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Reversed air staging — a method for reduction of N_2O emissions from fluidized bed combustion of coal

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Reversed air staging, a method for reduction of N_2O emissions, was studied in a 12 MW circulating fluidized bed boiler. The effect of combustor air-ratio, bed temperature, load and limestone addition ratio was investigated. The results indicate that if only the air distribution is changed, the emission of N_2O can be decreased to one fourth compared to normal air staging at a temperature of 850°C, with maintained low emissions of NO, SO_2 and CO. With increased bed temperature, 870°C, in combination with increased limestone addition, from Ca/S molar ratio = 3 to 4, it was possible to reduce N_2O by 90 vol% with low emissions of NO, SO_2 and CO. The method was less efficient at low load, because of the lower cyclone temperature. Also the effect of redistribution of some of the air added in the cyclone outlet to the cyclone inlet was studied. This did not, however, give any improvement in the boiler used compared to adding all of the air in the cyclone outlet. There was no significant difference in combustion efficiency between reversed air staging and normal air staging. © *1998 Elsevier Science Ltd. All rights reserved*

(Keywords: N_2O; fluidized-bed; combustion)

INTRODUCTION

N_2O is involved in both the greenhouse effect and in the destruction of the Earth's protective stratospheric ozone gas layer. Flue gases from fluidized bed combustion (FBC) of coal have significantly higher concentrations of N_2O than other combustion techniques and may, if not abated, constitute a problem if the use of FBC becomes widespread. The discovery of high N_2O-emissions from FBC has launched a considerable research effort and the knowledge in the area is accumulating rapidly. Important results are available in the literature[1–17].

Most of the work published is concerned with the understanding of formation/destruction mechanisms, although some work has been initiated on methods for N_2O reduction[6,18,19].

The two major parameters for controlling the environment in the combustion chamber of a fluidized bed boiler (FBB) are temperature and air supply. Air staging, i.e. primary air ratio, has only a small effect on N_2O emissions[16]. However, it is well known that N_2O emissions can be decreased by either increased temperature or by decreased air-ratio. The problem is that these two parameters also affect NO-emission, sulphur capture and combustion efficiency. The situation is summarized in *Figure 1*, which shows the effect of increased temperature and increased air-ratio (more oxidizing conditions) on the emissions of NO, N_2O, SO_2 and CO. (The latter also represents the general effect on combustion efficiency.) Normally, the N_2O-emission cannot be significantly decreased by re-optimizing the operation of a boiler because of the adverse effects on NO, SO_2 and combustion efficiency. Thus, the problem is not really how to reduce N_2O, but how to reduce N_2O without introducing adverse effects on other emissions and combustion efficiency. This means that a simple re-optimization of operation parameters, such as temperature and air-ratio, is not sufficient to solve the problem of N_2O reduction in FBC.

Two methods for N_2O reduction, afterburning and reversed air staging, have been studied in a 12 MW

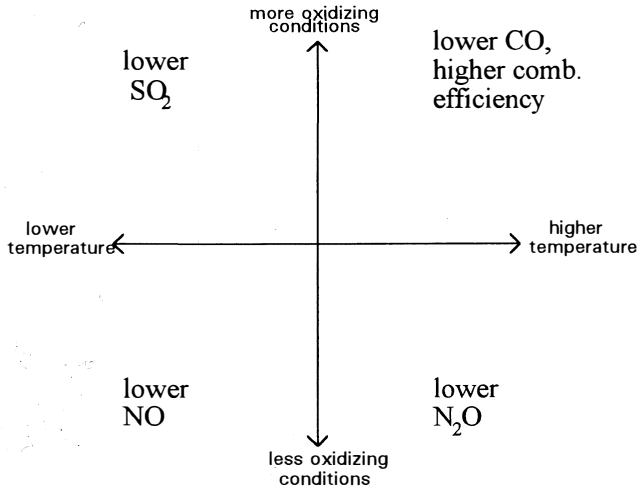

Figure 1 The optimization problem of fluidized bed combustion. The effect of changing temperature and air-ratio (oxidizing/reducing conditions) on various emissions and combustion efficiency is indicated

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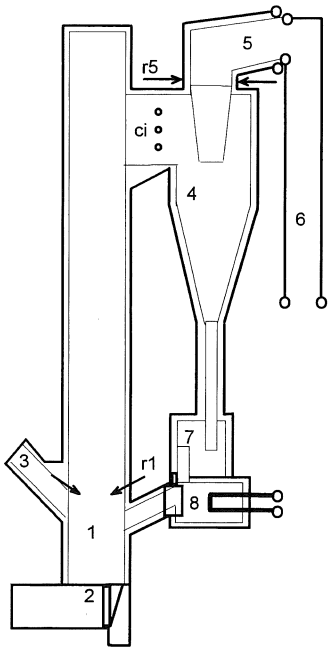


Figure 2 The Chalmers 12 MW boiler. (1) combustion chamber; (2) air plenum and start-up combustion chamber; (3) fuel feed chute; (4) cyclone; (5) exit duct; (6) convection cooling section; (7) particle seal; (8) particle cooler; →, °, secondary air nozzle inlets: r1 at 2.2 m, r5 in cyclone outlet, ci in cyclone inlet

Table 1 Fuel and sorbent data

Fuel	Bituminous coal
Size	< 16 mm (50% > 5 mm)
Moisture wt%	14.4 ± 0.5
Ash wt%	7.3 ± 0.3
Volatiles, wt% daf	36
Carbon, wt% daf	84.0 ± 0.5
Hydrogen, wt% daf	5.82 ± 0.06
Nitrogen, wt% daf	1.74 ± 0.02
Oxygen, wt% daf	≈ 7 (calculated)
Sulphur, wt% daf	1.58 ± 0.03
Sorbent	**Ignaberga limestone**
Size	0.2–2 mm
$CaCO_3$ wt%	90

circulating fluidized bed boiler[17,19,20]. With reversed air staging, the air-ratio in the bottom part is raised and the air-ratio in the upper part of the combustion chamber is lowered, as compared to normal air staging. More oxidizing conditions in the bottom part are beneficial for sulphur capture and do not affect the N_2O emission, while the lower air ratio in the upper part and the cyclone results in a significant lowering of the N_2O emission. It has recently been demonstrated[21], by oxygen probe measurements inside the combustion chamber, that the conditions in the riser are reversed compared to normal air staging. The present study is an extension of previous work on reversed air staging, which indicated that the effect of increased temperature and decreased load needs to be studied further[20]. In addition, the role of the conditions in the cyclone relative to the conditions in the riser was not clear. This aspect is also included in the present study with two test series showing the effect of redistributing some of the air added in the cyclone outlet to the cyclone inlet.

EXPERIMENTAL

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 *Figure 2*. 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 (5) to allow as long a gas residence time as possible before final air for completion of combustion is added. In this boiler there is a hot combustion space (5) before the gases are cooled in the convection path (6). In two of the test series some air was also introduced in the cyclone inlet (ci). 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). *Figure 2* 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).

Daily calibrated on-line gas analysers were used for continuous monitoring of O_2, CO, SO_2, NO, N_2O and total hydrocarbons. Except for one oxygen analyser which was sampling in the convection pass, all the analysers were supplied with cooled and dried gas sampled in 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 vol%. 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 ppmv). The mass fraction of combustibles in the ash samples was determined from mass loss during combustion at 815°C.

The total air-ratio of the boiler, λ_{tot}, defined as the air-ratio after addition of final air in the cyclone exit, was kept constant at 1.2 during all tests. By combustor air-ratio is meant the air-ratio corresponding to the conditions in the riser, i.e. before addition of air in the cyclone exit or inlet. The combustor air ratio, λ_c, is

$$\lambda_c = 1 + K_c\left(O_2 - \frac{21x}{1+y+(K_c-1)/\lambda_{tot}}\right)/(21-O_2) \quad (1)$$

where O_2 is percentage of oxygen in the flue gas (including moisture) as measured in the convection pass, $K_c \approx 1.07$ is the ratio of theoretical flue gas (including moisture) to theoretical air, y is the ratio of flue gas recirculation to total air flow, and x is the fraction of total air which is introduced in the cyclone outlet. If K_c is approximated by unity, the combustor air-ratio is given by the simpler relation

$$\lambda_c \approx \lambda_{tot}[1 - x/(1+y)]. \quad (2)$$

The air-ratio in the cyclone is equal to the combustor

Table 2 The reversed air staging series studied

Series	Symbol	Temperature	Limestone	Load	Air cycl entr	Ca/S (molar ratio)	Fig.
Ref.	○	850	normal	full	–	3.1	3(a)
High temp	T	870	normal	full	–	3.0	3(b)
Extra lime	E	870	high	full	–	4.4/3.8	3(c)
Low load	L	850	normal	70%	–	3.6	4(a)
Both T and L	B	870	normal	70%	–	3.6	4(b)
T, L and E	Z	870	high	70%	–	4.8	4(c)
	8	850	normal	full	8 vol%	3.2	5(a)
	3	850	normal	full	3 vol%	3.1	5(b)

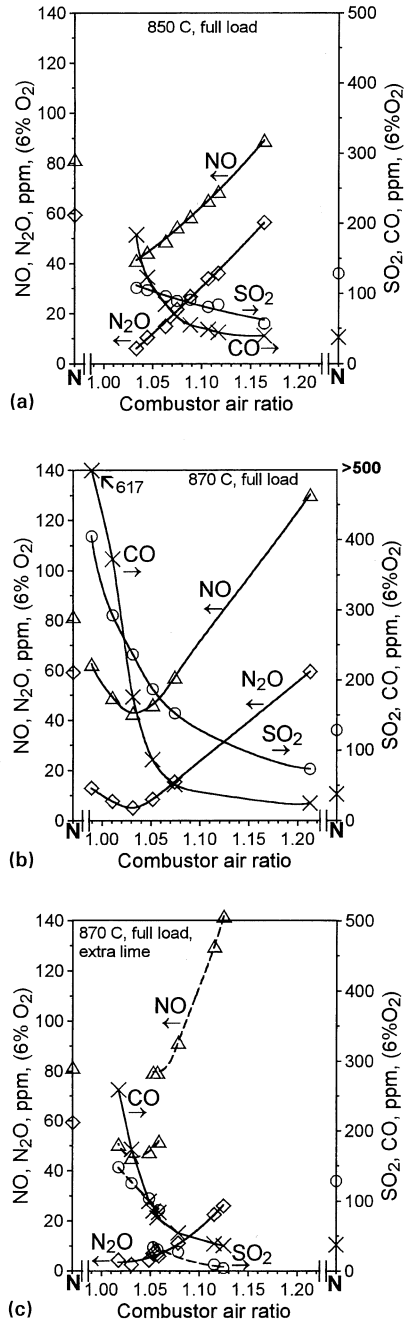


Figure 3 Full load: (a) reference series, i.e. 850°C and normal limestone addition (Emissions for normal air staging are shown on the vertical axes); (b) high temperature (Emissions for normal air staging are shown on the vertical axes); (c) high temperature and higher limestone addition. (Emissions for normal air staging are shown on the vertical axes)

air-ratio, except for the two cases where air is added both in the cyclone inlet and in the cyclone outlet. In these two cases a cyclone air-ratio is also needed, see below.

All test cases were run at constant load, i.e. the combustion air added was kept constant at 3.5 kg/s (full load), and the total air-ratio at 1.2 (3.5 vol% $O_{2,wet}$). The total pressure drop over the height of the riser was 6 kPa. The sorbent was Ignaberga limestone and the coal was a British bituminous coal with a medium sulphur content. Limestone and fuel data are shown in *Table 1*.

PROCEDURE

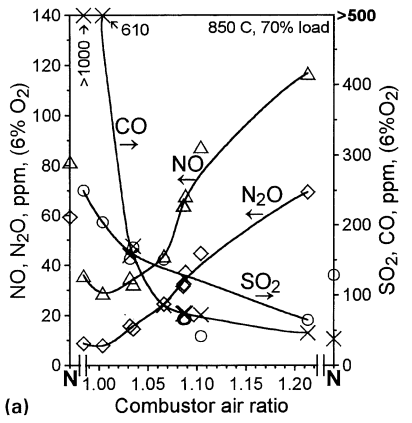
The reference test series was full load, a bed temperature of 850°C and a limestone addition rate of 200 kg/s corresponding to a Ca/S molar ratio of about 3. Eight test series were made, see *Table 2*. The first three series include (i) the reference series, (ii) increased temperature, and (iii) increased temperature combined with extra limestone addition. The next three test series are similar to the first three except that the load was reduced to 70% of full load. The two last series are based on the reference series, but with some air added in the cyclone entrance. The purpose of the latter two is to investigate how a rise in air-ratio from the riser to the cyclone affects the emissions under reversed air staging.

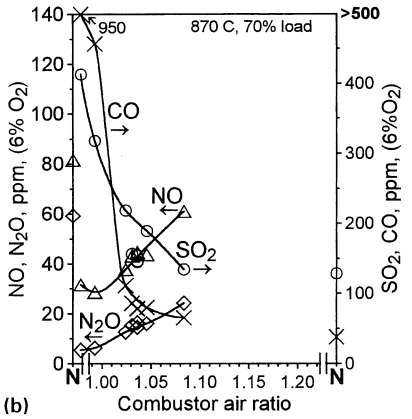
The procedure used in all the test series was to vary x, the fraction of the total air added in the cyclone exit, while keeping the total air-ratio, as well as other parameters of operation, constant. Thus, the total air flow to the boiler was constant, and an increase in the flow of air added to the cyclone exit (r5, *Figure 2*) resulted in an equal decrease in the air flow through the bottom plate (2, *Figure 2*). After each change of the air flow added to the cyclone exit the conditions were allowed to stabilize. The period between each change was typically one hour. The variation of the air flow added to the cyclone exit changes the combustor air-ratio, eqn (2), and the emissions are expressed as a function of the combustor air-ratio.

RESULTS

The emission data for the eight test series are shown versus combustor air-ratio in *Figures 3–5*. The results of reversed air staging should be compared to those of normal air staging at 850°C, shown on the two vertical axes in each diagram and denoted by **N** on the horizontal scale but, of course, not related to the horizontal scale. With normal air staging is meant that about 60 vol% of total air is added through the bottom plate (2, *Figure 2*), while 40% is added as secondary air at a height of 2.2 m (r1). The emissions under normal air staging were: 120–140 ppmv SO_2, 80 ppmv NO, 40 ppmv CO and 60 ppmv N_2O.

A comparison between the results of *Figures 3–5* may be





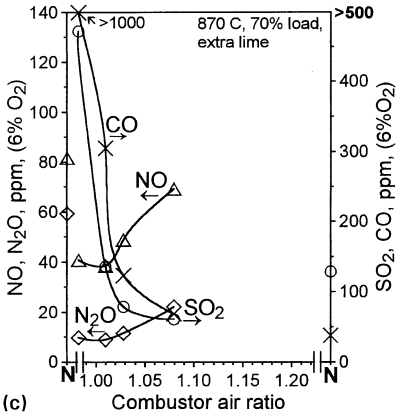
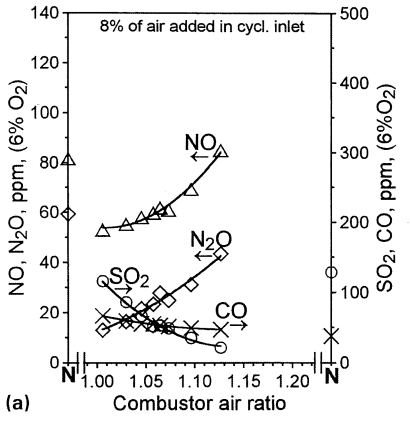


Figure 4 Low load: (a) 850°C; (Emissions for normal air staging are shown on the vertical axes); (b) high temperature (Emissions for normal air staging are shown on the vertical axes); (c) high temperature and higher limestone addition. (Emissions for normal air staging are shown on the vertical axes)



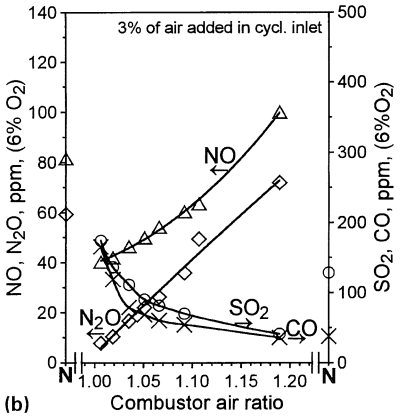


Figure 5 Part of the air is added in the cyclone entrance: (a) 8 vol% of total air in cyclone entrance (Emissions for normal air staging are shown on the vertical axes); (b) 3 vol% of total air in cyclone entrance. (Emissions for normal air staging are shown on the vertical axes)

difficult in some cases. Therefore, a more detailed analysis was made, where emission data for the various series were compared in the same diagram. Here, only an example is shown as in *Figure 6*, where CO is shown versus N_2O. Below the individual series are commented.

The reference test series shown in *Figure 3(a)* qualitatively resembles previous results. As the combustor air-ratio is lowered, the emissions of NO and N_2O decrease, while SO_2 and CO increase. At low combustor air-ratios CO increases dramatically. The figure indicates that N_2O can be reduced to one fourth with CO below 100 ppmv.

The high emissions of CO at low air-ratios can be suppressed by increasing the temperature as shown by the high temperature series in *Figure 3(b)*. The price paid for this decrease in CO emissions is higher SO_2 emissions. In addition, the N_2O emissions are further lowered while the emission of NO rises.

The higher SO_2 emissions at the higher temperature can be reduced by a higher limestone addition rate as shown in *Figure 3(c)*. The higher limestone addition also results in even lower N_2O emissions, below 5 ppmv. Thus, a

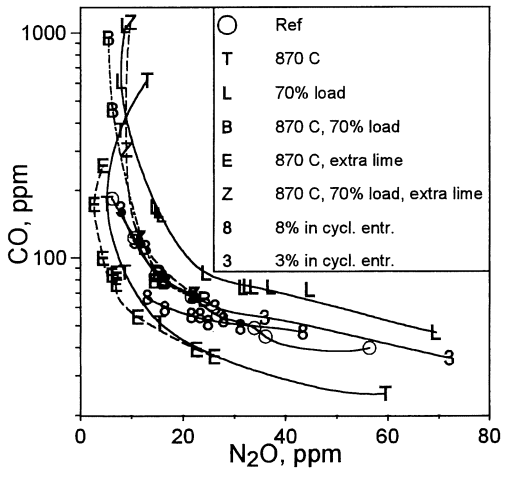


Figure 6 CO versus N_2O

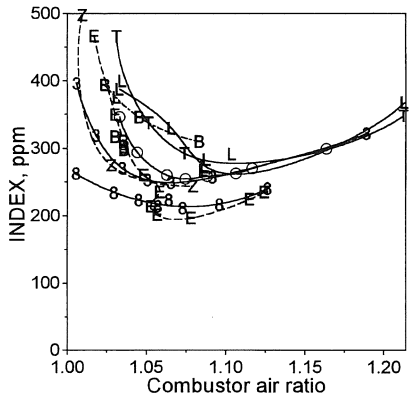


Figure 7 Emission index versus combustor air ratio. (Symbols, see *Figure 6.*)

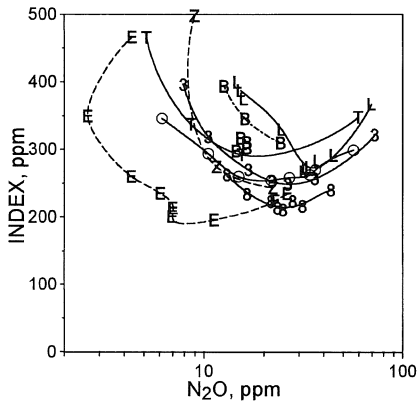


Figure 8 Emission index versus N_2O. (Symbols, see *Figure 6.*)

reduction of N_2O with 90 vol% compared to the normal air staging case is realized in combination with low emissions of NO, SO_2 and CO.

In *Figure 3(c)* there is a discontinuity in the graphs for NO and SO_2. This is caused by different Ca/S molar ratios. (In contrast to all other series, the series shown in *Figure 3(c)* includes data from two occasions with somewhat different limestone flows, cf. *Table 2.*) The highest Ca/S ratio gives lower SO_2 and higher NO.

In *Figure 4* the three series above are repeated at reduced load. The obvious effect of reduced load is higher N_2O emissions, while the effect on the other emissions is moderate. The increase in the N_2O emissions is a temperature effect. Although the bottom bed temperature is the same as in the normal load cases, there is a fall in temperature of about 15–20°C from the bottom bed to the riser top at reduced load, as opposed to normal load where there is instead a small temperature increase of 0–5°C.

In *Figure 5* the effect of adding some air to the cyclone entrance is shown. For the case with 8 vol% of the total air flow added to the cyclone inlet, the cyclone air-ratio is obtained by adding 0.09 ($\approx$ fraction of air $\times$ total air-ratio) to the combustor air-ratio. This means that the conditions are similar to those of the reference series, *Figure 3(a)*, except that the air-ratio is higher in the cyclone. The increased air-ratio in the cyclone has a distinct effect on N_2O and NO, which increase, and on CO and SO_2, which both decrease. The similar effect, but less strong, is seen in *Figure 5(b)* where the air in the cyclone entrance is kept constant at 3 vol% of the total air flow, giving a cyclone air-ratio which is 0.03 greater than the combustor air-ratio. A difference between the two series with air addition in the cyclone entrance compared to the other series is that the SO_2 emission increases relative to the CO emission. Although the addition of air in the cyclone entrance possibly gives some improvement when looking at N_2O and CO, *Figure 6*, it does, on the other hand, give a change for the worse when considering N_2O and SO_2. It can be concluded that, with the present boiler and the operating conditions used, addition of air in the cyclone entrance does not improve the results of reversed air staging. It can not be excluded, however, that the result would have been different in a larger boiler with a longer residence time available before the cyclone inlet.

To see how the lowest emissions can be obtained, an emission index is defined to express the total emission, but with a weight of two chosen for N_2O, to account for the two nitrogen atoms and also to emphasize the significance of N_2O:

$$\text{Emission index} = 2p_{N_2O} + p_{NO} + p_{SO_2} + p_{CO} \quad (3)$$

where p_{N_2O} etc. are the emissions in ppm normalized to a flue gas with 6 vol% oxygen. The value of the emission index for normal air-staging is about 370 ppmv. The emission index shown versus combustor air-ratio in *Figure 7*, indicates that the total emissions can be decreased compared to normal air staging in all the cases of reversed air staging studied. The two series giving lowest emissions are the series with 8 vol% air in the cyclone inlet and the series with increased temperature and limestone addition. However, of the two series only the latter reaches very low emissions of N_2O as seen in *Figure 8*, where the emission index is shown versus N_2O. The emission of N_2O can be decreased below 10 ppmv at the same time as the emission index is kept low.

In addition to CO also total hydrocarbons were measured (as methane equivalents). In connection with low CO emissions the amount of hydrocarbons was too low to give any safe results as the measured data varied from -1 to 4 ppmv. No significant difference in hydrocarbon emission compared to normal air staging was seen, but there was a very distinct correlation between hydrocarbon and CO emissions. The slope of hydrocarbons versus CO was 0.01, i.e. each increase in CO with 100 ppmv was accompanied by an increase in hydrocarbons of 1 ppmv.

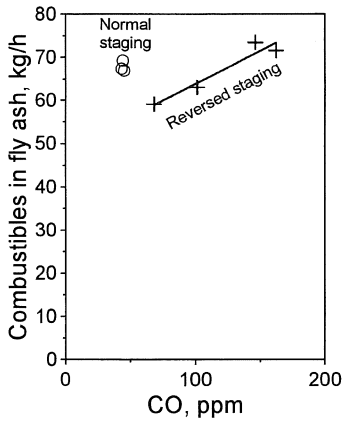


Figure 9 Loss of combustibles in fly-ash versus emission of CO. Normal air staging (○); reversed air staging (+)

Previous tests [20] indicated that the combustion efficiency was not affected by reversed air staging except in connection with high CO-emissions, above 200–300 ppmv. This is verified in *Figure 9* which shows the loss of combustible matter versus the emission of CO in the two staging cases: clearly there is no significant difference in combustion efficiency between the two staging cases. The data in *Figure 9* were obtained according to eqn (4)

$$\dot{L}_{comb} = \dot{m}_c c_c + \dot{m}_f c_f \quad (4)$$

where $\dot{m}_c$ and $\dot{m}_f$ are the exiting mass flows of ash from the secondary cyclone and the textile filter, c_c and c_f are the mass fractions of combustible matter in these flows and $\dot{L}_{comb}$ is the loss of combustible matter from the system.

DISCUSSION

The Ca/S ratios used, about 3.0, giving approximately 90 wt% sulphur retention in the normal air staging case, reflect the sulphur capture performance of the CFB boiler used. A comparison with two larger CFB boilers[22], clearly indicates that the larger boilers have a more efficient sulphur capture. This is also the case for the combustion efficiency where the smaller Chalmers boiler has a considerably lower combustion efficiency with coal firing, about 94–95%, compared to the two larger boilers.

A comparison of the emission results obtained for normal air staging from four different occasions gives an idea of the reproducibility of the results. Expressed in 1 h mean values these data typically show a small variation in CO and N_2O, $\pm$ 2 ppmv, a somewhat larger variation in NO, $\pm$ 5 ppmv, and the largest variation in SO_2, $\pm$ 20 ppmv. NO was measured with two analysers in many of the tests, and the standard deviation was about 1 ppmv. The variation in SO_2 when comparing the normal air staging runs is probably related to variations in Ca/S molar ratio caused by variations in fuel sulphur content, since the variations in limestone flow were small. Thus, it can be concluded that there is a good reproducibility for CO, N_2O and NO, while the reproducibility is lower for SO_2. Therefore the comparison of the SO_2 emission data of the various series should be made bearing in mind that the variation in SO_2 is in the order of $\pm$ 20 ppmv.

The emission of N_2O during normal air staging was considerably lower, about 60 ppmv, for the coal used in this study compared to coals of similar type previously used in the same boiler [17,20]. The reason for this is not clear, but is probably explained by a difference in the fuel properties.

CONCLUSIONS

In addition to reconfirming results previously obtained with reversed air staging, the following conclusions can be made from the study:

- A raised temperature suppresses the increase in CO observed during reversed air staging at lower air-ratios. However, the SO_2 emissions increased.
- By the combination of higher temperature and higher limestone addition rate it was possible to reach very low emissions of N_2O, in combination with low CO and SO_2 emissions. A reduction in N_2O of 90 vol% appears possible in combination with low emissions of CO, SO_2 and NO.
- Previous preliminary results of a positive effect of reduced load were not confirmed. Except for the emission of N_2O, which increased owing to the lower cyclone temperature, the effect of load was small on the other emissions.
- Tests where air is entered in the cyclone inlet, at the same time as the air-flow to the cyclone exit is varied, clearly demonstrate the important role of the cyclone. In order to obtain very low emissions of N_2O it was not sufficient to lower the air-ratio in the riser, but also the air-ratio in the cyclone had to be low.
- The combustion efficiency was not affected by reversed air staging.

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