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**16th International Conference on
Fluidized Bed Combustion**

May 13-16, 2001 • Reno, Nevada

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Co-Combustion of Dried Sewage Sludge and Coal - The Fate of Heavy Metals

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Abstract

The fate of some selected heavy metals has been investigated during co-combustion of sewage sludge and coal in two fluidised bed combustors: the 12MW_{th} circulating fluidized bed (CFB) boiler at CTH and the laboratory scale unit at IVD. The total and species balances were, in general, fairly well closed, and the increased amounts of heavy metals originating from the input sludge were also found in the exit flows. Volatile species such as Hg and Cd are sensitive to the temperature conditions. Thus, almost no mercury was found in the bottom ash, and a cyclone at 350 °C captured little Hg in comparison to a cyclone and filters at 150 °C. The high particle loading in a CFB appears to contribute essentially to the reduction of the flue gas mercury concentration; the emission is reduced by orders of magnitude compared to a plant with low particle concentration. In most cases investigated the emissions of heavy metals were below the limits related to co-combustion set by the European Union.

Introduction

The volume of sewage sludge can be reduced by gasification or combustion. Provided that drying is organised in an energy efficient way, energy can be gained from the conversion process. Here we focus the attention on the conversion process and deal with combustion of dried sludge. The simplest way to utilize the dried sludge is in co-combustion with coal or some other base fuel in an existing boiler. This means that not only volume reduction of the sludge is achieved, but also that the energy is utilized in an efficient way in a power process. Furthermore, coal is replaced, and the effective CO₂ emission is reduced. Although sludge can be converted in a dedicated plant, co-combustion is an attractive option due to its simplicity. Industrial power plants exploiting co-combustion of sewage sludge and lignite with additional flue gas clean-up have been recently implemented [1]. However, municipal sludge contains a high content of nitrogen, which may lead to increased emissions of nitrogen oxides, and, depending on the sources of sludge, higher concentrations of trace elements than those of coal [2]. This means that, if the sludge fraction is too high in co-combustion, emission limits may be exceeded. Recent studies have investigated the environmental issues dealing with sewage sludge FB incinerators [3], but less work is available on co-combustion with coal. The latter aspect of municipal sludge combustion, with coal as a base fuel in fluidized bed (FB), is the subject of the present paper. Related work concerning sewage sludge combustion can be found in ref. [4], combustion of biomass in a moving grate furnace [5], and coal combustion, [4]. The paper presents measurements of trace element emissions in the stack and in the ashes

of two FB combustors, one CFB boiler and one laboratory scale unit, fired with the same municipal sludge and coal and using the same silica sand and limestone (Ignaberga) as bed material.

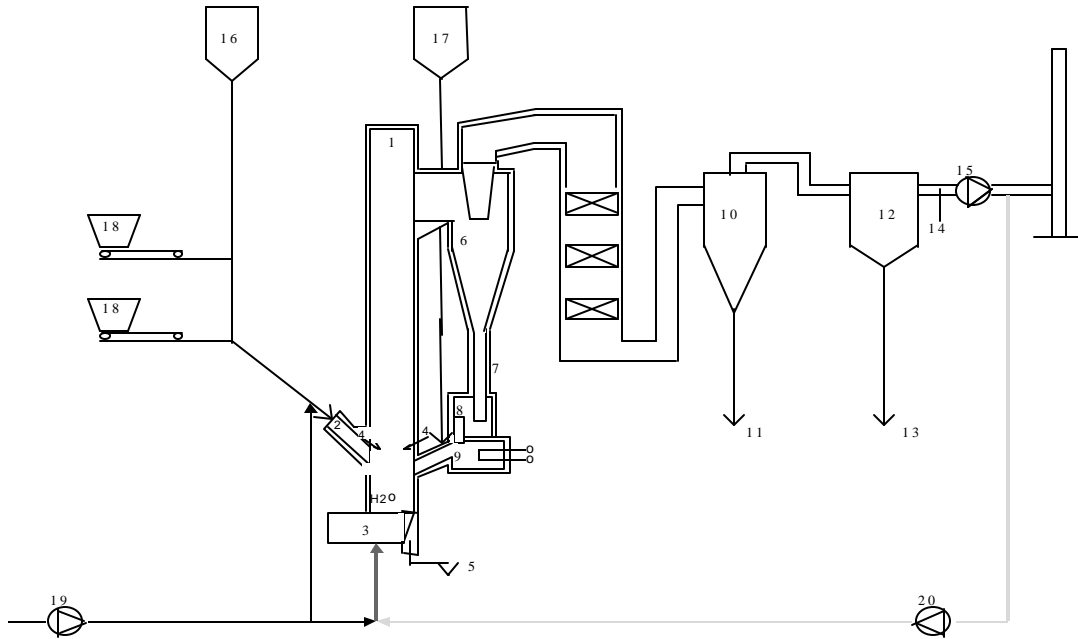


Figure. 1 The 12-MW_{th} CFB boiler at Chalmers University of Technology (1) combustion chamber; (2) fuel feed chute; (3) primary air to air plenum; (4) secondary air inlet at 2.1m; (5) bottom ash removal (6) hot primary cyclone; (7) particle return leg; (8) particle seal; (9) heat exchanger; (10) cold secondary cyclone; (11) secondary cyclone ash removal; (12) bag house filter; (13) bag house ash removal (14) probe for flue gas extraction and dust sample collection (15) flue gas fan; (16) sand bin; (17) lime bin; (18) fuel bunkers; (19) air fan; (20) flue gas recirculation fan.

Experimental background

Test units and operation

The two combustors used are the 12MW_{th} CFB boiler located at the Chalmers University of Technology (CTH), Fig. 1, and the fluidised bed test unit at the Institute of Process Engineering and Power Plant Technology (IVD), Fig. 2.

The CTH plant consists of a 14 m high furnace (1) (cross-section 2.4m²), primary loop (1-9), secondary cyclone (10), from where the ashes were not recirculated to the furnace, and a bag-house filter (12). During the present tests, samples were taken from bottom ash (5), return leg (7), secondary cyclone (10) and from the filter (12) as seen in Fig. 1.

The IVD reactor is built of a heat resistant tube (108 mm inner diameter in the bed region and 135 mm in the freeboard), of a total height of 3 m (1). Electrical heaters allow adjustment of the reactor temperature. The general features of the plant are similar

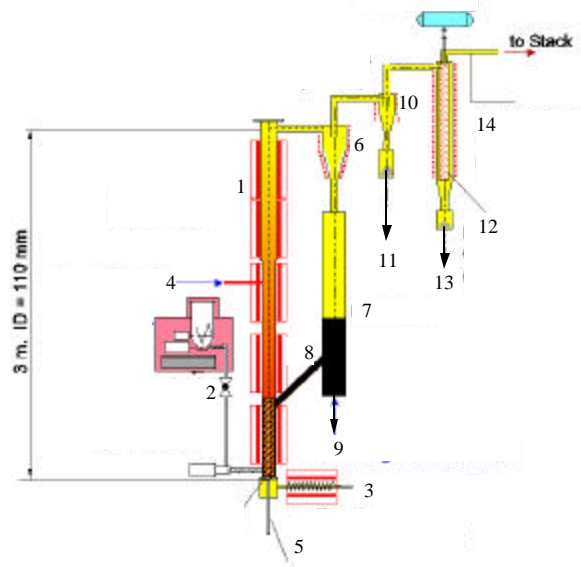


Figure 2. The test unit at IVD. (1) Combustion chamber; (2) fuel feed chute; (3) primary air; (4) secondary air inlet; (5) bottom ash removal (6) hot primary cyclone; (7) particle return leg; (8) particle seal; (9), ash removal, cyclone leg, (10) cold secondary cyclone; (11) secondary cyclone ash removal; (12) hot gas filter (ceramic); (13) ash from hot gas filter (14) probe for flue gas extraction and dust sample collection.

to the larger one at CTH: primary loop (1-9), secondary cyclone (10) and a hot gas filter (12). Samples were taken in the same locations as at CTH: (5), (11), (13), and from the return leg (9) according to Fig. 2. Sampling of flue gases leaving the test units was made in position (14) in both units.

The operating conditions in the two units were kept as constant as possible during the three tests and as similar as possible between the units. Apart from a base case with coal without sludge, two runs were performed with a moderate and with a large fraction of sludge. Normal operating conditions were used. Table 1 shows that, despite the effort to keep conditions similar, differences exist. The most significant differences, reflecting themselves in the results, are related to fluidization velocity. The larger boiler was run as a conventional circulating fluidized bed boiler with a high fluidization velocity and a high rate of recirculation of bed material. The smaller unit was operated a little above 1 m/s, and although the size of the bed material had been reduced, the degree of recirculation was small and less bed material passed the primary cyclone with the flue gases. Furthermore, as will be discussed below, the temperatures of the secondary cyclones were different in the two plants. Otherwise the differences were insignificant for the results to be presented here.

Table 1 Operating conditions

	Coal, CTH EU1	Coal, CTH EU2	Coal, CTH EU3	Coal, IVD EU1	Coal, IVD EU2	Coal, IVD EU3
Sludge,% of total energy input	0	15.4	58.1	0	15	50
Sludge,% of total fuel flow	0	23.2	69.4	0	34	68
Load, MW	7.9	8.0	8.1	0.015	0.015	0.015
Bed temp. °C (bottom)	822	851	823	875	870	860
Bed temp. °C (top)	846	860	833	850	860	845
Temp. inlet of sec. cyclone, °C	150	150	150	350	350	350
Temp. inlet of bag house/ hot gas filter, °C	150	150	150	150	150	150
Excess air-ratio	1.22	1.23	1.24	1.35	1.40	1.35
Primary air flow/total air flow, %	43	49	50	100	100	100
Superficial velocity, top of riser, m/s	6.0	6.0	6.4	1.3	1.3	1.35
Ca addition, Ca/S molar ratio	1.9	2.4	1.8	2.0	2.0	2.0
Ca/S with Ca in fuel included	2.0	2.7	2.4	2.1	2.3	2.6

Fuels

The same fuels were used in both plants, Table 2, a bituminous Polish coal and municipal sewage sludge originating from Himmerfjärden waste-water treatment plant outside Stockholm. The sludge had been subject to primary digestion before drying. Despite the drying, the water content was 25 to 30 % based on raw substance. The composition of the sludge was analysed by a specialized laboratory (CTH data) and at the University of Stuttgart (IVD-data). The sludges used at CTH and IVD were taken from the waste-water treatment plant at different occasions (considering the rather complex logistics: transport from Stockholm to Stuttgart and to Göteborg). The factors mentioned reflect themselves in the table, where differences in the data are found. The differences are however generally insignificant and do not prevent the interpretation of the results.

The selection of trace elements in this presentation has been done to compare with the regulations of the European Commission for emissions of trace elements related to co-combustion of sludge with conventional fuels. These trace elements are Cd, Tl, Hg, Sb, As, Pb, Cr, Co, Cu, Mn, Ni and V. For the comparison between the units a selection of the trace elements: Hg, Pb, Cd, Cr, Cu, Ni, and Mn were made. Zn is not on the “EU-list” and was not

Table 2. Properties of the fuels

	Bituminous coal,CTH	Bituminous coal,IVD	Sewage sludge,CTH	Sewage sludge,IVD
<i>Proximate analysis</i>				
Water (wt-%, raw)	8.9	10.1	23.9	29.1
Ash (wt-%, dry)	17.5	14.2	37.2	47.1
Combustibles (wt-%, dry)	82.5	85.8	62.8	52.9
Volatiles (wt-%, daf)	33.6	33.5	87.3	91.6
<i>Ultimate analysis (wt-%, daf)</i>				
C	82.5	81.4	53.4	45.7
H	5.0	4.2	7.1	4.3
O	9.4	12.43	30.4	44.8
S	0.80	0.78	1.90	1.98
N	1.60	1.58	7.10	3.15
Cl	0.08	0.08	0.05	0.09
<i>Lower heating value (MJ/kg)</i>				
Hu, daf	32.74	33.25	20.96	22.1
Hu, raw	24.38	28.60	9.4	8.4
<i>Ash analysis (mg/kg dry ash)</i>				
K	543	562	1700	4013
Na	123	122	575	1735
Al	2500	1238	19000	20602
Si	54667	23551	49500	52752
Fe	6933	4405	67500	48000
Ca	4467	2785	25000	16270
Mg	2333	1686	4000	3061
P	591	529	35000	4714
Ti	300	97	500	2824
<i>Trace elements (mg/kg dry fuel)</i>				
Hg	0.06	<0.1	0.75	1.86
Pb	21.0	35.7	29.0	40.5
Cd	0.1	<1	0.91	2.7
Cr	12.7	12.6	105	96.3
Cu	32.0	52.6	335	322
Ni	19.3	200.5	26.5	60.9
Mn	130	96.6	235	256
Zn	na	37.0	na	816

daf= dry and ash free, raw= as received, na=not analysed

analysed at CTH. It is, however, of interest and is therefore included in the IVD data. Particularly important for the present tests are the differences in feed of trace elements when substituting coal with sludge. A comparison of this supply of trace elements with the sludge, accounting for the concentrations according to Table 2 and the energy contents of coal and sludge, is found in Fig 3. For these particular fuels, the replacement of coal with sludge leads to an enrichment of Hg, Cu Cr and Cd in the input ash flow, whereas Mn, Ni and Pb are not significantly enriched compared to a case of pure coal.

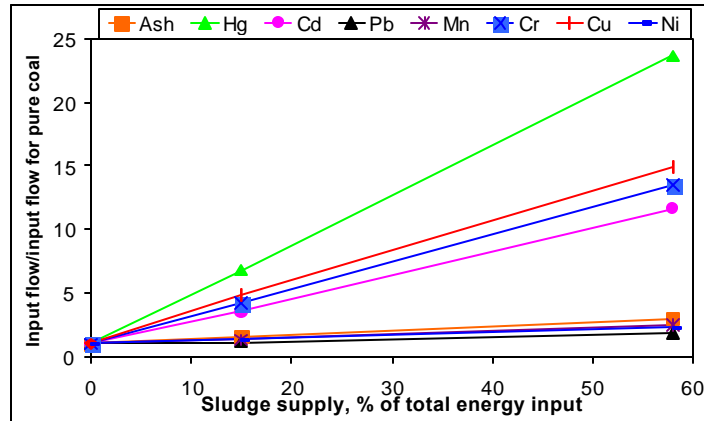


Figure 3. Relative increase of input flow to the CTH boiler of ash, Hg, Pb, Cd, Cr, Cu Ni, Mn as function of sludge supply with bituminous coal as fuel.

Test conditions and sampling methods

The test program at CTH started with the EU2 test, (see Table 1) operated for 21 hours prior to the collection of ash samples. Then followed the EU3 test that was successfully operated for 51 hours prior to the sample collection. After the EU3 test the sludge supply was shut off and 28 hours of undisturbed operation was maintained before the last samples were taken for the EU1 test. The IVD test unit is not operated continuously for more than one day at the time. The duration of the test runs at IVD under stable operating conditions was typically 4-6 hours. Between each test the bed was replaced. This could not be done in the larger boiler.

The emissions of trace elements in the flue gases leaving the boiler were determined by a standard procedure for waste incinerators. This involves isokinetic sampling at position (14) in Figs. 1 and 2, and dust collection on a heated filter. Trace elements in the gas phase were trapped in acidic solutions (in the case of Hg in acidic KMnO_4 -solution) kept in wash bottles through which the flue gas was passed.

Special attention was taken measuring Hg. Apart from being included as one of the trace elements, Hg was also measured in position (14) by an on-line flue-gas measurement system (SEMTECH Hg 2000 from Boliden Contech.) This instrument measures elemental Hg (Hg^0), but by reducing the ionic form of Hg in for example HgCl_2 (Hg^{2+}) by SnCl_2 prior to the entrance of the gas into the SEMTECH analyser, it is possible to also measure the total concentration of Hg (Hg^{tot}). This is called the gaseous concentration of Hg.

Results and discussion

Balances of solids and species

The total mass balance of solid material over a fluidised bed combustor can be expressed as:

$$\text{Mass flow of fuel ash and lime } (M_{\text{in,ash}}) = \text{Mass flow of bottom ash and fly ash } (M_{\text{out,ash}})$$

Here conversions of the lime and unburned char in the exit flows are taken into account. From these mass flows we define the ash balance ratio or the ash recovery rate = $M_{out,ash} / M_{in,ash}$. The species balances are based on total mass flows and a further measurement: the species concentrations. This balance can be expressed:

$$\text{Mass flow of species X} = \text{Mass flow of total solids} * \text{concentration of X}$$

Only small amounts (from a mass balance point of view) of species are found in the gas phase, so the closure of this balance depends on the total solids mass balance and on the measurements of species concentration. The species recovery ratios are defined for each species X as the ratio of mass flow out of X to mass flow in of X.

The total solids mass balance is difficult to close for fluidised bed combustors, and so even more the species balances. The better the closure of ash balances becomes, the firmer the conclusions of the behaviour of the trace elements. However, even a poorly closed ash balance may be acceptable for some conclusions if taken into consideration in the final judgement.

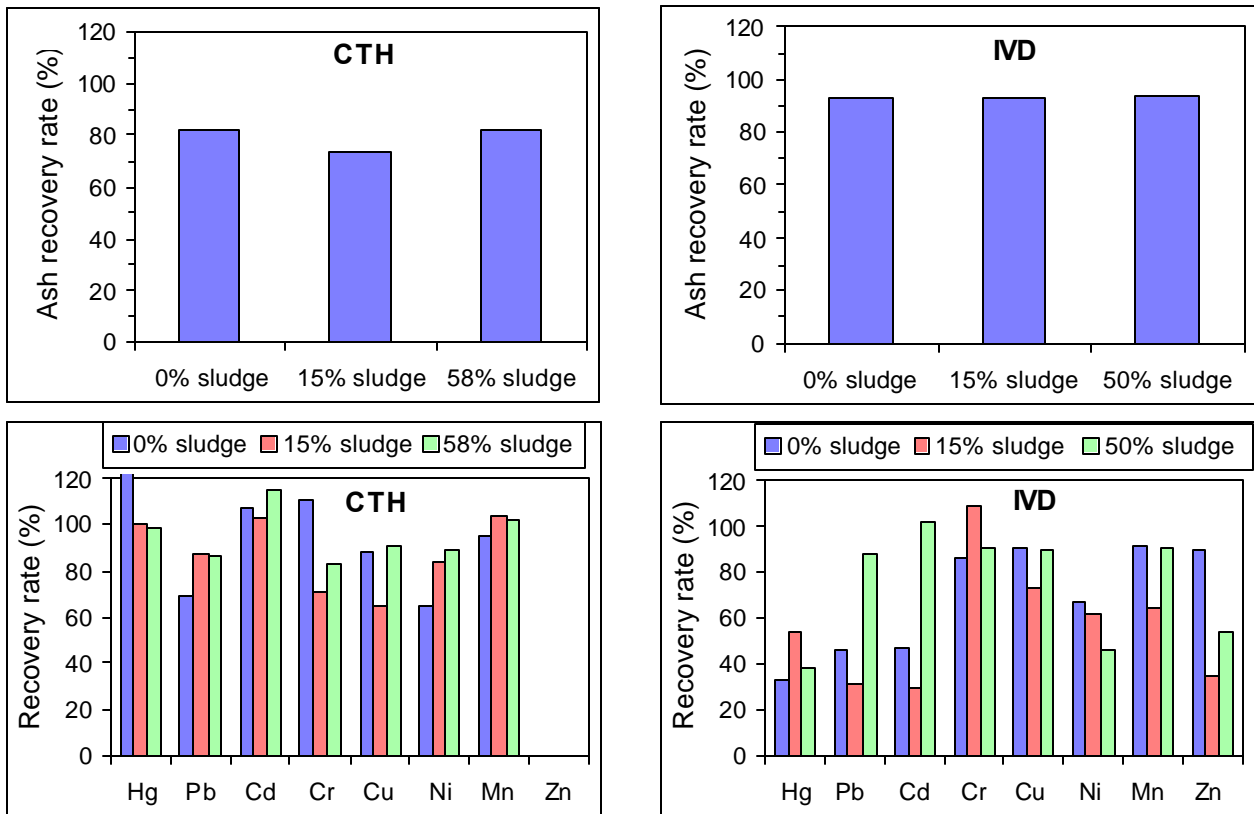


Figure 4. Comparison between the CTH and IVD tests units of the recovery rates (out/in*100) of ash and the selected trace elements. Zn was not analysed at CTH. Recovery rate for Hg, 0% sludge, CTH unit is 213%

The measurement results, presented in Fig. 4, show that the closure of the total ash balances has been fairly good in both plants. In order not to bias the species balances due to deviations in the total mass the deviation from 100% is corrected for by a correction factor in the calculation of the species balance.

The species balance is reasonably good in the CTH boiler except for Hg when no sludge was added. The reason that more Hg was found in the exit flows than added with the fuel in this test is most probably that this test was performed after the other tests, and that 21 hours is not sufficient to achieve steady state regarding Hg. Consequently, it also takes time to build up the concentration of Hg after starting the supply of sludge. This explains the low degree of closure of the Hg balance in the IVD unit, since only 4-6 hours could be used for building up the concentration of Hg before the reactor was turned down. In the IVD unit the balances are not closed for all of the other species either: more material has entered the unit than what has left. If the analyses of concentrations are correct, this means that an accumulation of these species in the bed still took place during the test runs.

Material flows

The solids flows leaving the units (now expressed in the actual flow, including: ash, CaSO₄ and CaO) are shown in Fig. 5. In the CTH boiler 71-80% of the ash is fly ash, split into secondary cyclone and bag filter flows, whereas only 20-29% is bottom ash (bed ash). This picture is typical for a CFB boiler. In the IVD unit most of the ash is bottom ash. Although the IVD unit has some recirculation of bed material from the primary cyclone, the material flows leaving the plant through the secondary cyclone and the filter were very small. This situation is more typical for a stationary, non-circulating fluidized bed combustor.

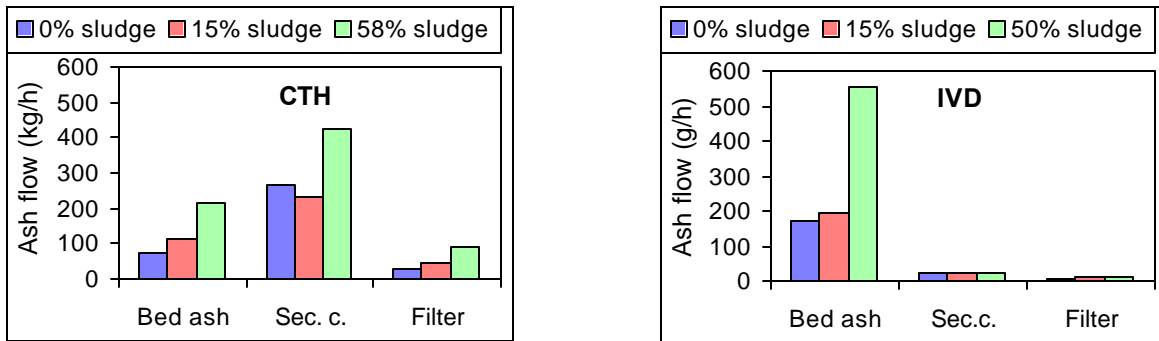


Figure 5. The solids flows through the CTH and IVD units

In both cases the increased ash flows resulting from the sludge addition are reflected in the bar diagrams.

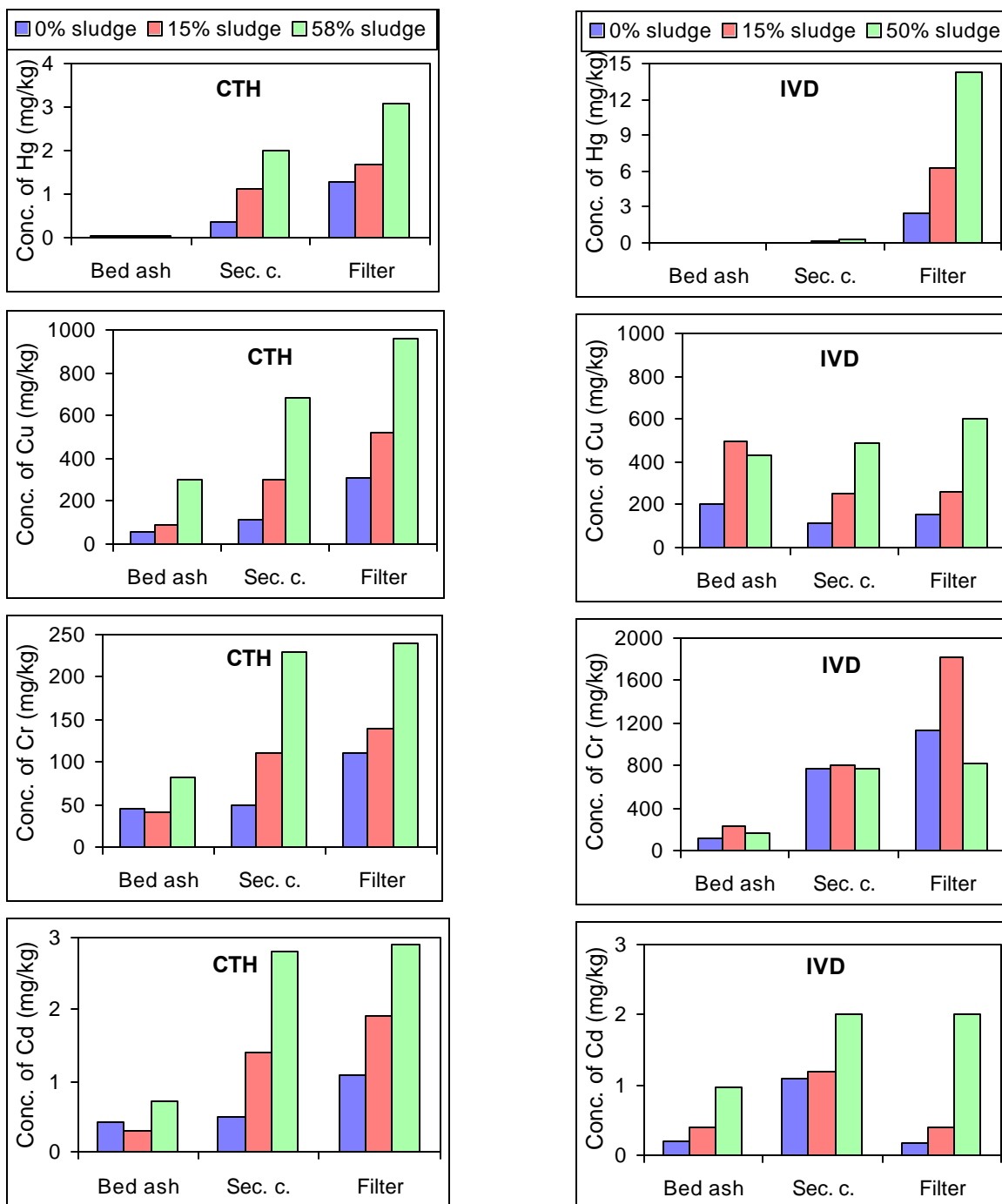


Figure 6. Concentrations of enriched species (Hg, Cu, Cr and Cd) in the exit streams. Comparison between the CTH and IVD test units for the three tests with 0, 15 and 50-58% dried sludge in coal.

Species concentration

Not only the solids flows, but also the species concentrations, are affected by the increased sludge fraction. This is caused by the concentrations of the enriched species (cf Fig. 3) Hg, Cu, Cr and Cd shown in Fig. 6 in contrast to the non-enriched species Pb, Ni and Mn shown in Fig. 7. For Cr and Ni the species concentration is an order of magnitude higher in the IVD samples than the corresponding CTH samples. This could be an effect of a contamination of the IVD samples by the alloy metals of the steel tube of the reactor. Also the higher

concentration of Ni in the fuel analysis of IVD, Table 2, could be explained in this way, since a mill of the same material was used for preparing the samples prior to analysis.

The enrichment of the species in the ash is clearly reflected in an increased concentration in all exit streams. The volatile metal species are predominant in the fly ash. Most evidently, this is the case of Hg, whose concentration is very small in the bottom ash and high in the fly ash, especially in the filter ash. It may be surprising that there is a considerable Hg concentration in the ash from the secondary cyclone of CTH but not from that of IVD. The most reasonable explanation is in the temperature of the two cyclones: 150 °C at CTH and 350 °C at IVD. (see Table 1). The volatility of mercury increases with temperature, and little was found in the beds and in the 350 °C cyclone ash. A similar tendency is not seen for the other species.

Very little work is available on the behaviour of Mn for coal and waste combustion. EPA reports that Mn generally is equally distributed between fly ash and bottom ash for FB coal combustion [7], and that for sewage sludge combustion, it is found mostly in particles larger than 4 µm. In our study Mn behaves as a non-volatile element, as well.

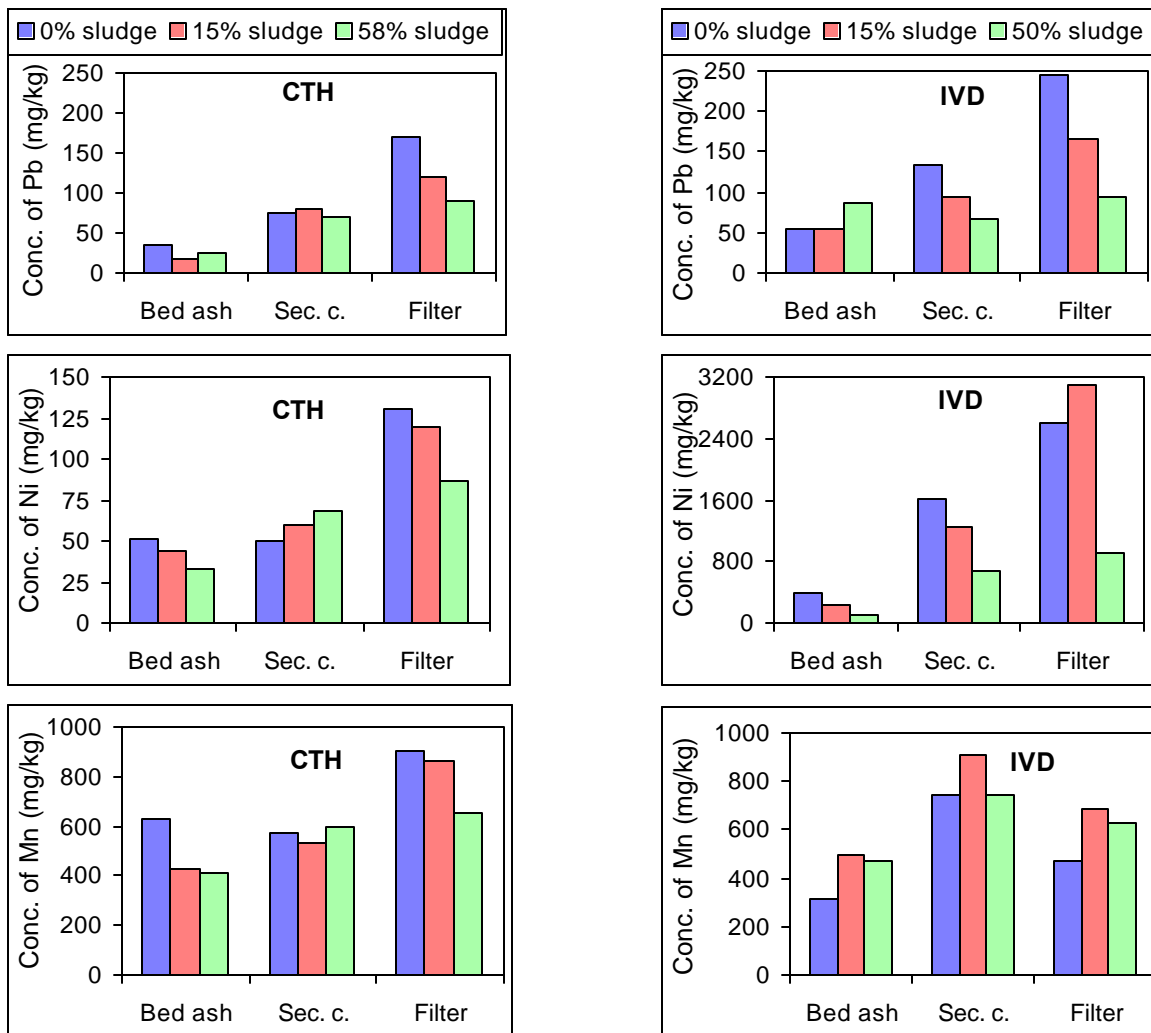


Figure 7. Concentrations of non-enriched species (Pb, Ni, and Mn) in the exit streams. Comparison between the CTH and IVD test units for the three tests with 0, 15 and 50-58% dried sludge in coal.

It is quite clear from Fig. 5 that the two plants perform in different ways as far as the ash flows are concerned. Despite these differences, the over-all picture given by the species concentrations in Fig. 6 and 7 is fairly similar. The principal differences may be found in the higher concentrations in some of the very small flows of the IVD units (compare the scales in the CTH and IVD diagrams!)

An important property of the CFB is the homogeneity of the circulating bed. This has been clearly verified by the fact (not shown in the diagrams) that the concentrations of all species in the CTH unit are the same in the bottom ash ((5) in Fig. 1) and in the circulating bed material ((7) in Fig. 1).

Emissions of heavy metals

The results of the measurements of emissions of heavy metals in the 2x3 test cases are shown both in Table 3 and Fig. 8.

Table 3 Emissions of heavy metals

	CTH EU1	CTH EU2	CTH EU3	IVD EU1	IVD EU2	IVD EU3
Energy from sludge, %	0	15.4	58.1	0.0	15.0	50
<i>Emissions, $\mu\text{g}/\text{Nm}^3$ @ 6% O₂ dry</i>						
Hg	0.679	0.061	0.120	1.50	19.00	59.00
Pb	0.772	1.366	1.830	5.30	4.80	1.94
Cd	0.029	0.038	0.028	1.40	2.13	1.39
Cr	1.69	3.36	2.56	< 1	< 1	< 1
Cu	4.59	4.37	7.97	0.400	11.42	12.78
Ni	5.75	2.35	2.51	1	< 0.6	< 0.6
Mn	3.74	5.76	27.16	38.60	11.92	29.44
Zn	na	na	na	21.35	11.33	20.78

na=not analysed

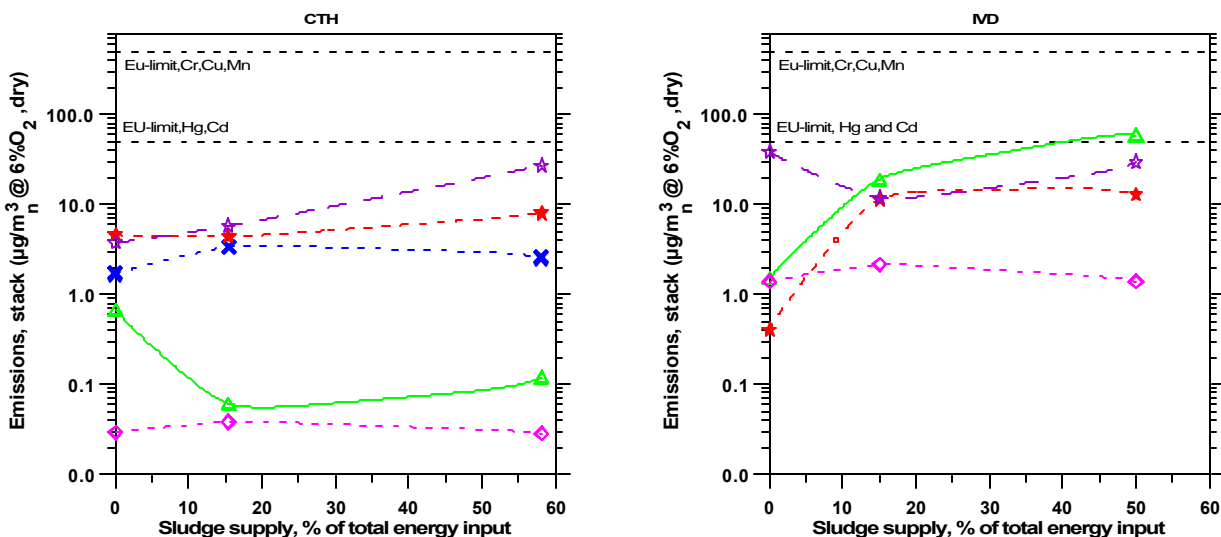


Figure 8. Emissions of heavy metals in relation to emission limits proposed by the European Commission for co-combustion of sludge with bituminous coal.

Symbols:

△ Hg ◇ Cd × Cr ★ Cu ☆ Mn

From these results the following conclusions can be made:

- 1) The emissions are usually below the emission limits set by the European Union [8] even at a high sludge fraction.
- 2) The emissions of the most volatile species, Hg and Cd, are much higher in the IVD unit than at CTH.

The principal differences between the two units lie in the temperature of the secondary cyclones, (Table 3) and in the solids flux (Fig. 4). It is likely that that the higher particle concentration (particles in general; not only submicron particles) in the CTH unit may be responsible for the differences. The results show that mechanisms like adsorption or chemical surfaces reactions of the volatile heavy metals on fly ash particles may play a major role in the heavy metal capture. However, at present this is a hypothesis and the mechanisms for removal of Hg and Cd in the fluidised bed application need to be further investigated. It is important to find out the role of various types of particles in the particle flow, including those of the textile filters. Finally, it should be emphasized that the gas concentration of Hg has been measured by two independent methods. Both methods give the same results, and this verifies the large difference in Hg emission between the IVD plant and the test unit at CTH.

On-line measurement of mercury

The SEMTECH analyser shows the instantaneous Hg concentrations. Figure 9 gives an example of a one-hour recording of the flue gas concentration in each unit. The periods measuring elemental and gaseous Hg are indicated in the figures. In the CTH unit the range of Hg is on the lower limit of calibration and no difference between the Hg-forms is detected. Apart from this, the concentration of Hg fluctuates between zero and $8 \mu\text{g}/\text{Nm}^3$ both gradually and as spikes, One could relate this as an effect of variation of the feed of Hg in the sludge. In the IVD unit corresponding variations are not clear because of the different scale.

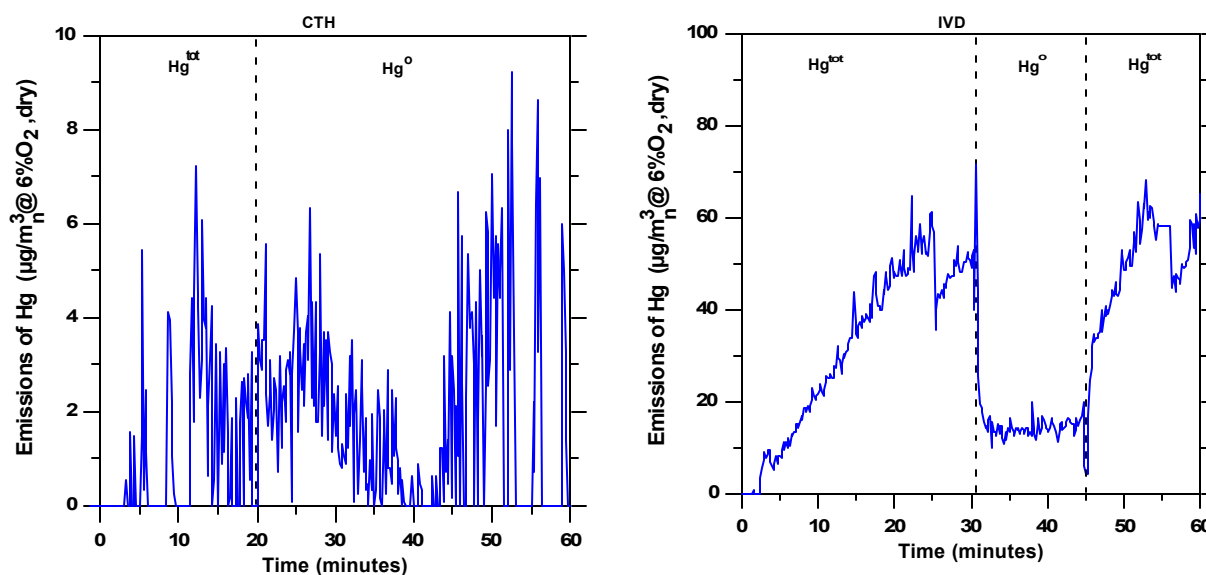


Figure 9. Comparison of the concentrations of Hg measured with SEMTECH analysers. Hg^{tot} = gaseous Hg concentration; Hg^{o} = elemental hg concentration. Fuel: 50/58% sludge with bituminous coal as base fuel. Note the different scales regarding the Hg emission.

In the IVD unit two long transients are seen. At $t=0$ the SEMTECH analyser was connected to the flue gas duct. At $t=2.5$ minutes the feed of fuel started, so the first transient is related to the start-up of the unit. But this is not the whole story. At $t=45$ minutes a shift between registration of Hg^o and Hg^{tot} was done without any change of the operating conditions. Again a delay of the signal can be seen. This delay is explained as an adsorption effect of Hg in the system. Similar transients are not observed in the CTH system, but there the signal is close to the detection limit, and it is difficult to trace this effect. The important conclusion from the SEMTECH measurement is that the difference in levels of Hg concentration between the two units is confirmed independently. This further stresses the difference between the units; Hg is captured by the secondary cyclone ash in the CTH unit but not in the IVD unit, Fig. 6. More on this issue is found in ref. [9]

Conclusions

Total mass and species balances have been made for co-combustion of sludge and coal. In general a good closure in two fluidized bed units, different in design and size, is achieved.

Both the influence of the increased ash content and the increased concentration of some species in the sludge are reflected in the effluent ash streams.

The distribution of volatile species, especially Hg, depends on the temperature. Especially important is the temperature of the separation devices. A separation device operating at 150 °C captures Hg, whereas a device operating at 350 °C does not perform well in this respect.

It appears that the high particle loading in the cold part of the gas path of a CFB has a reducing effect for Hg and Cd gaseous emissions.

Acknowledgements

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