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DETERMINATION OF POLYCHLORINATED DIBENZO-P-DIOXINS AND FURANS IN THE EMISSIONS OF DISCONTINUOUS MUNICIPAL INCINERATORS

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SUMMARY

The concentrations of polychlorinated dibenzo-p-dioxins (PCDD) and furans (PCDF) in emissions (particulate matter, flue gases) from 3 discontinuous municipal incinerators (batch-wise burning process) were determined. The incinerators were located at Ulsteinvik, Vadsø and Senja. Only low levels of PCDD and PCDF were found (20 pg to 370 ng/Nm^3 for individual compounds). 2,3,7,8-tetrachlorodibenzo-p-dioxin was found in two samples (30 and 197 pg/m^3). The influence of the incineration temperature on the formation and destruction of PCDD and PCDF is discussed and the results are compared with other measurements.

LIST OF ABBREVIATIONS AND TOXICITY DATA

TCDD : tetrachlorodibenzo-p-dioxin
HCDD : hexachlordibenzo-p-dioxin
OCDD : octachlorodibenzo-p-dioxin

TCDF : tetrachlorodibenzofuran
HCDF : hexachlorodibenzofuran
OCDF : octachlorodibenzofuran

Compound	LD_{50} (µg/kg)
1,3,6,8-TCDD	> 100.000 (rat)
1,3,7,9-TCDD	
2,3,7,8-TCDD	22-45 (rat)
	0.6-2 (guinea pig)
1,2,3,6,7,8-HCDD	1250 (rat)
	70-100 (guinea pig)
OCDD	> 1.000.000 (rat)
2,3,7,8-TCDF	7 (guinea pig)

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DETERMINATION OF POLYCHLORINATED DIBENZO-P-DIOXINS AND FURANS IN THE EMISSIONS OF DISCONTINUOUS MUNICIPAL INCINERATORS

1 INTRODUCTION

Polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) are highly toxic trace compounds in the emission of municipal incinerators. A large number of investigations have been carried out to determine these substances in fly ash precipitated by electrostatic filters [1-4]. Based on these results, the burden of the emitted fly ash was estimated. However, the load of the fly ash, collected by the electrostatic filter, with PCDF and PCDD is highly dependent on the temperature of the passing flue gases and the adsorption characteristics of the ash. In addition, the precipitated fly ash contains mainly coarse particles, while the emitted part consists mainly of the fine particle fraction. Therefore, the determination of PCDD and PCDF in fly ash is not a very reliable method to estimate their emissions from incinerators.

Furthermore, most investigations were carried out on big continuous incinerators. Very little is known about PCDD and PCDF emissions from small municipal incinerators burning the garbage batch-wise. At these incinerators the operation parameters such as temperature etc. vary more than for continuous types. It is therefore of interest to investigiate whether the formation of PCDD and PCDF is favored or suppressed under these conditions. In the present investigation the content of PCDD and PCDF in the true emissions (particulate matter, flue gases) from 3 discontinuous incinerators was determined. In addition, polycyclic aromatic hydrocarbons and some selected chlorinated compounds were quantified in the same samples (5).

2 FORMATION OF PCDD AND PCDF BY COMBUSTION PROCESSES

Different investigations have shown that both PCDD and PCDF are formed during combustion processes or by thermolysis (6,7). PCDF are formed by fusion of chlorinated phenols at low temperatures and by rearrangement reactions of chlorinated benzenes and phenols at higher temperatures. PCDD are also formed by these processes. Moreover, PCDD can be created by thermal combustion from pentachlorophenol, 2,4,5-trichlorophenoxy acetic acid, polychlorinated diphenylethers, polychlorinated biphenyls etc. The highest yield is obtained in the temperature range between 300 and 620^{0}C.

In municipal incinceratorS PCDF and PCDD can be formed by the same reactions as mentioned above. In addition, different hypothesis propose a "de novo" synthesis by chlorination and oxidation of benzene and phenols followed by thermal rearrangements.

NaCl, HCl, $CHCl_3$ and chlorinated hydrocarbon radicals are considered as main sources for chlorine radicals, which are necessary for the chlorination processes. All these processes, which may form PCDD, PCDF and their precursors, are highly temperature dependent. Different investigations have shown that the formation of PCDD and PCDF is strongly suppressed at incineration temperatures above 770^{0}C. In addition the most toxic tetrachlorodibenzo-p-dioxins (TCDD) are thermolytic decomposed at temperatures > 850^{0}C. The most critical temperature range with an increased risk for PCDD and PCDF formation is between < 500 and 700^{0}C. Furthermore, at these temperatures most of the PCDD and PCDF are quite stable. Therefore, one recommends to maintain incineration temperatures above 850-900^{0}C for municipal waste incineration. A more complete survey is given in references (6, 8).

3 EMISSION SAMPLING AND QUANTITATIVE ANALYSIS OF PCDD AND PCDF

Both the applied sampling technique and the analytical method are described in details in ref. (9). A short summary is given in Table 1. The efficiency of the used extraction method was >80% and is compared with other technique in ref. (10). The efficiency of the total clean-up and preseparation procedure was tested with fly ash samples obtained from an intercalibration (11). Comparable or even higher values for 2,3,7,8-TCDD and octachlorodibenzo-p-dioxin were found. At the end of the intercalibration the mean values for the analysed fly ash sample were corrected to higher values, which agreed with our results within 20% deviation.

4 RESULTS FROM DISCONTINUOUS MUNICIPAL INCINERATORS IN NORWAY

4.1 Sampling and incineration conditions

3 discontinuous incinerators placed at Ulsteinvik, Vadsø and Senja were investigated. Table 2 summarizes the sampling conditions for the 3 incinerators. More details are given in ref. 12. The Figures 1-3 show the temperature profiles of the primary and secondary burning chamber and of the stack gases for the respective sampling days.

Table 1: Sampling and analysis of PCDD and PCDF. Step-by-step procedure.

-Isokinetic stack sampling using the following elements:

- cyclon dust collector
- heated glass fiber filter
- cooling train consisting of water cooler, 4 impingers in series at 0^0 C
- solid adsorbent cartridges (150 ml) filled with XAD-2
- sampling speed: 1-2 m^3/hour

Carried out by Central Institute for Industrial Research, Oslo.

- toluene extraction (soxhlet or liquid/liquid)

- Clean-up on florsil followed by high performance liquid chromatography on nucleosil 5 with chemically bonded NO_2-groups

- Separation by high resolution gas chromatography on SP 2330, H_2 as carrier gas

- Identification and quantification of TCDD isomers by OH^--negative ion chemical ionization mass spectrometry using multiple ion detection. Methane was used to determine PCDF and hexa- to octachlorodibenzo-p-dioxins. Alternatively electron impact ionization mass spectrometry was used to quantify PCDD and PCDF

- Detection limits (signal-to noise ratio 2:1) for 2,3,7,8-TCDD: 20 pg or 0.8 ng/m^3 (6 m^3 sample, 250 µl sample extract) for Ulsteinvik; 9 pg or 0.02 ng/m^3, 15 µl sample extract) for Vadsø and Senja.

Table 2: Sampling conditions for particulate matters and flue gases at the discontinuous incinerators of Ulsteinvik, Vadsø, Senja.

Place	Date	Sampling period	Dry gas volume [Nm^3]	Amount collected Cyclone mg	Filter mg	Condensate g
Ulsteinvik	10.11.83	11.09 to 18.30	7.8*	31*	3141	855
Vadsø						
No. 6	15.3.84	8.52 to 10.46	7.57	735	412	116
No. 7	"	11.24 to 13.26	7.89	491	565	93
No. 8	"	14.01 to 16.13	8.78	479	556	88
Senja						
No. 1	27.3.84	10.46 to 12.00	3.17	79	1173	207
No. 2	"	15.13 to 20.15	6.43	103	1946	441
No. 6	29.3.84	14.10 to 16.30	0.535	18	385	30

*Uncertain due to leakage

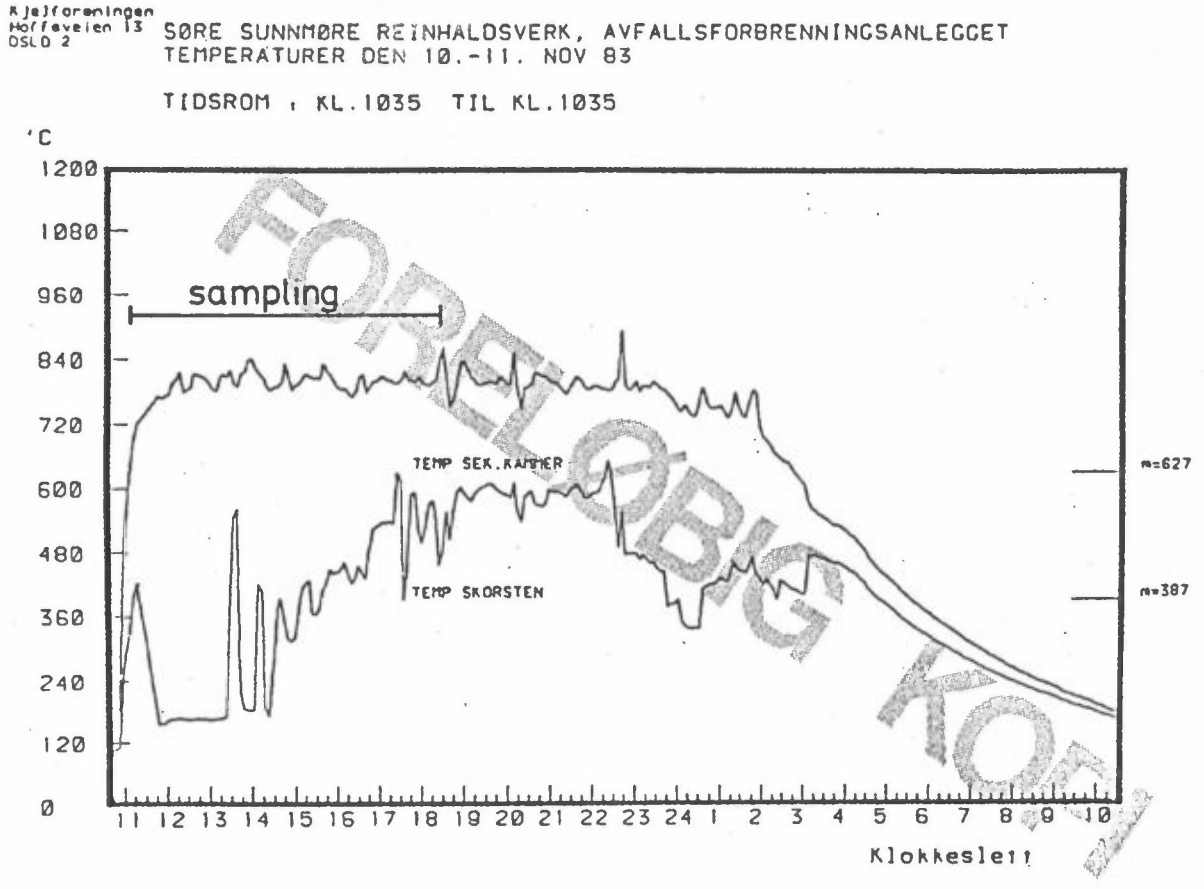

Figure 1: Temperature profile of Ulsteinvik discontinous incinerator at 10/11 november 1983. The temperature of the primary chamber was not recorded. The sampling period is marked (from Dampkjelforeningen, with permission).

BEDRIFT: VADSØ KOMMUNE	* * * K J E L F O R E N I N G E N * * *
ANLEGG : FORBRENNINGSANLEGGET	NR PARAMETER ENHET
VEDR. : DOK. AVFALL SFT	8 TEMP PRIMÆRKAMMER `C
TEMPERATURER 15.3.84	9 TEMP SEKUNDÆRKAMMER `C
ORDRENR: 10904	10 TEMP SKORSTEIN `C
DATO : 13.-15. MARS 1984	

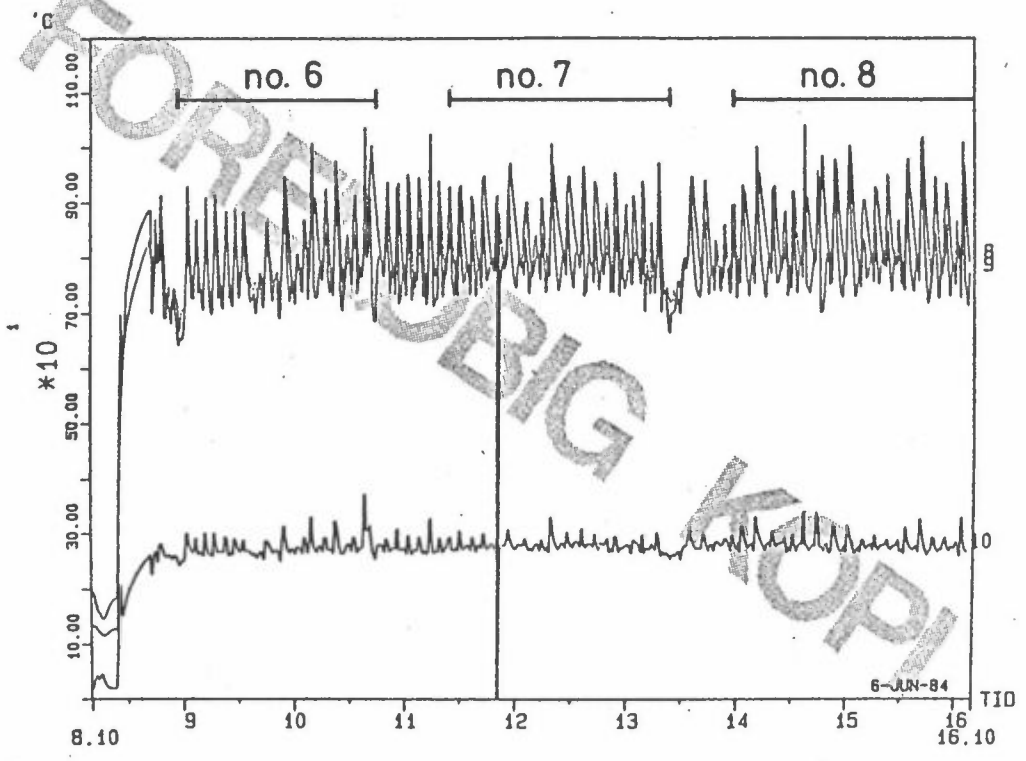

Figure 2: Temperature profile of Vadsø discontinuous incinerator at 15 March 1984. The sampling periods are marked (from Dampkjelforeningen, with permission).

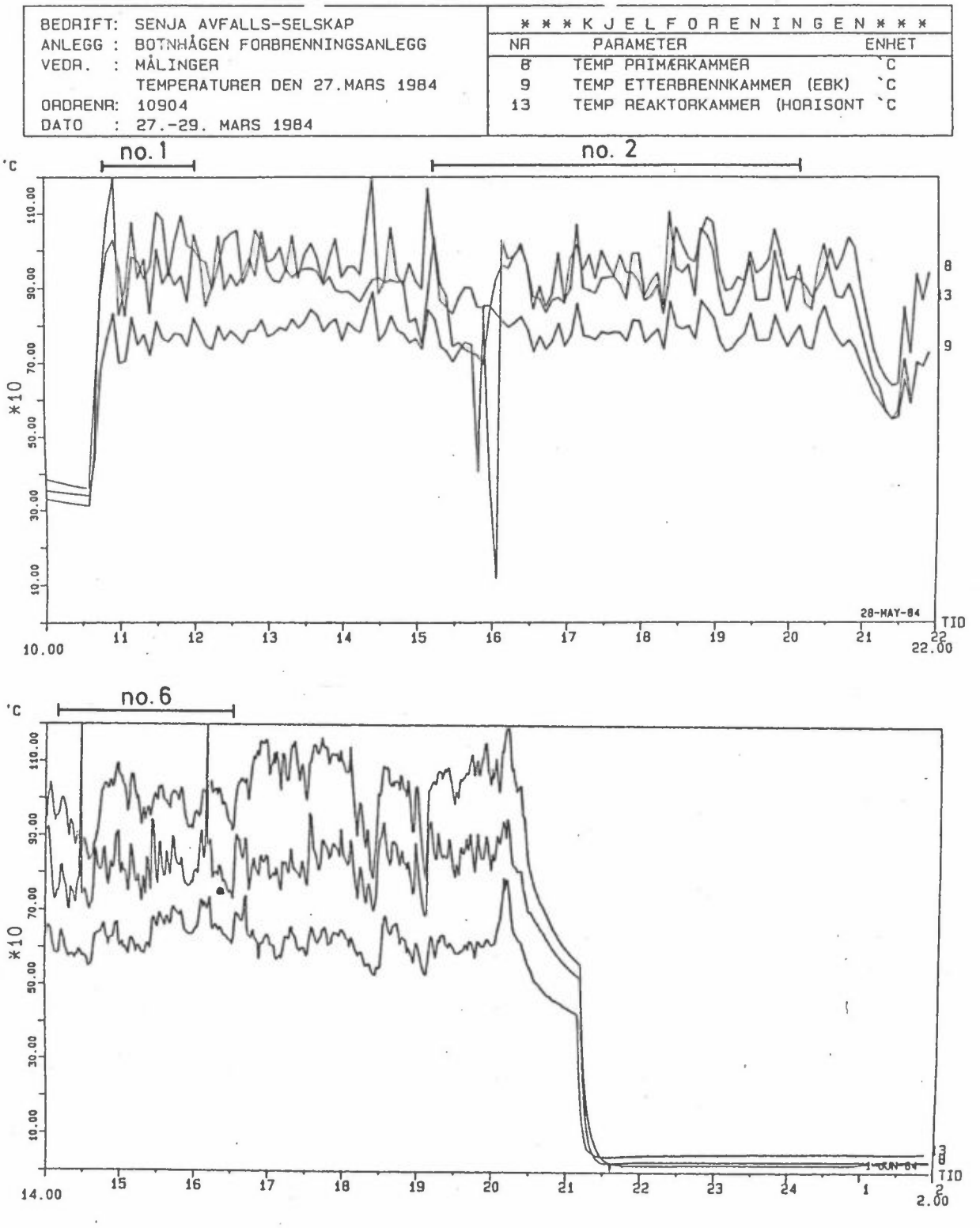

Figure 3: Temperature profiles of Senja discontinuous incinerator at 27 and 29 March 1984. At 29 March only a part of the profile was recorded. The sampling periods are marked (from Dampkjelforeningen, with permission).

The Tables 3-5 summarize the results for PCDD and PCDF in the emissions.

Table 3: Quantitative results for PCDD and PCDF for Ulsteinvik. The sample was taken 10 November 1984, 11.00 to 19.30 h.

	Filter (ng/m^3)	Condensate (ng/m^3)	XAD-2 (ng/m^3)
2,3,7,8-TCDD	n.d. (detection limit 0.2 ng/m^3 (S/N2:1))		
other TCDD	n.d.	n.d.	n.d.
2,3,7,8-TCDF	n.d.	10.1	n.d.
1,2,3,6,8,9 + 1,2,3,6,7,9-HCDD	n.d.	53	n.d.
other HCDD	n.d.	137	n.d.
1,2,3,4,7,8 + 1,2,3,4,7,9-HCDF	0.3	116(108)	n.d.
other HCDF	0.3	251(261)	n.d.
OCDD	1.6(1.9)	161(153)	n.d.
OCDF	5.1(3.3)	369(341)	n.d.

Number in parantheses are results quantified by negative ion chemical ionization mass spectrometry.

Table 4: Quantitative results for PCDD and PCDF for Vadsø. The samples were taken 15 March 1984 from 8.52 to 16.13 h.

	Cyclon + Filter (ng/m^3)	Condensate (ng/m^3)	XAD-2 (ng/m^3)
Sample 6			
2,3,7,8-TCDD	n.d.	(detection limit ⩾0.02 ng/m^3)	
other TCDD	n.d.	traces	n.d.
2,3,7,8-TCDF	n.d.	n.d.	n.d.
1,2,3,6,8,9+ 1,2,3,6,7,8-HCDD	n.d.	0.13	n.d.
other HCDD	n.d.	0.17	n.d.
1,2,3,4,7,8+ 1,2,3,4,7,9-HCDF	n.d.	0.05	n.d.
other HCDF	n.d.	0.23	n.d.
OCDD	trace	n.d.	n.d.
OCDF	0.57	n.d.	n.d.
Sample 7			
2,3,7,8-TCDD	n.d.	n.d.	n.d.
other TCDD	n.d.	traces	n.d.
2,3,7,8-TCDF	n.d.	n.d.	n.d.
1,2,3,6,8,9+ 1,2,3,6,7,8-HCDD	n.d.	n.d.	n.d.
other HCDD	n.d.	0.77	n.d.
1,2,3,4,7,8+ 1,2,3,4,7,9-HCDF	n.d.	0.18	n.d.
other HCDF	n.d.	0.57	n.d.
OCDD	trace	trace	n.d.
OCDF	0.57	0.62	n.d
Sample 8			
2,3,7,8-TCDD	n.d.	n.d.	n.d.
other TCDD	n.d.	traces	n.d.
2,3,7,8-TCDF	n.d.	0.1	n.d.
1,2,3,6,8,9+ 1,2,3,6,7,9-HCDD	trace	n.d.	n.d.
other HCDD	n.d.	0.09	n.d.
1,2,3,4,7,8+	n.d.	0.19	n.d.
1,2,3,4,7,9-HCDF other HCDD		0.52	n.d.
OCDD	n.d.	n.d.	n.d.
OCDF	0.39	0.43	n.d.

Table 5: Quantitative results for PCDD and PCDF for Senja. The samples were taken 27 March 1984 from 10.46 to 16.30 h and 19 March from 14.10 to 16.30.

	Cyclon + Filter (ng/m^3)	Condensate (ng/m^3)	XAD-2 (ng/m^3)
Sample 1			
2,3,7,8-TCDD	n.d.	traces	n.d.
other TCDD	n.d.	6.1	n.d.
2,3,7,8-TCDF	n.d.	1.9	n.d.
1,2,3,6,8,9+ 1,2,3,6,7,8-HCDD	n.d.	1.9	n.d.
other HCDD	n.d.	2.8	n.d.
1,2,3,4,7,8+ 1,2,3,4,7,9-HCDF	n.d.	5.8	n.d.
other HCDF	0.3.	24.63	n.d.
OCDD	n.d.	1.3	n.d.
OCDF	0.5	2.8	n.d.
Sample 2			
2,3,7,8-TCDD	n.d.	0.03	n.d.
other TCDD	n.d.	0.76	n.d.
2,3,7,8-TCDF	n.d.	2.8	0.07
1,2,3,6,8,9+ 1,2,3,6,7,8-HCDD	n.d.	4.2.	n.d.
other HCDD	n.d.	3.1	n.d.
1,2,3,4,7,8+ 1,2,3,4,7,9-HCDF	n.d.	4.9	n.d.
other HCDF	0.12	30.7	n.d.
OCDD	trace	3.3	n.d.
OCDF	0.3	8.4	n.d
Sample 6			
2,3,7,8-TCDD	traces	0.20	n.d.
other TCDD	0.06	4.3	n.d.
2,3,7,8-TCDF	n.d.	1.6	n.d.
1,2,3,6,8,9+ 1,2,3,6,7,9-HCDD other HCDD	n.d.	n.d. 13.7	n.d.
1,2,3,4,7,8+ 1,2,3,4,7,9-HCDF	n.d.	11.9	n.d.
other HCDD	n.d.	81.1	n.d.
OCDD	n.d.	n.d.	n.d.
OCDF	n.d.	2.4	n.d.

4.2 Results from other investigations

Only a few investigations have been carried out earlier to determine the level of PCDD and PCDF in true emissions such as particulate matter and flue gas condensate. In most cases the same sampling procedure as in the present study has been employed. Large variations between the emissions of different incinerators were observed. The concentration range for the different PCDD and PCDF isomer groups is summarized in Table 6.

Table 6: Concentration range of PCDD and PCDF isomer groups in the emissions of continuous incinerators.

	Particulate matter (ng/m^3)			Condensate (ng/m^3)		
	TCDD	HCDD	OCDD	TCDD	HCDD	OCDD
Italy, ref. (13)	Only sum dust + condensate reported			17-1127	19-3805	19-631
Italy, ref. (14)	0.04-172	0.28-12000	0.5-7300	9.6-60	178-26600	63-2700
Italy, ref. (15)	Not reported			46	18	n.d.
Switzerland, ref. (16)	3.4	22.7	47.5	0.6	2.4	1.6
	TCDF	HCDF	OCDF	TCDF	HCDF	OCDF
Italy, ref. (13)	Only sum dust + condensate reported			17-2846	22-2928	17-382
Italy, ref. (14)	n.d.-75		n.d.-2883	n.d.-1814		24-4390
Italy, ref. (25)	Not reported			994	135	n.d.
Switzerland, ref. (16)	15.5	14.3	7.7	6.8	4.3	0.5

More information about incinerator type, incineration conditions etc. are normally only given in restricted reports. Furthermore, no values for individual isomers especially for 2,3,7,8-TCDD are available in open publications. Nevertheless, Table 6 shows that the range for different incinerators varies between < 1 ng/m^3 to > 10,000 ng/m^3 for the respective isomer groups.

4.3 Discussion of the results from discontinuous incinerators

The content of PCDD and PCDF in the emissions of the 3 investigated discontinuous incinerators can be considered as very moderate. The values for Vadsø and Senja are lowest and often close to the detection limit. As can be seen from Figures 2 and 3, the incineration temperature in both the primary and secondary chamber was always around 850^0 for Vadsø and 900-1000^{0}C for Senja. At these temperatures PCDD and PCDF are very rapidly decomposed. A similar temperature profile was registered for the secondary chamber of the incinerator at Ulsteinvik. The average temperature was slightly lower (~800^{0}C). Unfortunately, no temperature profile for the primary chamber was recorded. However, the profiles of the primary chamber registered in March 1983 showed a very slow rise from 200 to 900^{0}C over a 24 hours period, while the secondary chamber temperature was more or less constant at 850^{0}C. Under these conditions, PCDD and PCDF can be formed in the primary chamber. Obviously the residence time in the secondary chamber is not long enough to decompose all PCDD and PCDF and consequently a higher emission rate was observed for Ulsteinvik than for Vadsø and Senja.

Since one did not expect rather low concentrations of PCDD and PCDF in the emissions of discontinuous incinerators, a detection limit of 1 ng/m^3 for individual TCDD isomers was considered as sufficient. This is the main reason that individual TCDD isomers were not detected in the Ulsteinvik samples. For

the analysis of the samples from Vadsø and Senja the detection limit was lowered by more than a factor of 10 (see Table 1).

Most earlier investigation have been carried out using fly ash samples. However, the loading of the fly ash with PCDD and PCDF is highly dependent on the position of the electrostatic precipitator in the stack, the flue gas temperature at that point and the adsorption characteristics of the fly ash. Furthermore, different investigations have shown that more than 90% of the PCDD and PCDF are present in the vapor phase or adsorbed to the smallest particles. Therefore the PCDD and PCDF content of fly ash samples can only be a semiquantitative indicator to compare incinerators. Based on the concentration levels found in fly ash a general rule was outlined that the content of 2,3,7,8-TCDD is about 1 to 10% of the OCDD concentration (4). However, this is only valid for particulate matter. For flue gases little is known about the ratios between different PCDD isomer classes. In addition, the distribution ratio between particulate matter and flue gases can be influenced by the sampling conditions such as the temperature of the heated filter holder, the amount of dust collected on the filter surface (acts as an adsorbent) and the flow conditions.

All TCDD isomers including 2,3,7,8-TCDD could be detected in the samples from Senja. However, the concentrations were very low. The pattern of the TCDD group was very similar to that reported for some Canadian incinerators (2,17,18). Figure 4 shows the TCDD pattern of sample No. 2 from Senja. Simultaneously polycyclic aromatic hydrocarbons (PAH) were determined in all samples. No correlation between PCDD, PCDF and PAH-concentrations was observed, which is in accordance with earlier findings (2). Other investigations which tried to compare PCDD/PCDF- and HCl-emissions together with the H_2O and O_2 content of the stack gases concluded that the PCDD/PCDF-level does not correlate with any of these parameters(4).

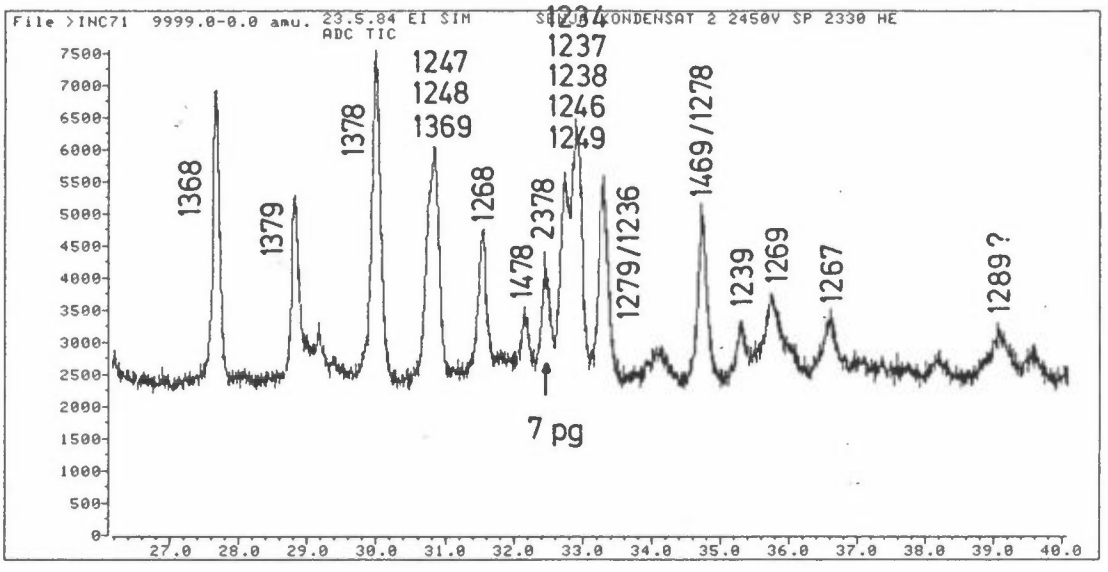

Figure 4: TCDD profile in condensate sample No. 2 from Senja discontinuous incinerator. The individual isomers are marked.

The PCDD/PCDF concentrations in the Vadsø samples were even lower than for Senja. The differences between both incinerators are difficult to explain considering the low concentration level.

The sample from Ulsteinvik contained more OCDD compared to HCDD, while in the samples from Senja more HCDD was found. Differences in the composition of the burned garbage might be an explanation. Differences in the flame chemistry caused by a possibly lower temperature in the primary chamber at Ulsteinvik is another possibility.

For all samples higher amounts of PCDF than PCDD were found. PCDF are of similar toxicity as PCDD. However, due to the increased number of isomers (135 instead of 75) and the lack of standard compounds no detailed studies about the formation and occurrence of PCDF in incinerator emissions have been carried out. Nevertheless, quantification of TCDF isomers,

which have a comparable toxicity as TCDD, should be part of any further investigation.

Some other chlorinated compounds such as polychlorinated benzenes and biphenyls as well as PAH have been quantified in some samples. The results from all samples will be discussed in a further report (5).

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ADDENDUM TO NILU REPORT OR 67/84

The following additional information was given by Lauritzen, Kjelforeningen, Oslo after the final version of the report was printed.

1. Senja (Lenvik): The garbage is continuously feeded into the primary incineration chamber and moved through different sones with increasing temperature. The residence time at the highest temperatures (> 800 ^{0}C) is longer than for comparable incinerators. In addition a considerable part of the garbage consisted of impregnated military equipment, batteries etc.

2. Vadsø: The garbage is continuously feeded into the primary incineration chamber.

3. Ulsteinvik: The whole batch of garbage is placed directly in the primary incineration chamber.

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TITTEL Determination of polychlorinated dibenzo-p-dioxins and furans in the emissions of discontinuous municipal incinerators		PROSJEKTLEDER M. Oehme	
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REFERAT (maks. 300 anslag, 7 linjer)
Konsentrasjonsnivået av polyklorerte dibenzo-p-dioxiner (PCDD) og furaner (PCDF) i utslippet av tre diskontinuerlige søppel-forbrenningsanlegg (Ulsteinvik, Vadsø, Senja) ble bestemt. Bare lave konsentrasjoner av PCDD og PCDF ble funnet.

TITLE

ABSTRACT (max. 300 characters, 7 lines)
The concentrations of polychlorinated dibenzo-p-dioxins (PCDD) and furans (PCDF) in emissions from 3 discontinuous municipal incinerators (Ulsteinvik, Vadsø, Senja) were determined. Only low levels of PCDD and PCDF were found.

*Kategorier: Åpen - kan bestilles fra NILU A
Må bestilles gjennom oppdragsgiver B
Kan ikke utleveres C