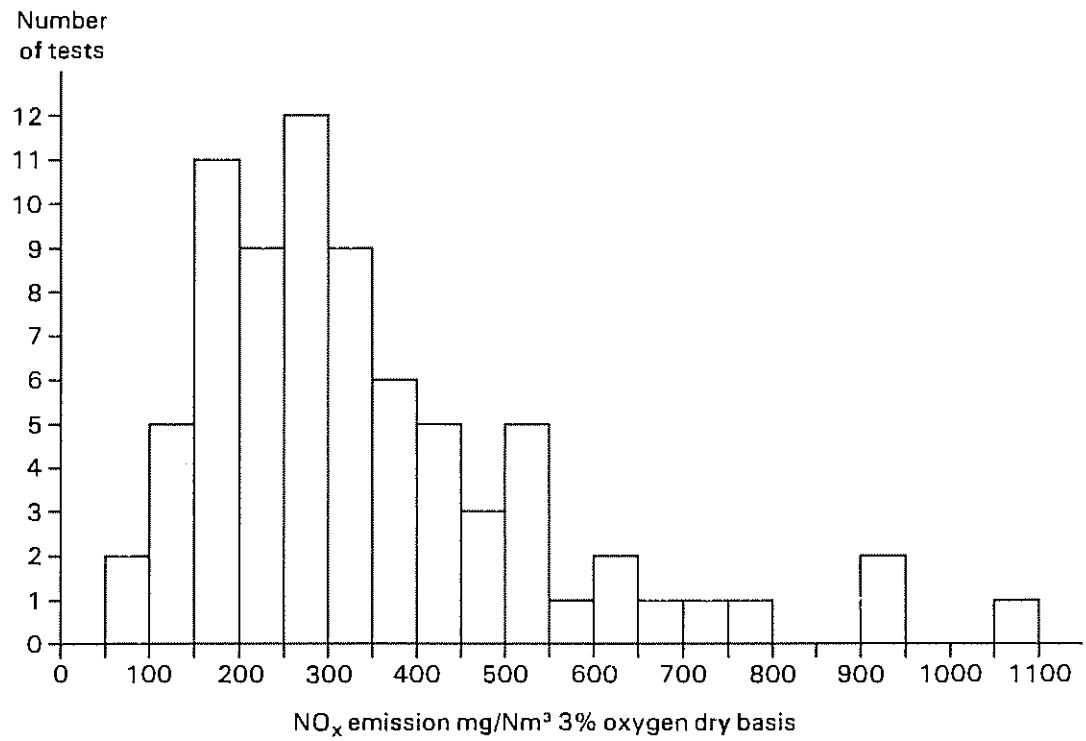

Fig. 2 NO_x emissions from refinery boilers