# COMBUSTION OF KRAFT MILL BIOSLUDGE

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#### **ABSTRACT**

During trials at kraft mills, waste water treatment sludge mixed with bark was burned and samples were taken from the flue gases and ashes. The emphasis was on the determination of chlorinated dioxins, phenols and benzenes as well as PCB compounds.

The results showed that burning biosludge mixed with bark can cause the formation of chlorinated compounds in amounts that vary with different bark-fired boilers. The concentrations of chlorinated dioxins in the flue gases were lower than the guidelines applied to the old municipal waste incinerators in Sweden. All the bark-fired boilers studied were old, and some of them were converted recovery boilers. It can thus be assumed that modern bark-fired boilers fitted with advanced combustion technology are likely to give rise to less chlorinated substances than those found in this study. The risk will be reduced still further as the chlorine content of sludge falls as changes are introduced in bleaching processes.

The contents of heavy metals in the flue gases were low. Burning biosludge did not significantly affect the emissions normally encountered on burning bark.

#### **INTRODUCTION**

The activated sludge treatment of effluents from pulp and paper mills produces large amounts of biosludge. Of the total sludge produced annually by the Finnish pulp and paper industry - some 350,000 tonnes in terms of dry weight - around 50,000 tonnes is biosludge. Based on a present estimate, by the year 2000, when almost all pulp and paper

mills in Finland will be using activated sludge treatment, this figure will rise to around 65,000 tonnes a year. About 35 to 45% of all biosludge will come from the treatment of waste waters at mills producing bleached kraft pulp.

Before disposal, biosludge from activated sludge treatment is mixed with fiber-containing primary sludge from the mechanical clarification of waste water. The only feasible way to dispose of this sludge is to burn it together with bark. However, little is known about the emissions to the air caused by burning sludge. Although the chlorine content of pulp mill biosludge is lower than that of municipal waste, burning biosludge can result in the formation of chlorinated compounds such as chlorinated phenols, benzenes and dioxins (PCDD/PCDF). Some studies have reported low or non-detectable contents of PCDDs and PCDFs in flue gases and ashes from mill-scale bark and sludge-fired grate boilers (1,2,3). Recently reported results (4) from circulating fluidized bed pilot tests also show much lower PCDD/PCDF formation during combustion of biosludge than during combustion of municipal waste. According to mill-scale tests PCDDs/PCDFs possibly entering with fuel are destroyed during the combustion (3,5). However, other recently reported studies indicate that the introduction of chlorine into fuel by bark from wood previously immersed in sea water or by wood waste containing chlorinated substances would substantially increase PCDD/PCDF formation during combustion (6,7).

In the present study the burning of kraft pulp mill biosludge in bark-fired boilers was investigated. The main aim was to study the gaseous emissions from combustion, with the emphasis on chlorinated compounds, and to determine safe combustion conditions.

#### **MILL TRIALS**

# **Description of boilers**

In this study, emissions from three different types of barkfired boiler were measured: bubbling fluidized bed (BFB), circulating fluidized bed (CFB) and grate. Each of the boilers was rather old and had undergone conversion. The bubbling fluidized bed boiler (105 MW with bark fuel) was a modified sulfite waste liquor boiler. It had a rather large freeboard volume over the bed and consequently the flue gases had a long residence time in the high-temperature combustion zone. The flue gases were passed through an electrostatic precipitator (ESP). The circulating fluidized bed boiler (70 MW with bark fuel) was a converted kraft recovery boiler. It had a less than optimal bed height for bark burning. In addition, the electrostatic precipitator of this boiler operated at a high temperature (320-340°C) exceptional for a bark boiler. The grate furnace (55 MW with bark fuel) had been equipped with a moving

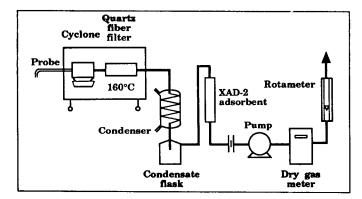


Fig. 1. Schematic drawing of the train used for sampling organic substances in flue gas.

mechanical grate. The boiler's electrostatic precipitator was under-dimensioned for full-capacity operation.

### Trials and sludge contents of fuels

Three different fuel compositions were tested in each boiler: bark alone, and bark mixed with sludge from effluent treatment with and without auxiliary fuel. The sludge used was biosludge from activated sludge treatment mixed with primary sludge and sludge from separate barking effluent sedimentation. The sludge composition and its content in the fuel were in the normal range for each mill, although in all cases the fuel sludge content was somewhat higher than average. A different auxiliary fuel was used in each boiler, as shown in Table 2.

An additional mill-scale trial (Table 3) was performed on the bubbling fluidized bed boiler in which emissions during the burning of extremely moist and high sludge-content bark on the one hand, and high loading with auxiliary fuel (coal and gas) on the other, were compared to those during the burning of fuel of normal composition.

The content of sludge in the bark mixture was estimated from the amounts of wood barked and material flows in sludge handling during the periods when the fuel mixture burned was produced.

Each trial, in which fuel composition and combustion conditions were kept constant, took about 10 hours. Two consecutive samples were taken, each over a period of about 3 hours, which was sufficient for PCDD/PCDF sampling of flue gases.

### Sampling

During the trials, the fuels, flue gases and ESP ash were sampled and flue gas composition (SO<sub>2</sub>, NO<sub>X</sub>, CO, CO<sub>2</sub>, O<sub>2</sub>) monitored continuously. The samples were analyzed for polychlorinated dibenzo-p-dioxins and dibenzofurans

Table 1. Characteristics of mixed sludge burnt.

Boiler			BFB	BFB	CFB	Grate
			Trial 1	Trial 2		
Bioslud	ge	%	80	70	29	36
Moistur	е	%	72	74	69	69
Chlorine mg/kg		14000	11500 3400		8900	
PCDD/F	PCDF	pg/g	1150	65	18	280
Metals	Pb	mg/kg	4.6	11	6.0	6.7
	Cd	00	4.3	5.3	1.1	2.0
	Cu		14	7.6	8.7	14
	Cr		20	69	23	19
	Ni		11	29	8.4	12
Al		1400	2600	1300	7800	
Fe		12700	8100	2300	4100	
Zn		260	350	200	250	
As		0.9		1.0	9.5	
	Hg		1.0		0.13	0.05

Contents based on dry solids, PCDD/PCDF as Eadon TEQ.

(PCDD/PCDF), for chlorinated phenols and benzenes, polychlorinated biphenyls (PCB) and polyaromatic hydrocarbons (PAH), and heavy metals.

Flue gas samples for analysis of organic compounds were taken with a special glass device, which is presented schematically in Fig. 1. Sampling involved sucking the flue gas isokinetically through a heated quartz probe into a chamber maintained at  $160^{\circ}$ C and containing a cyclone and a quartz fiber filter for particle separation (penetration of 0.3  $\mu$ m particles <0.002%). The gas was then cooled and condensed water collected. Finally, the gas was passed through XAD-2 adsorbent.

## **Analysis**

Particulate matter, condensed water and XAD-2 adsorbent were extracted with a suitable solvent, combined with solvent rinsings of the glass parts of the sampling device. and analyzed for PCDDs/PCDFs by high-resolution GC/MS according to refs. 8 and 9. In most cases, the particulate matter extract combined with acetone rinsings of the probe and glass parts in the chamber was analyzed separately to determine the amount of organics bound to particles; however, in some cases, extracts of all fractions - particles, condensed water and XAD resin - were combined before analysis. Altogether 17 PCDD and PCDF congeners were determined and used to calculate the Eadon (86) TCDD toxicity equivalent (TEQ). Other chlorinated compounds were analyzed by gas chromatography and PAH compounds by GC/MS using the SIM technique. The extracts were the same as in the analysis of PCDDs/PCDFs.

Table 2. Results of flue gas analysis for the bark boilers studied .

Boiler		В	FB, Trial	1		CFB			Grate	
Fuel mix		Bark	Bark sludge	Bark sludge	Bark	Bark sludge	Bark sludge	Bark	Bark sludge	Bark sludge
				gas			coal			peat
Boiler										
Output	MW	72	65	83	62	59	61	49	47	55
Bed temp.	°C	865	845	855	905	840	900			
Furnace temp.	°C	875	830	880	920	900	950	925	900	725
ESP temp.	o <sub>C</sub>	145	150	150	320	330	320	155	155	150
Fuel **										
Mixed sludge	%	İ	8.7	8.2		13.9	19.5		6.5	5.6
Moisture	%	52	57	56	55	60	58	57	58	55
Chlorine	mg/kg	250	1340	1310	120	570	520	110	1000	580
PCDD/PCDF, Eadon	pg/g	5.9	99	78	<5	1.0	1.8	2	12	13
Aux. fuel, of heat input	%			25			20			15
Flue gas, reduced to 10% oxyg	gen content		4.							
Volume	m³n/s	43	40	48	41	41	41	33	32	37
CO	mg/m³n	25	37	5	450	800	160	260	210	180
SO <sub>2</sub>	mg/m³n	26	26	25	29	21	26	17	36	49
$NO_x$ (as $NO_2$ )	mg/m³n	260	300	310	250	270	280	160	200	230
Particulate	mg/m³n	38	20	20	50	110	45	180	210	260
HCI	mg/m³n	2	5	34	2	2	5	4	5	15
Cl-phenols	μg/m³n	_	_	-	1.61	1.9	2.6	0.51	0.24	0.48
Cl-benzenes	μg/m³n	0.08	0.56	0.11	0.34	0.43	1.37	0.4	0.67	0.55
PCBs	μg/m³n	0.04	-	-	0.32	0.63	0.63	0.27	0.30	0.05
PAHs	μg/m³n	4.1	4.6	6.4	1370	850	450	8.5	6.6	1.1
PCDD/PCDF, Eadon	L-Q		-,-							
Stack	ng/m³n	0.04	0.14	0.07	0.39	0.71	1.21	0.09	1.09	0.91
Before ESP	ng/m³n	1			0.04	0.15	0.43 *			

<sup>\*</sup> Exceptionally high value of 1.8 ng/m³n in parallel sample disregarded.

### **RESULTS AND DISCUSSION**

#### Sludge characteristics

The amount of mixed sludge formed in effluent treatment at a kraft pulp mill is usually about 10% of the amount of wood waste from debarking. At the mills participating in this study the sludge content of the bark varied from about 5% to about 20%, as shown in Tables 2 and 3. The mixed sludge usually contained about one-third biosludge from the activated sludge plant. However, as shown in Table 1, at the kraft mill with the bubbling fluidized bed boiler, the biosludge dry solids represented 70-80% of the mixed sludge, because no barking sludge was added and some of the primary sludge was recovered for reuse. Because of the high biosludge content, the moisture content of the mixed sludge was high at all three mills. A high moisture content results in poor burning economy and tends to lower the bed

temperature and freeboard temperature, necessitating the use of auxiliary fuel to compensate.

The trials were conducted at a time when the trend in bleaching practice was to reduce the use of elemental chlorine, which is seen in Table 1 as the varying chlorine and PCDD/PCDF contents of sludge. The metal contents of all sludges were fairly similar, except in the cases of iron and aluminum, which at the mill with the bubbling fluidized bed boiler reflect the use of iron compounds in sludge handling, and at the mill with the grate furnace some aluminum-containing effluent coming from integrated papermaking.

# Effect of sludge

The results show that mixing sludge from effluent treatment with bark increases the formation of PCDD/PCDF compounds in the flue gases, Fig. 2. This took place at all

<sup>\*\*</sup> Contents are those for dry bark-sludge mixture.

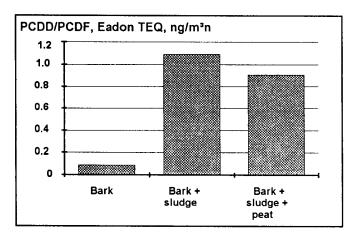


Fig. 2. Effect of inclusion in bark of sludge on flue gas PCDDs/PCDFs. Trials with the grate furnace.

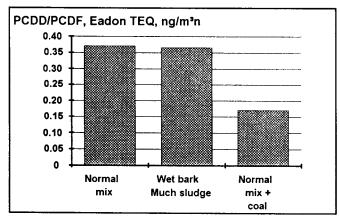


Fig. 3. PCDD/PCDF emissions during combustion of different bark sludge mixes and during increasing the boiler output with auxiliary fuel. Trial 2 with the BFB boiler.

three mills. The reason is probably the much higher chlorine content of bark fuel caused by introduction of sludge: as shown in Table 2, the chlorine content increased from 100-250 mg/kg in bark without sludge to 600-1300 mg/kg in the bark-sludge mixture, the level depending on the chlorine content of the sludge and the proportion of sludge in the fuel.

The PCDD/PCDF contents of the flue gases and ashes could not be explained by the amount of chlorinated dioxins introduced into the fuel with the sludge. This is clear from the results with the BFB boiler: during the first trial, the sludge had a high dioxin content but emissions were very low; during the second trial (Table 3) at the same mill, the PCDD/PCDF content of the sludge was low but the flue gas PCDD/PCDF emissions were higher. Balance calculations showed that the BFB boiler destroyed chlorinated dioxins but that the CFB boiler and the grate boiler were net producers of dioxins.

Table 3. Effect of fuel on emissions from combustion of bark sludge mix. BFB boiler, Trial 2.

		Normal	Wet	Normal
Fuel mix		mix	Much	mix
			sludge	+ coal
Boiler				
Output	MW	84	75	115
Bed temp.	oC	815	690	795
Furnace temp.	oC	880	825	915
ESP temp.	oC	170	175	175
Fuel *				
Mixed sludge	% ds	8	19	8
Moisture	%	57	65	59
Chlorine	mg/kg	880	1810	1310
PCDD/PCDF,Eadon	pg/g	7	15	
Gas, of heat input	%	7	14	15
Coal, of heat input	%			15
Flue gas, reduced to 10	% oxygen	content		
Volume	m³n/s	52	51	70
CO	mg/m³n	73	35	100
$SO_2$	mg/m³n	34	68	290
$NO_x$ (as $NO_2$ )	mg/m³n	290	240	350
Particulate	mg/m³n	31	29	320
Cl-phenols	μg/m³n	0.36	0.46	1.08
Cl-benzenes	μg/m³n	0.26	0.4	0.43
PCBs	μg/m³n	0.24	-	-
PAHs	μg/m³n	20	3	40
PCDD/PCDF,Eadon	ng/m³n	0.37	0.37	0.17

<sup>\*</sup> Contents for dry bark-sludge mixture.

## **Combustion conditions**

The formation of PCDDs/PCDFs in certain bark-fired boilers seems to be somewhat insensitive to combustion conditions. As shown in Table 3 and Fig. 3, the burning of a very moist (65%) bark-sludge mixture with a high (19% ds) sludge content did not result in higher formation than burning fuel of normal composition. This was the case regardless of the fact that moist fuel lowered the bed temperature to 650-700°C from its normal level of 800-850°C. However, by using gas as auxiliary fuel the freeboard temperature was maintained near normal. As shown in Table 2 and Fig. 2, the use of auxiliary fuel (15-25% on heat input to boiler) reduced only slightly the PCDD/PCDF emissions from the levels resulting from burning barksludge mixture alone. During these trials the combustion temperature was rather high, even without auxiliary fuel, and therefore rose only slightly when auxiliary fuel was used.

Trials with a 4 MW pilot incinerator were conducted during the project to determine the effect of combustion conditions on the PCDD/PCDF emissions. The pilot plant was operated in both bubbling and circulating fluidized bed incineration modes. As shown in Table 4, the PCDD/PCDF emissions to the stack were very small because of the very effective dust separation; on the other hand, PCDD/PCDF levels were high in the ESP ash and hot flue gases before dust separation. Clearly, combustion at this pilot plant was not as complete and stable as in the mill boilers, as indicated by the high carbon monoxide content of the flue gases. This explains the much higher PCDD/PCDF formation during the pilot tests. However, the results of these experiments agree with the mill trials in suggesting the insensibility of combustion conditions - as expressed by conventional control parameters - to PCDD/PCDF formation. No correlation was found between formation on the one hand, and bed temperature. excess air or carbon monoxide content on the other. In these experiments the lowest bed temperature (about 700°C) resulted in the lowest PCDD/PCDF concentration in ESP ash, which is inconsistent with the general theories about formation of chlorinated dioxins during the burning of chlorine-containing wastes.

# **Boiler specificity**

The results showed that the bark-fired boilers studied differed in respect to dioxin formation. Figure 4 shows the difference in the PCDD/PCDF content of the flue gases, which was much lower with the BFB boiler than the other two boilers. Further, the distribution of PCDDs and PCDFs in the flue gases seemed to depend on the boiler: as shown in Figure 5, about 90% of PCDDs/PCDFs were bound to particles separated in sampling in the grate furnace, whereas in the two fluidized bed boilers most of them had passed the filter. This result is consistent with the finding that the ESP ash from both fluidized bed boilers exhibited low dioxin contents, whereas the grate furnace gave a significantly higher content, Table 6.

It can be concluded from the results that the formation of PCDDs/PCDFs is more dependent on factors characteristic of each boiler and combustion technique than on the parameters used to control boiler operation. Although the results do not clearly show which factors should be considered in the prevention of dioxin formation, likely candidates include uniform and complete combustion, residence time of flue gases over different temperature ranges, and the contact of flue gases with catalytic surfaces.

Fluidized bed technology gives more uniform combustion than grate furnaces both during fluctuations in fuel heat value and in regard to freeboard cross-section. When fuel is unevenly distributed on the grate, the hot gases in the freeboard can form channels and some fuel can pass the

Table 4. PCDD/PCDF formation during pilot combustion tests.

Bed		PCDD/PCI			
temp.	$O_2$	CO	Flue	Flue gas	
			Hot	Stack	ash
°C	%	ppm	ng/m³n		pg/g
Circulating f	luidized l	oed			
708	6.1	1670	0.8	0.0	635
813	4.3	490	0.6	0.0	870
783	6.5	425	0.6	0.0	670
856	4.3	165	0.3	0.0	495
Bubbling flui	dized be				
700	3.6	1260	NA	0.0	565
803	4.6	510	5.3	0.2	3150
793	6.4	245	4.9	0.2	3800
871	4.1	620	7.2	0.0	3500

Bark-sludge mixture:

Moisture, %

Sludge content, % ds
Peat, of heat input, %

Chlorine in sludge, mg/g ds
PCDD/F in sludge, pg/g ds

Chlorine in sludge, pg/g ds

Chlorine in sludge, pg/g ds

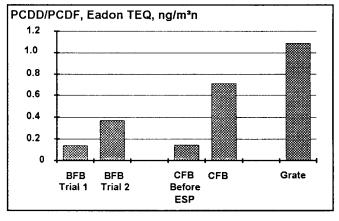


Fig. 4. PCDD/PCDF emissions from bark-fired boilers studied.

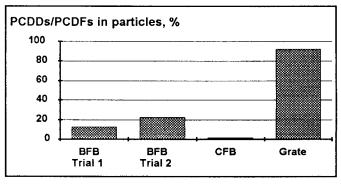


Fig. 5. Proportion of PCDD/PCDF compounds bound to particles separated in flue gas sampling.

freeboard without burning completely. In this study this was reflected in the high content of unburnt matter in ESP ash from the grate furnace, where the channeling was probably the reason for the high formation of PCDDs/PCDFs.

In the BFB boiler, which exhibited the smallest dioxin formation, the residence time of the gases in the flame temperature range 900-1150°C was relatively long, that is 3 to 3.5 seconds at the boiler outputs used. This resulted in complete combustion. On the other hand, the reactor part of the CFB boiler was too short to achieve complete combustion during incineration of bark fuel, as verified by the high PAH content of the flue gas.

On the other hand, with the pilot incinerator, the residence time of the flue gases in the hot combustion zone was estimated to be shorter during trials using bubbling fluidized bed combustion than with circulating fluidized bed combustion. Consequently, combustion might have been less complete during the bubbling fluidized bed pilot trials than during the circulating fluidized bed pilot trials, as indicated by the higher unburnt matter content of the ESP ash. It could also explain the higher PCDD/PCDF content of the ash in the former case (Table 4).

In the mill-scale CFB boiler, the formation of PCDDs and PCDFs was found to take place mainly in the hot (315-335°C) electrostatic precipitator. The same has been observed in the burning of municipal waste in incinerators with hot electrostatic precipitators. Flue gases have long residence times in hot electrostatic precipitators at temperatures at which PCDD/PCDF compounds have been found to be formed. The electrostatic field and the catalytic surface of ash particles may contribute to their formation.

The results of this study cannot be used to rank the combustion techniques in order of superiority. All three boilers studied were old, and the fluidized bed boilers had originally been designed to burn spent liquors.

## Other organic compounds

Flue gases and ESP ashes were also analyzed for chlorinated phenols and benzenes, and for PCB compounds. The levels found were low (Tables 2 and 3) and no significant difference was found between boilers. The emissions of PAH compounds were low, too, both from the grate furnace and from the BFB boiler, whereas in the CFB boiler their formation was significant, suggesting incomplete combustion in that boiler

### Heavy metal emissions

Emissions of heavy metals in the flue gases were determined during trials in which bark was mixed with sludge without

Table 5. Heavy metal emissions (10% O<sub>2</sub>).

	BFB *	CFB	Grate						
Heavy metals,	Heavy metals, mg/m³n								
Pb	0.087	0.10	0.18						
Cd	0.004	0.012	0.011						
Cu	0.084	0.13	0.042						
Cr	0.12	0.057	0.043						
Ni	0.096	0.042	0.042						
Al	0.85	7.0	3.1						
Fe	0.69	4.8	2.6						
Zn	0.32	0.69	1.9						
As	0.001	0.010	0.008						
Hg	0.004	0.005	0.004						

<sup>\*</sup> Trial 1

auxiliary fuel (Table 5). The results obtained in these short trials cannot be considered very reliable, because the scatter in the results of heavy metal emissions is usually fairly great. However, they indicated low emissions during the combustion of fuel containing sludge. The emissions measured were about one-tenth of the limits allowed in the EC directive for new municipal waste incineration plants.

#### ESP ash

During the trials, ESP ash from each boiler was analyzed to check its suitability for land applications. As shown in Table 6, the PCDD/PCDF content of the ashes from the fluidized bed boilers was low, whereas that from the grate furnace was 0.6-1.2 ng/g, which would hinder its use in land applications.

Although the sludges generally contained higher levels of heavy metals than bark, the inclusion of sludge in the fuel mix was not reflected in the heavy metal contents found in the ash from the electrostatic precipitator (with the exceptions of iron, nickel and chromium, which increased somewhat). In terms of its metal content, it can be concluded that the ash obtained from burning sludge mixed with bark is as good for land applications as the ash from burning ordinary hog fuel.

### Conventional emission parameters

As shown in Table 2, the inclusion in the fuel of normal amounts of sludge did not cause SO<sub>2</sub> emissions to rise above the very low level resulting from burning bark alone. On the other hand, the nitrogen oxide content of the flue gases rose by about 20 mg NO<sub>2</sub>/MJ.

# **CONCLUSIONS**

The content of PCDD/PCDF compounds in the flue gases varied from 0.07 to 1.2 ng/m<sup>3</sup>n as expressed in Eadon (86)

Table 6. Electrostatic precipitator ash.

Boiler			BFB, '	Trial 1	C	FB	Gı	rate
Fuel			Bark	Bark sludge	Bark	Bark sludge	Bark	Bark sludge
Unburnt		%	8	6	9	10	62	55
PCDD/PCD	F, Eadon	pg/g	71	96	22	24	390	1200
Chlorine		mg/kg	9600	5600	6900	8500	2400	6400
Metals	Pb	mg/kg	50	54	53	45	54	55
	Cd	mg/kg	12	14	9.5	8.3	4.2	5.4
	Cu	mg/kg	91	85	75	69	57	57
	Cr	mg/kg	32	59	81	120	40	56
	Ni	mg/kg	32	47	52	74	37	50
	A1	mg/kg	18000	17000	18000	22000	7400	15000
	Fe	mg/kg	16000	24000	10000	14000	6600	10000
	Zn	mg/kg	5600	3700	3600	2500	1100	1200
	As	mg/kg	17	16	15	17	7.5	17
	Hg	mg/kg	0.5	0.9	0.01	0.004	0.4	0.5
Nutrients	P	g/kg	9.2	10	9.3	7.0	3.7	4.3
	K	g/kg	34	28	16	22	15	14
	Ca	g/kg	190	170	230	210	110	98
	Mg	g/kg	18	19	16	14	9.4	8.1
	Mn	g/kg	9.6	7.9	12	8.1	4.4	4.2
	В	g/kg	0.31	0.26	0.32	0.21	0.1	0.1

equivalents. The results lead to the assumption that the PCDD/PCDF levels in the flue gases can vary from one boiler to the next, although they probably fall within the range of values measured in this study.

The emissions fell between the target values set in Sweden for waste incineration, namely 0.1 ng/m³n for new plants and 0.5 to 2.0 ng/m³n for old ones. In assessing the emissions measured in the present study, it should be remembered that all three bark-fired boilers were old and that the two fluidized bed boilers had originally been designed to burn spent liquors. New boilers employing more sophisticated combustion techniques can be expected to produce lower PCDD/PCDF emissions than those measured here. Fluidized bed boilers provide more uniform combustion conditions than grate boilers, and therefore presumably cause less formation of PCDDs and PCDFs.

Further, it should be remembered that the measurements were made at a time when pulp mills were using almost the traditional amounts of chlorine. This means the sludges had higher chlorine contents than is at present the case. In the future, as bleaching processes change, sludge chlorine levels are likely to become very low indeed, which means the emission of chlorinated compounds from burning sludge will also be very low.

## **ACKNOWLEDGEMENTS**

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