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N_2O EMISSIONS FROM COMBUSTION PROCESSES

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Abstract

N_2O emissions from the Dutch power plants, chemical industry, oil refinery and a waste incineration plant were determined. During sampling special precautions were taken in order to avoid the well known artifacts. N_2O emissions at the level of ambient air concentrations of 0.3 ppm(v) were determined at power plants fired with coal, oil and gas, refinery furnaces fired with a variety of different fuels and at chemical industry furnaces fired with a variety of high calorific fuels. Low N_2O concentrations were determined in flue gases from gasturbines without additional firing in a subsequent boiler and also at a waste incineration plant.

1. INTRODUCTION

In 1987 EPA estimated that the N_2O concentration in flue gases from coal combustion amounted 20-25% of the NO_x concentration. For gas a value of 3-7% was estimated [1]. In 1988 it became clear that these high values were due to sampling artifacts [2]. For reliable estimation of the N_2O emissions from power plants a method was developed in which these sampling artifacts were avoided. Then N_2O concentrations in the flue gases of different power plants were measured. Secondly the N_2O emissions of several other combustion processes were determined. This last part of the work was carried out and financed within the framework of the National Research Programma on Global Air Pollution and Climate Change.

2. DEVELOPMENT OF AN ARTIFACT FREE SAMPLING METHOD

A method was chosen in which grab samples were taken from flue gases which were analysed on a gaschromatograph after some time. During a series of extensive tests carried out with flue gases from a pilot plant gas burner in which additional gases could be added an optimal method was achieved. The SO_2 in the flue gases was removed with a series of washing bottles filled with a H_2O_2 solution. Then the flue gases were dried by means of a permeation dryer and stored in glass sampling bottles under overpressure (0.5 bar). By analyzing the flue gas directly and after several periods of storage no increase in N_2O concentrations (up to one week) could be detected. This was also the case during the emission measurements in which the samples were analyzed as soon as possible (mostly within 24 hours) and as a check also after 3-7 days. The tests showed also that storage in stainless steel cilinders in some cases give a rise in N_2O concentration.

3. N_2O EMISSIONS FROM ELECTRICITY GENERATION

Measurements were carried out at a variety of power plants fired with coal, gas and oil. Also the influence of load, firing method and flue gas cleaning equipment was investigated.

3.1. Coal fired power plants

Measurements were carried out at six power plants with a capacity in the range between 115 and 650 MW. N_2O concentrations were below 0.2 ppm for three boilers with a conventional firing method at full load (90 - 96%). For two-stage combustion at three other boilers (180 - 520 MW) N_2O concentrations ranged from below 0.2 ppm to 0.4 ± 0.2 ppm in the load range between 45 - 95%. No enhancement of N_2O concentrations was found at two boilers (650 and 520 MW) equipped with a flue gas desulphurization plant (concentrations <0.2 ppm). One boiler (115 MW) equipped with a $DeNO_x$ installation (selective catalytic reduction - SCR) showed no increase in N_2O concentration (<0.2 ppm). As the N_2O concentrations are as well as below or slightly above ambient air concentrations an emission factor of 0 ± 0.1 g N_2O-N/GJ was calculated.

3.2. Gas fired power plants

The N_2O concentration from a 640 MW conventional gas fired power plant ranged from below 0.1 ppm to 0.4 ± 0.2 ppm N_2O at a loads between 22% and 95%. The N_2O concentration from a conventional fired plant with natural and blast furnace gas (460 MW) was below 0.2 ppm. At one boiler (180 MW) with two-stage firing N_2O concentrations were 0.2 - 0.3 (± 0.2) ppm with a load between 33% and 95 %.

N_2O concentrations at five gasturbines (40 - 130 MW) were in the range between 0.4 and 2.8 ppm. However when gasturbines were used in combination with a boiler ("boiler with topping gasturbine") the N_2O concentrations were below 0.2 ppm. Apparently any N_2O present in the flue gases from the gasturbine is destroyed in the boiler. The emission factor for boilers is 0 ± 0.1 g N_2O-N/GJ. The emission factor for gasturbines was estimated to be in the range between 0.8 - 3.2 g N_2O-N/GJ. When the total amount of gas burnt in the Dutch power plants is taken in account than the emission factor for gas fired power plants (boilers plus gasturbines) is 0.1 - 0.4 g N_2O-N/GJ.

3.3. Oil fired power plants

Although oil is almost not fired in the Netherlands in power plants, measurements could be done at one boiler (180 MW) at three different loads (33% - 95%). N_2O concentrations ranged from below 0.2 ppm to 0.4 ± 0.2 ppm. The N_2O concentration from one oil fired gasturbine (23 MW) was 0.9 ± 0.2 ppm. From these data an emission factor of 0 ± 0.1 g N_2O-N/GJ for oil fired boilers was estimated.

4. N_2O EMISSIONS FROM OTHER COMBUSTION PROCESSES

4.1. Refinery

The N_2O emissions from a series of 13 refinery furnaces fired with vacuum residue, refinery gas (consisting of mainly H_2 and CH_4) and fuel oil were all 0.3 ± 0.1 ppm.

4.2. Chemical industry

At a chemical industry N_2O emissions of <0.2 ppm were measured at two boilers fired with either methane gas (98% CH_4) or high calorific gas (88% CH_4 plus higher hydrocarbons). Also N_2O concentrations of <0.2 ppm were measured at a gasturbine with steam injection and at a combination block.

4.3. Waste incineration

At a municipal waste incineration samples were taken every hour during two days. The 14 measurements showed varying N_2O concentrations between 0.5 and 5.5 ppm with an average of 2.8 ppm. From this an emissionfactor of 20 g N_2O per tonne of domestic waste was calculated.

4.4. Fluidized bed combustion

Data were obtained from the company which runs a FBC. From the sampling procedure that was used it was concluded that no artifacts were present. As expected a strong dependence upon the freeboard temperature was found. N_2O concentrations in the range between 8-85 ppm were measured at the freeboardtemperature range of 790-940 °C. From these data a N_2O emission factor of 4.5 - 49 g N_2O/GJ was calculated.

5. CONCLUSIONS

The measurements indicate that the N_2O concentrations from most combustion processes such as in power plants, refineries and chemical industry are very low and around ambient air concentration (0.3 ppm). N_2O concentrations are low for gasturbines and waste incineration. N_2O concentrations range from 0.4 to 2.8 ppm for gasturbines without additional firing and 0.5 to 5.5 ppm for waste incineration. If the gasturbines are used with additional firing such as in a combination block N_2O concentrations are very low and around or even below ambient air concentration (0.3 ppm). As expected N_2O concentrations for fuid bed combustion (FBC) are high and range from 8 - 85 ppm depending upon the freeboard temperature.

The N_2O emissions from the Dutch power plants are in well agreement with a Japanse study in which 43 power plants were investigated. Yokoyama [3] found for coal, oil and gas fired power plants values of 0.5, 0.3 and 0.1 ppm respectively. As in a number of cases the N_2O concentrations were just below ambient air concentrations, N_2O from the ambient air could be destroyed in the combustion process and thus resulting in a negative N_2O emission.

As a summary the emission factors are shown in figure 1.

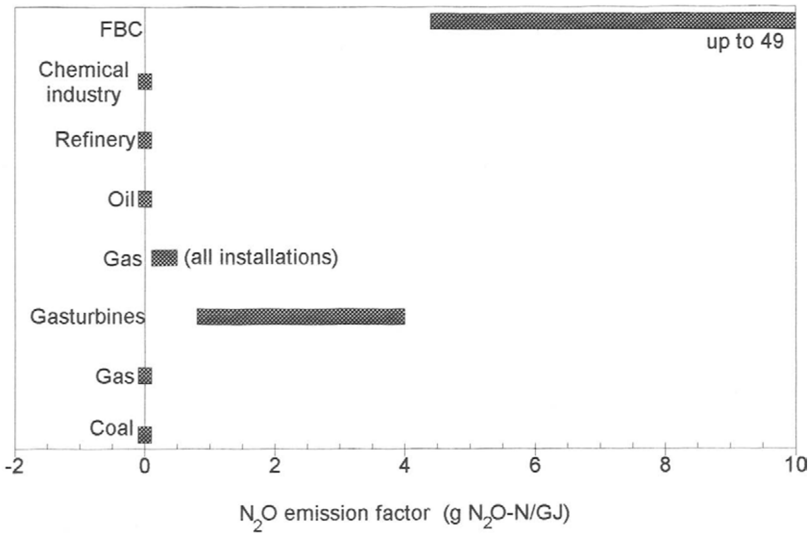

Figure 1. Range of N_2O emission factors for several combustion processes.

6. REFERENCES

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3 T. Yokoyama and S. Nishinomiya, Environ. Sci. Technol. 25 (1991) 347.