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REDUCTION OF N_2O EMISSIONS FROM FLUIDISED BED COMBUSTION BY REVERSED AIR STAGING.

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A new method, reversed air staging, for decreasing N_2O from fluidised bed combustion, was investigated in a 12 MW circulating fluidised bed boiler. Previous measurements, showing that the N_2O is decreased to one fourth (25 ppm), and NO to half (40 ppm) with maintained sulphur capture, were confirmed. Supplementary information on reversed air staging was obtained from additional measurements. By adjusting the temperature and the air ratio it was possible to reduce the CO emission. Thus, the N_2O emission can be reduced to the level of 25 ppm, with the emissions of NO, SO_2 and CO maintained at about same level as with normal air staging. Low load in combination with reversed air staging resulted in further reduced emissions. The effect of reversed air staging on the emissions from peat is similar to that of bituminous coal. There is no negative effect of reversed air staging on the combustion efficiency.

emissions, nitrous oxide, fluidized-bed combustion, air-staging, coal, peat

INTRODUCTION

N_2O is a greenhouse gas and is also believed to contribute to the ozone depletion in the stratosphere. The discovery of considerable emissions of N_2O from fluidised bed combustion, compared to other types of combustors, shifts the image of fluidised bed combustion technology from "clean" (low NO_x and SO_2) to "dirty" (N_2O unresolved). To benefit from the low emissions of NO_x and SO_2 from fluidised bed boilers (FBBs), a solution is needed for the N_2O emissions.

The processes involved in formation and destruction of NO and N_2O are complex and not completely understood. The same also applies to the sulphur capture process, which involves release of sulphur from the combustibles, reaction with CaO to $CaSO_4$ and reductive decomposition of $CaSO_4$.

It is well known that the emissions of NO_x, SO_2 and N_2O can be significantly affected by changes in operational parameters like bed temperature and air supply. The problem is that, while a measure taken to decrease one of the emissions may prove successful, it has the opposite effect on one or two of the others. The situation can be summarised as follows:

Raised bed temperature: N_2O decreases, but NO increases and the sulphur capture efficiency is considerably reduced.

Lowered air-ratio: N_2O and NO decreases, but the sulphur capture efficiency is considerably reduced.

Lowered fraction of primary air (increased degree of air staging): NO decreases, N_2O decreases somewhat, but the sulphur capture efficiency is considerably reduced .

This coupling of positive effects on N_2O to negative effects on other pollutants can be circumvented by addressing the conditions in the upper and the lower parts of the combustion chamber separately. While the conditions in the upper part are very important for the N_2O emission, the conditions in the lower part seem to be relatively more important for the sulphur capture. Thus a shift of conditions which provides more oxygen in the bottom part and less in the upper part may cause a reduced N_2O emission, while the sulphur capture remains unaffected. Such a reversal of the conditions compared to normal staging can be accomplished as follows:

The air-ratio in the combustion chamber and the cyclone is kept close to unity. No secondary air is used in the combustion chamber and all air is added in the bottom zone, except for some air which is supplied for final combustion after the cyclone, giving a total air-ratio of 1.2.

The increased air-ratio of the bottom part will make this part more oxidising compared to normal air staging. The gradual consumption of oxygen with height decreases the average oxygen concentration from the bottom and upwards approaching very low oxygen concentrations in the top zone of the combustion chamber and the cyclone, since the combustor air-ratio is kept at about unity.

With a bituminous coal of normal sulphur content the method, reversed air staging, has been shown to reduce the N_2O emission to one fourth (25 ppm), and NO to half (40 ppm) with a maintained sulphur capture efficiency of 90%.[1,2] In the present work the method is investigated further. The effects of load, temperature, combustor air-ratio and different rates of limestone addition are studied. Also, a run without limestone addition, i.e. with a sand bed, is included, as well some tests with peat and wood-chips as fuel.

EXPERIMENTAL CONDITIONS

The 12 MW circulating FBB used for the experiments has the features of a commercial boiler, but was built for the purpose of research. The boiler is equipped for various types of measurements and has facilities that make it possible to vary parameters independently and in a wider range than in a commercial boiler. The boiler can also be operated under extreme conditions inappropriate to commercial boilers.

The boiler is shown in Fig. 1. The height of the combustion chamber is 13.5 m and the square cross-section is about 2.5 m^2. Fuel is fed to the bottom zone of the combustion chamber through a fuel chute (3). Primary air is introduced through nozzles in the bottom plate (2) and secondary air can be injected through several nozzle registers located horizontally on both sides of the combustion chamber. In the case of reversed air staging air is introduced through the bottom plate (2) and in the cyclone outlet (r5). Entrained bed material is captured in the hot, refractory-lined cyclone (4) and returned to the combustion chamber through the return leg and particle seal (7). After the cyclone the flue gas passes an uncooled duct (5). Fig. 1 does not show the flue-gas recycling system, which supplies flue gas to the combustion chamber for fine tuning of the temperature in the boiler. Large, intentional changes in temperature can be made using the external, adjustable particle cooler (8).

The sorbent was Ignaberga limestone and the coal was a bituminous coal with a medium sulphur content. Limestone and fuel data are shown in Table 1. The peat used had a relatively high content of both sulphur and nitrogen.

Daily calibrated on-line gas analysers were used for continuous monitoring of O_2 (Magnos 7G and Magnos 5T, both paramagnetic, and Westinghouse 132/218, zirconia cell), CO (Uras 3G, i.r.), SO_2 (Uras 3G, i.r. and Binos, vis./u.v.), NO (two Beckman 955, chemiluminescence) and N_2O (Spectran 647, non-dispersive i.r.) in cold and dry gases. Except for an analyser called $O_{2,wet}$ (Westinghouse) which was sampling in the convection pass, all the analysers were connected to the gas duct downstream of the baghouse filter. The emissions of SO_2, NO, N_2O and CO have been normalised to a flue gas with an oxygen concentration of 6%. Before the gas was introduced to the N_2O analyser, the SO_2 in the flue gas was captured in a solution of carbonate, since the N_2O analyser is affected by high SO_2 concentrations (above 500 ppm).

The *total* air-ratio, λ_{tot}, is defined

$$\lambda_{tot} = 1 + K_c \frac{O_2}{21 - O_2} \tag{1}$$

where O_2 is per cent oxygen in the flue gas (including moisture) as measured in the convection pass, and the correction factor K_c is the ratio of theoretical flue gas (including moisture) to theoretical air (*i.e.* moles of flue gas over moles of air at stoichiometric conditions). K_c is 1.07 for the bituminous coal and about 1.2 for peat and wood-chips (depending on the moisture content).

By *combustor* air-ratio is meant the air-ratio corresponding to the conditions in the flue gas leaving the cyclone, *i.e.* before the addition of final combustion air in the reversed staging case. The combustor air-ratio is obtained as

$$\lambda_c = 1 + K_c \frac{O_{2,c}}{21 - O_{2,c}} \tag{2}$$

where $O_{2,c}$ is the oxygen concentration in the cyclone outlet, prior to air addition, derived from an oxygen mass balance

$$O_{2,c} = \frac{O_2(K_1 + y) - 21x}{K_1 + y - x} \tag{3}$$

where y is the ratio of flue gas recirculation to total air flow, x is the fraction of total air which is introduced in the cyclone outlet, and K_1 is

$$K_1 = 1 + \frac{K_c - 1}{\lambda_{tot}} \tag{4}$$

RESULTS FOR BITUMINOUS COAL

All test cases were run at constant load, *i.e.* the combustion air added was held at 3.54 kg/s, and the total air-ratio at 1.2 (3.5% $O_{2,wet}$). The bed temperature was normally 850°C, the total pressure drop 6 kPa. Symbols within brackets are used in Figs. 4-8 to denote the various cases.

Normal limestone addition (N): In this case the combustor air-ratio was varied between 1.0 and 1.2 by varying the fraction of air introduced in the cyclone outlet. The total air-ratio was kept at 1.2, and thus the highest combustor air-ratio, 1.2, indicates the extreme condition when all air is introduced in the bottom bed. The limestone addition was 180 kg/h, corresponding to a molar Ca/S ratio of about 2.7.

Extra limestone addition (E): In this case the optimal region was studied in more detail, i.e. the combustor air-ratio was varied between 1.0 and 1.07. The limestone addition was increased to 240 kg/h corresponding to a Ca/S of 3.6.

Previously published data, for comparison (C): In this case the limestone addition was 217 kg/h, corresponding to a Ca/S of 3.3.

Low load (L): In this case the load was reduced by 30%. The limestone addition was reduced to 100 kg/h, corresponding to a molar Ca/S ratio of 2.1.

Sand bed, i.e. no limestone addition (S): This case was run with a new sand bed, i.e. with a bed material containing silica sand and some fuel ash.

High temperature (H): These cases were run at about 870°C, but as opposed to the N, C, and L series they were not run in sequence and with varying limestone addition.

The effect of the combustor air-ratio on the four emissions is shown for normal and extra limestone addition in Figs. 2 and 3, and the emissions for the various cases are compared in Figs. 4-7. Fig. 8 shows the four emissions, as well as an emission index, versus the combustor air-ratio in all the cases. The results verify the previously obtained data and demonstrate that it is possible, with reversed air staging, to obtain low N_2O and NO emissions without increasing SO_2 emissions. The N_2O and CO emissions were somewhat higher compared to the previously published data (C), because of a difference of 5-10°C in the temperature of the *upper part* of the combustion chamber. The different temperature profile is probably caused a difference in flue gas recirculation rate.

Below, the results are commented with respect to influencing parameters and emissions:

Combustor air-ratio: The emissions are much affected even by small changes in the combustor air-ratio, λ_c. The CO and SO_2 emissions decrease with increased λ_c, while the NO and N_2O emissions increase. The NO and N_2O emissions have a minimum for a combustor air-ratio of about 1.03.

Temperature: The temperature is also a very important parameter for all the four emissions, which is demonstrated by the marked drops of N_2O and CO, and the rises of NO and SO_2 caused by an increase of 20°C.

Limestone addition: Increased limestone addition lowers SO_2, but probably also CO and N_2O, while the NO emission rises. In the absence of limestone, the N_2O and CO emissions are considerably increased, while the NO emission is decreased, demonstrating the important catalytic effect of the sorbent.

Load: An interesting result is the lower total emissions obtained with reduced load: all the four emissions decrease simultaneously despite the low molar Ca/S ratio used, cf. Fig. 8. This suggests that a lower fluidising velocity should be optimal for reversed staging.

The N_2O emissions are at a minimum for a combustor air-ratio of about 1.03, below which they again increase. The N_2O emissions are reduced by an increased temperature and probably also by an increased limestone addition. The effect of limestone addition is clearly seen if a comparison is made to a sand bed. Also the NO emissions show a minimum at about 1.03. NO is increased by limestone addition and by increased temperature. The SO_2 emissions decrease with increased combustor air-ratio, increase with temperature and decrease with increased limestone addition. The CO emissions increase dramatically at combustor air-ratios approaching unity and thus provide a lower limit for the combustor air-ratio. CO is considerably reduced at high temperatures. Also, increased limestone addition seems to decrease CO. The effect of limestone is clearly seen if a comparison is made to a sand bed with much higher CO emissions.

In order to see how the lowest emissions can be obtained, a total emission index is defined as the average emission, but with a weight of two chosen for N_2O:

$$\text{Emission index} = \frac{2p_{N_2O} + p_{NO} + p_{SO_2} + p_{CO}}{5} \qquad (5)$$

where p_i is the emission of compound i in ppm (normalised to 6% O_2). This index shows that there is an optimum combustor air-ratio, Fig. 8. With increased limestone addition, which lowers SO_2 and probably also N_2O and CO, the optimum is shifted towards lower values of combustor air-ratio. At the higher temperature the emission index is about unchanged, since the lower CO and N_2O emissions are compensated by higher SO_2 and NO emissions than at normal operating temperature.

The qualitative effect of changed weights in the emission index is evident from the effect of temperature and combustor air-ratio. Thus, a decreased emphasis on CO and SO_2 reduces the optimum combustor air-ratio, and a decreased emphasis on NO and SO_2 increases the optimum temperature.

The temperature and air-ratio can be used to control the emissions, for instance to decrease the CO emission to the level achieved with normal air staging, below 50 ppm, *cf.* case H 1.059 in Figs. 4-7. The example indicates that the N_2O emission can be reduced to the low level of 25 ppm, with all the other emissions - NO, SO_2 and CO - at the about same level as with normal air staging.

The emissions obtained with reversed staging should be compared with those obtained for normal staging[1], approximately 100 ppm N_2O, 80 ppm NO, 120 ppm SO_2 and 50 ppm CO, yielding an emission index of 90.

RESULTS FOR PEAT

All test cases were run at constant load, *i.e.* the combustion air added was held at 3.68 kg/s, and the total air-ratio at 1.2 (3.15% $O_{2,wet}$). A slightly lower bed temperature was used, 845°C, since a higher temperature was expected in the upper part of the furnace. The total pressure drop was 5.6 kPa.

Two test series, where the combustor air-ratio was varied, were performed without limestone addition, i.e. with a sand bed. These are shown in Figures 9 and 10. The two series were carried out on different days and differences in the levels of the emissions are probably explained by differences in the fuel composition. Although no limestone was present, a slight effect on SO_2 is seen which could be explained by a varying self-absorption of the fuel ash.

Two test series were performed with limestone addition at a molar ratio of about 2.5-3, the first at normal load, and the second at 70% load. The series at full load, Fig. 11, qualitatively resembles both the peat series without limestone and those of coal. One of the data points indicates a minimum of SO_2 and this is not expected. This result should perhaps not be given too much attention, since it could be caused by variations in fuel properties.

The emissions from the reversed staging case shown in Fig. 11, should be compared with those obtained for normal air staging, i.e. 63 ppm N_2O, 108 ppm NO, 101 ppm SO_2 and 26 ppm CO. As with coal it is possible to reduce N_2O and NO significantly with maintained sulphur capture efficiency.

The series at reduced load, Fig. 12, indicates a similar decrease of emissions with load as is seen with coal. Only the emission of SO_2 is lower for all combustor air-ratios at the lower load. For the other emissions the decrease seems to be dependent on the combustor air-ratio: N_2O and NO are considerably lowered below their previous optimum point, $\lambda_c \approx 1.03$, and no minimum is seen, while CO seems to decrease just around this point.

The results of the peat runs are qualitatively very similar to those of the coal runs with a distinct minimum for NO and N_2O at a combustor air-ratio of about 1.03 at normal load, and CO emissions increasing at low combustor air-ratios. The CO emissions are considerably lower than with coal which most likely is caused by a lower char concentration in the cyclone.

RESULTS FOR WOOD-CHIPS

The test was run at similar conditions as with peat, *i.e.* the combustion air added was held at 3.68 kg/s, the total air-ratio at 1.2 (3.15% $O_{2,wet}$), the bed temperature at 845°C, and the total pressure drop was 5.1 kPa.

The emission of N_2O is below the detection limit of the analyser and there is no need for sulphur capture because of the low SO_2 emissions. Thus, only NO and CO remain for a comparison with the other fuels. Both have a similar dependence on air-ratio as the other fuels, see Fig. 13, although the CO emission is much lower for the same reason as in the case of peat and the NO emission does not have a distinct minimum, such as was observed with the other fuels at full load.

Reversed air staging is not needed for N_2O reduction from wood combustion and the results are only included for comparison. The distinct effect of reversed air staging on NO, however, is interesting in view of the small effect of primary air stoichiometry and bed temperature on the NO emission from wood combustion.[3]

COAL COMBUSTION EFFICIENCY

A previous investigation of the combustion efficiency of coal under reversed air staging indicated that the loss of combustibles with fly-ash increased with about 25% compared to normal air staging.[1] These results, however, were obtained in conjunction with high CO emissions, 300-400 ppm. In Fig. 14 the estimated loss of combustible matter is shown versus the CO emission. For comparison the level for normal air staging is also shown. The data indicate that there is no significant difference in combustion efficiency between reversed air staging and normal air staging as long as the CO emission is not allowed to be too high.

DISCUSSION

The influence of temperature and combustor air-ratio on the emissions indicates different possibilities to obtain low total emissions, although with different emphasis. Thus, these parameters can be used to obtain a "tailor-made" combination of emission levels. For example, increased emphasis on the N_2O emission could be met by increased temperature; increased emphasis on CO could be met by increased temperature or combustor air-ratio, etc.

The parameters may be chosen to suit complementary measures for emission reduction. For instance, higher temperature and combustor air-ratio, which gives lower CO and N_2O but higher NO, may be combined with complementary measures downstream the furnace for further NO reduction. The combination of higher temperature and combustor air-ratio would also be positive for combustion efficiency.

The N_2O and CO emissions are sensitive to the temperature in the upper part of the combustion chamber. Although it cannot be safely shown by the present data, it is likely that a temperature profile, which increases with height of the combustion chamber, would further improve the reduction of total emissions. This could be obtained by moving cooling surfaces from the upper part of the combustion chamber.

The lower emissions obtained for decreased load indicate that operation at lower fluidisation velocities would be advantageous. (Note that it is only in conjunction with reversed staging that all emissions decrease at lower load.) If the reduction of the emissions is partly caused by a longer residence time, a higher combustion chamber would be beneficial for low emissions.

An additional advantage with reversed staging is that scaling-up is simplified, since the difficulties with secondary air penetration in the particle-laden gas of the combustion chamber are eliminated. On the other hand, it has to be recognised that introduction of more air in the bottom results in a higher auxiliary power consumption. The present results are related to the boiler used with its 13.5 m high combustion chamber. Translation of the results to combustors with other sizes has not yet been treated.

CONCLUSIONS

The following conclusions can be drawn:

- Previous measurements on the effect of reversed air staging have been confirmed.
- Part load in combination with reversed air staging results in further reduced emissions.
- The temperature and air-ratio can be used to control the emissions under reversed air staging, for instance to decrease the CO emission to the level achieved with normal air staging. Thus, the N_2O emission can be reduced to the low level of 25 ppm, with all the other emissions - NO, SO_2 and CO - maintained at about same level as with normal air staging.
- When reversed air staging is applied on a sand bed, the emissions of N_2O and CO are considerably higher and NO is lower than with limestone addition, indicating the important catalytic role of the sorbent surface.
- The effect of reversed air staging on the emissions from a fuel with a considerably higher volatiles content, i.e. peat, is similar to that of bituminous coal.
- No significant difference of the combustion efficiency between normal air staging and reversed air staging is seen provided that the CO emission is not allowed to be too high.

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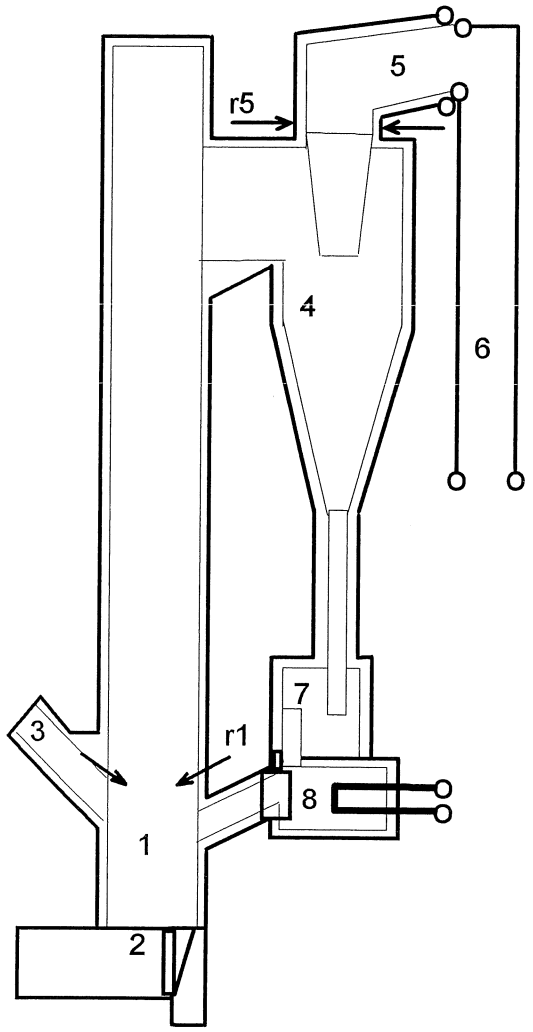

Fig. 1. The Chalmers 12 MW boiler.

1, combustion chamber;
2, air plenum and start-up combustion chamber;
3, fuel feed chute;
4, cyclone;
5, exit duct;
6, convection cooling section;
7, particle seal;
8, particle cooler;
→, secondary air nozzle inlets:
r1 at 2.2 m and r5 in cyclone outlet.

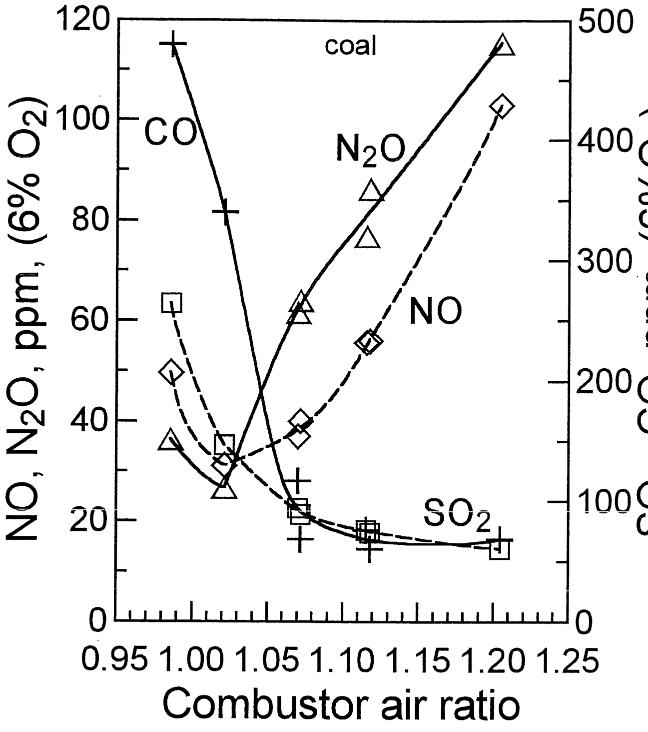

Fig. 2. Emissions versus combustor air ratio for normal limestone addition (N).

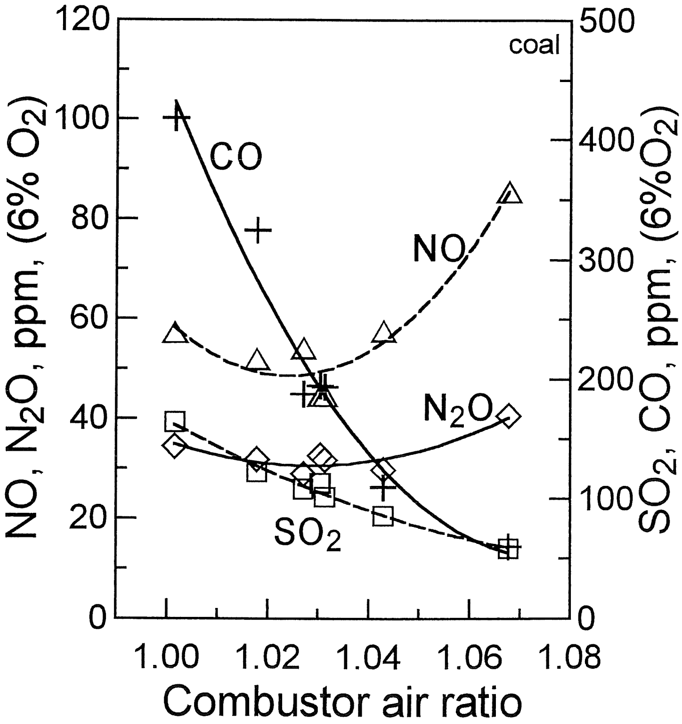

Fig. 3. Emissions versus combustor air ratio for increased limestone addition (E).

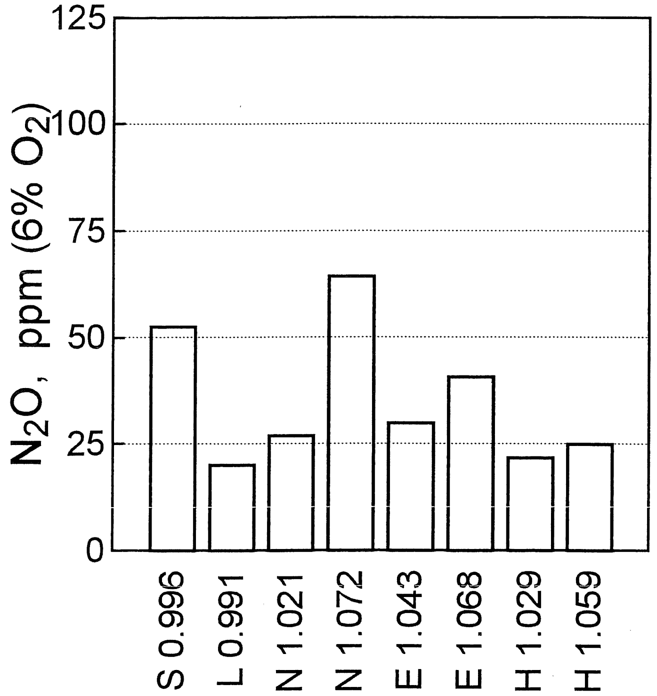

Fig. 4. Emission of N_2O versus case. Letter and number indicate case and combustor air ratio.

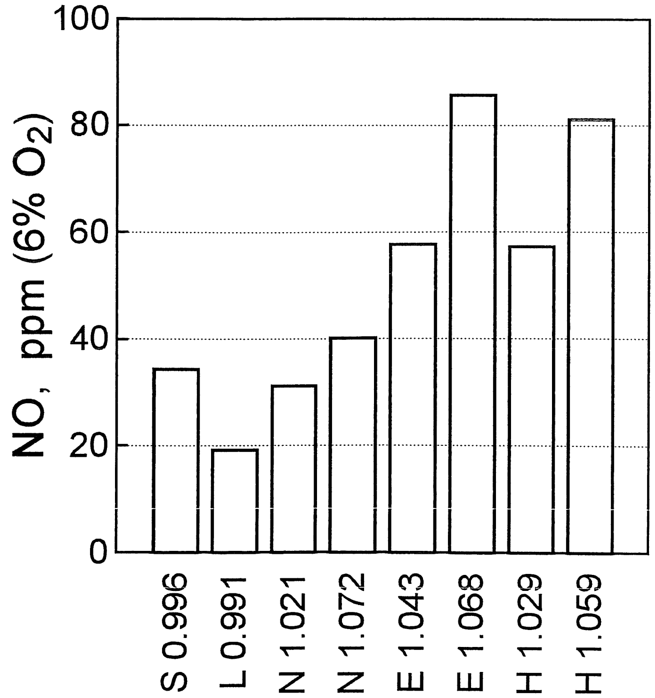

Fig. 5. Emission of NO versus case. Letter and number indicate case and combustor air ratio.

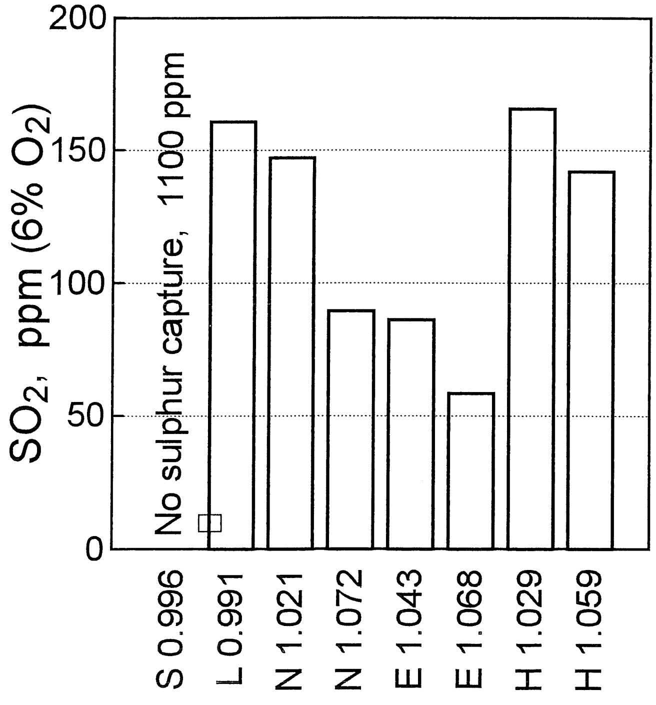

Fig. 6. Emission of SO_2 versus case. Letter and number indicate case and combustor air ratio.

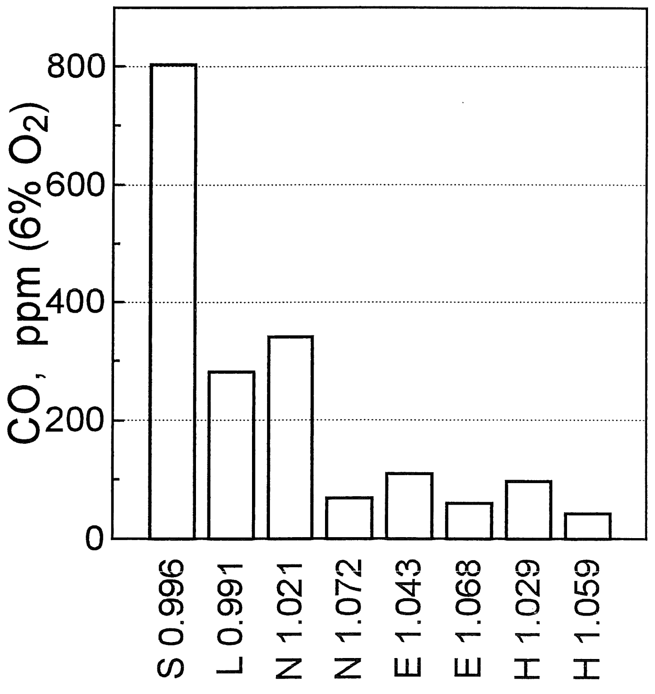

Fig. 7. Emission of CO versus case. Letter and number indicate case and combustor air ratio.

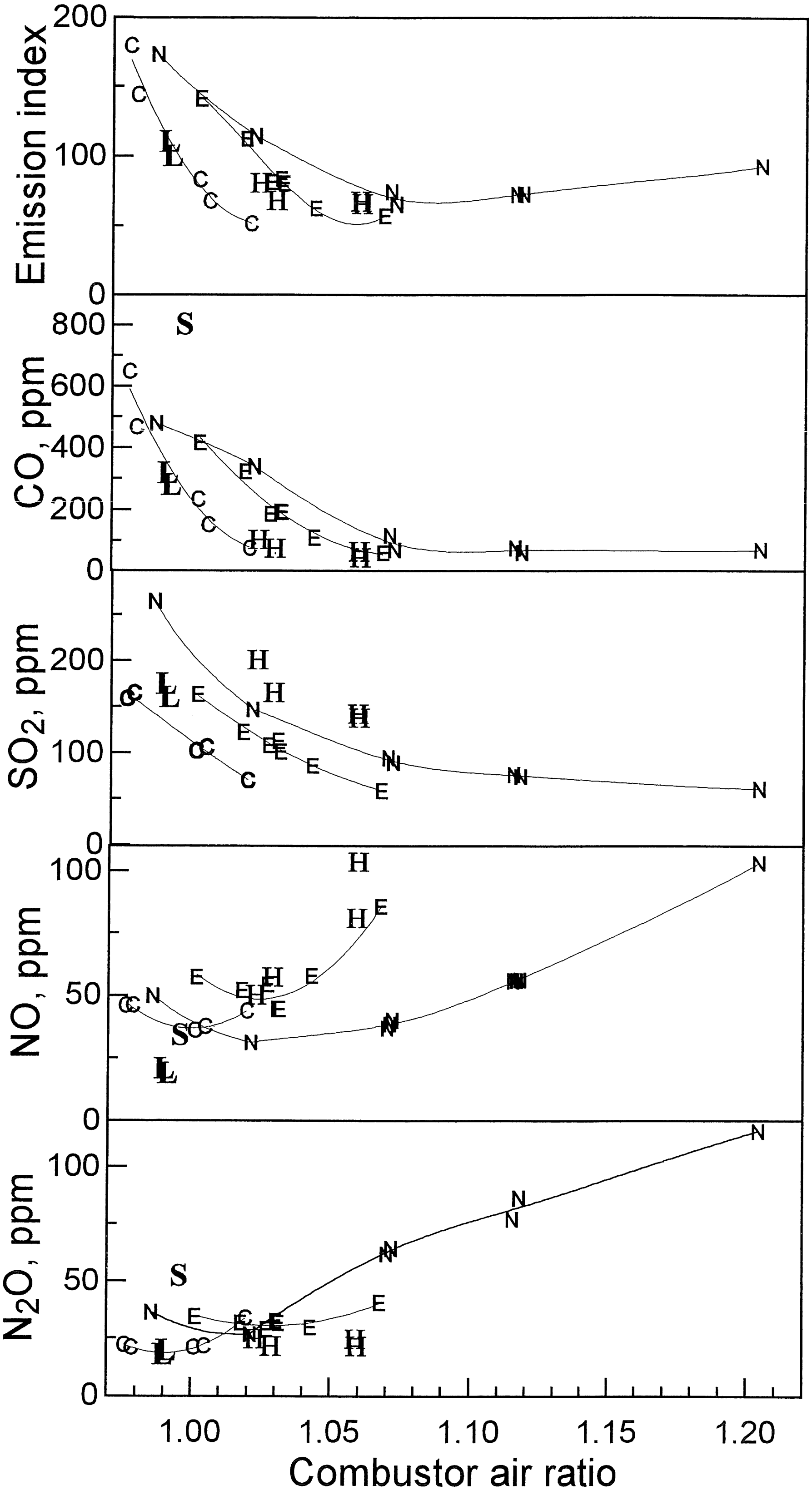

Fig. 8. Emissions and emission index versus combustor air ratio for coal runs (symbols see text).

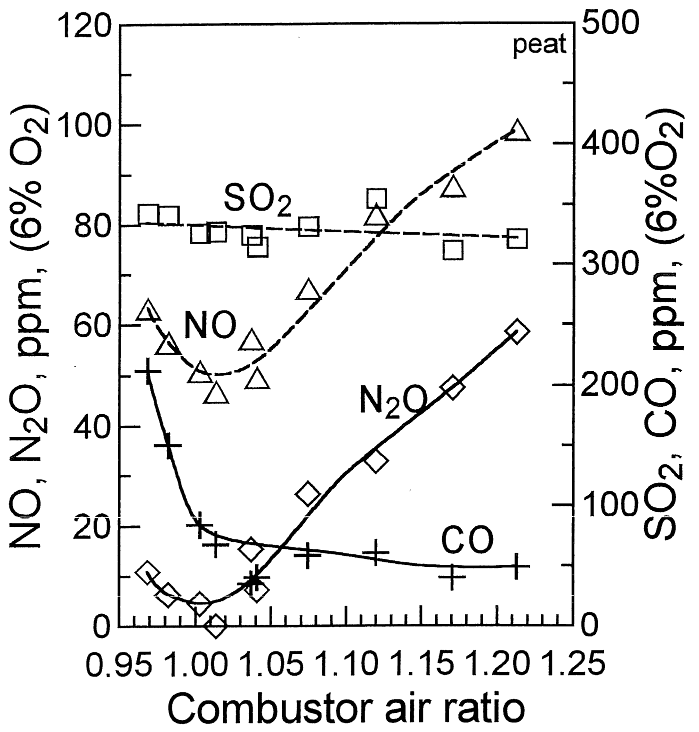

Fig. 9. Emissions versus combustor air ratio for peat. No limestone addition.

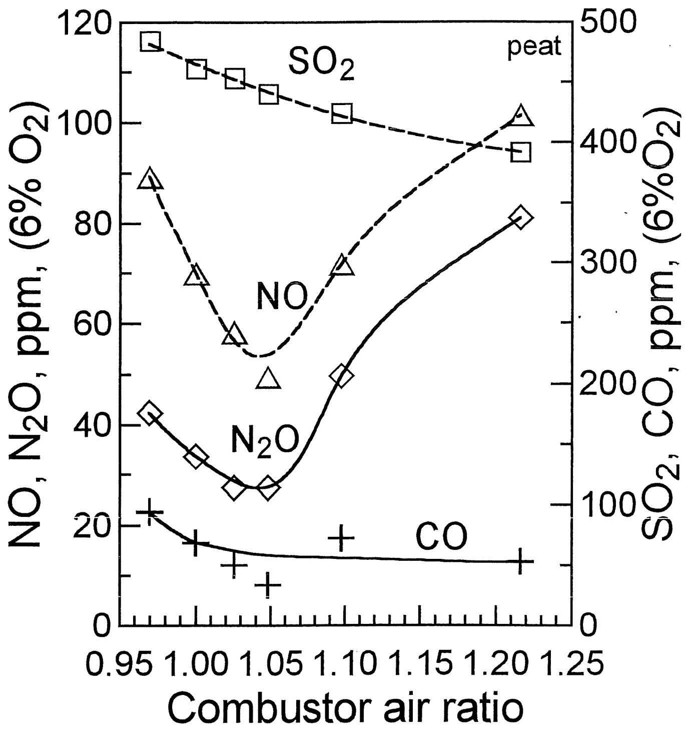

Fig. 10. Emissions versus combustor air ratio for peat (repeated). No limestone addition.

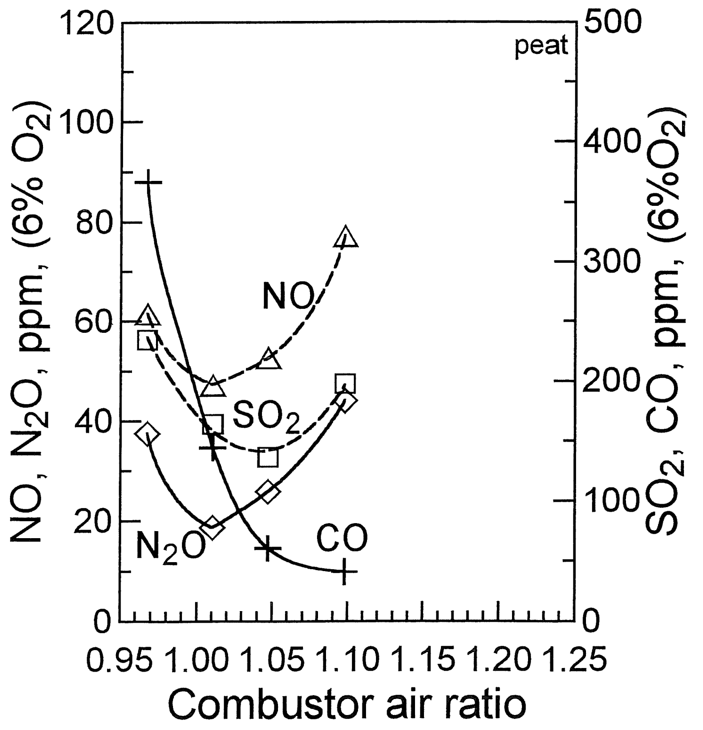

Fig. 11. Emissions versus combustor air ratio for peat with limestone addition.

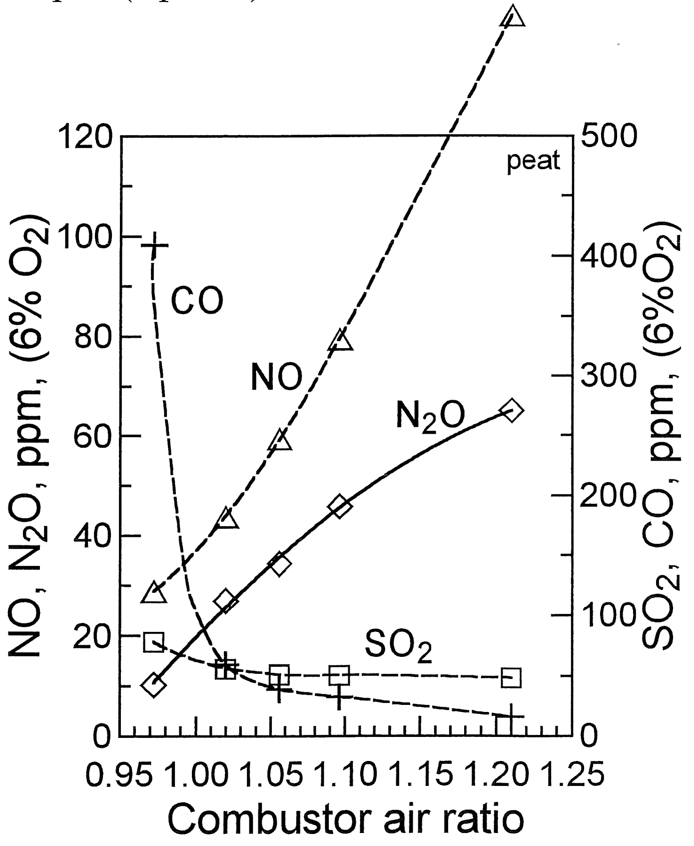

Fig. 12. Emissions versus combustor air ratio for peat with limestone addition at reduced load.

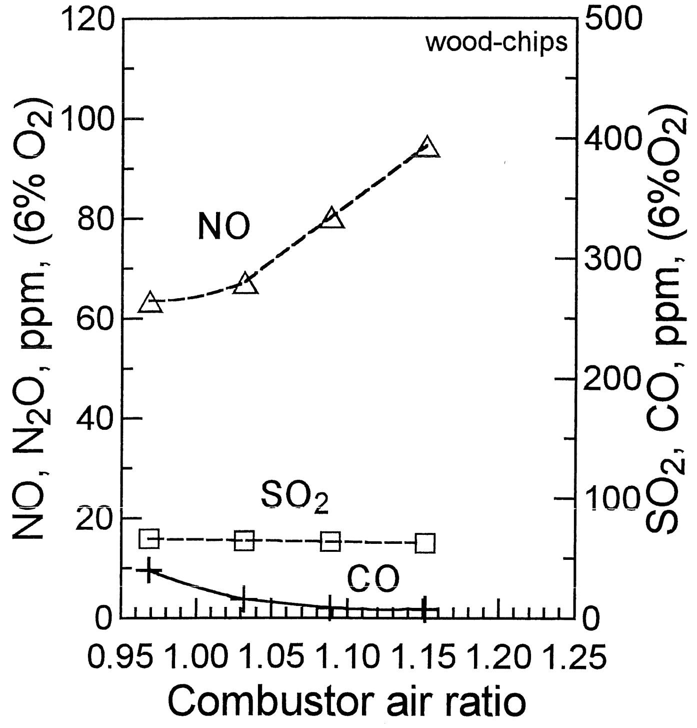

Fig. 13. Emissions versus combustor air ratio for wood-chips. (No limestone addition.)

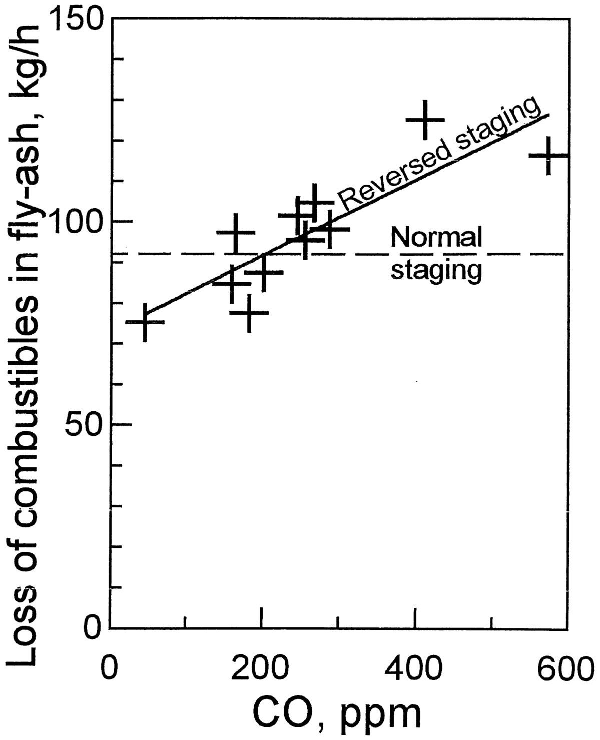

Fig. 14. Estimated loss of combustible matter in fly-ash versus CO. Level for normal air staging indicated for comparison.

TABLE 1. FUEL AND LIMESTONE MASS FRACTIONS IN % (daf=dry, ash-free).

Fuel	Bituminous coal	Peat	Wood-chips
size	<20 mm (50% <10 mm)	(not analysed)	(not analysed)
moisture	14	27	40
ash	9	4	0.7
volatiles, daf	40	69	82
carbon, daf	80	59	51
hydrogen, daf	5.7	6.3	5.9
nitrogen, daf	2.1	2.2	0.2
oxygen, daf	10 calculated	32 calculated	43 calculated
sulphur, daf	1.8	0.6	0.03
Sorbent	Ignaberga limestone	Ignaberga limestone	None
size	0.2-2 mm	0.2-2 mm	-
$CaCO_3$	90	90	-