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CONCENTRATION LEVELS OF C₁-C₁₀
HYDROCARBONS AND
POLYHALOGENATED COMPOUNDS IN
THE EMISSIONS OF SMALL
HOUSEHOLD WASTE STOVES AND
A HOSPITAL WASTE INCINERATOR

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SUMMARY

High emission factors for C_1-C_4 aldehydes, C_1-C_{10} hydrocarbons, CO and particulate matter were found for a small household waste incinerator (about 10 kg waste capacity). For these compounds most emission factors were in the order of 1-10 g/kg waste. Therefore, small stove incineration as well as burning on land-fills (even poorer efficiency) can locally be a considerable source for such type of substances. Due to the poor incineration efficiency (starved air conditions, O_2 -deficiency) only occasionally traces of polyhalogenated compounds could be found in the flue gases.

Polyhalogenated compounds including polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) were identified and quantified in the emissions of a hospital waste incinerator (about 3.5 t/10 hours capacity). The emission factors per kg waste were at least one to two orders of magnitude higher than for municipal waste incinerators and were in the order of 1-10 $\mu\text{g}/\text{kg}$ for PCDD/PCDF and several hundreds of μg to mg for chlorobenzenes (CLB), polychlorinated biphenyls and chlorophenols (CP). Except for CLB and CP the total annual emissions were low due to the relatively low total amount of waste burned.

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CONCENTRATION LEVELS OF C₁-C₁₀ HYDROCARBONS AND POLYHALOGENATED
COMPOUNDS IN THE EMISSIONS OF SMALL HOUSEHOLD WASTE STOVES
AND A HOSPITAL WASTE INCINERATOR

1 INTRODUCTION

In recent investigations it has been shown that a large variety of polyhalogenated compounds are present in the emissions of small municipal waste incinerators (1,2). One major conclusion was that even at high incineration temperatures the formation of toxic polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) still is possible. Furthermore, a large amount of different polychlorinated and brominated compounds could be identified.

The aim of the present work was to study the presence of polyhalogenated compounds in the emissions of other types of waste incinerators such as hospital waste incinerators and small stoves frequently used for incineration of packing materials, household waste etc. These stoves are often found in the backyards of smaller shops or are used by households on the countryside to burn all types of waste. A previous study concluded that burning of garden refuses (leaves, twigs etc.) leads to large emissions of volatile compounds (aldehydes, C₁-C₆-hydrocarbons) due to very poor incineration conditions (3). On the basis of these results, in this study the volatility range was extended to highly volatile compound groups (halogenated and non-halogenated) to get a better survey over the total emissions of organic material in the flue gases.

Some recently published work showed that incineration of hospital waste can be a major source for PCDD/PCDF (4). Therefore, the screening of the emissions of a hospital waste incinerator for polyhalogenated substances was included in this work. The results of the described measuring program are discussed in details and compared with earlier studies.

2 EXPERIMENTAL

2.1 SMALL STOVES FOR HOUSEHOLD WASTE

Incineration of different types of waste in small stoves was simulated using the following set-up. A commercial oven type which is frequently used in Norway was employed (Typ 100, 100 l volume, about max. 10 kg waste capacity, NT3 Industrier A/S, N-3161 Stokke). It had a lid on the top equipped with a short chimney (~50 cm) and a chimney-pot. The bottom of the oven was covered with a grate and had some openings as air-intake. Three vertical, perforated channels were mounted on the interior side of the walls to increase the air supply during incineration.

Sampling of the flue gases was carried out as shown in Figure 1. The waste gases were collected and diluted in a large tube of 3 m length and 80 cm diameter, which was placed at the end of the oven chimney. A fan with a capacity of 5200 m³/h (Type ECDQ 400-4, Luftkondisjonering A/S, N-1473 Skårer) was placed on the top of tube. The high flow rate allowed to collect all flue gases and to dilute them properly before sampling. The incineration conditions and the air flow in the oven were not influenced by the large sampling tube. Samples were taken close to the top of the tube maintaining isokinetic flow conditions as far as possible.

Less volatile polyhalogenated and polycyclic compounds were collected on a standard polyurethane (PUR) foam sampler using a 3 m x 10 cm i.d. sampling tube made from aluminium. Particles were collected on glass fiber filters (Gelman, Type 61635, Ann Arbor, MI., USA) and compounds present in the vapor phase were trapped by the PUR-foam. Details of the sampling procedures are described elsewhere (5). Sampling was carried out during the whole incineration period.

CO was monitored using a continuous CO-monitor with a measuring range from 1-300 ppm (portable monitor constructed by Rockwell for Environmental Protection Agency, USA). Aldehydes were sampled using small adsorption tubes impregnated with 2,4-dinitrophenylhydrazine (6) during the whole incineration period.

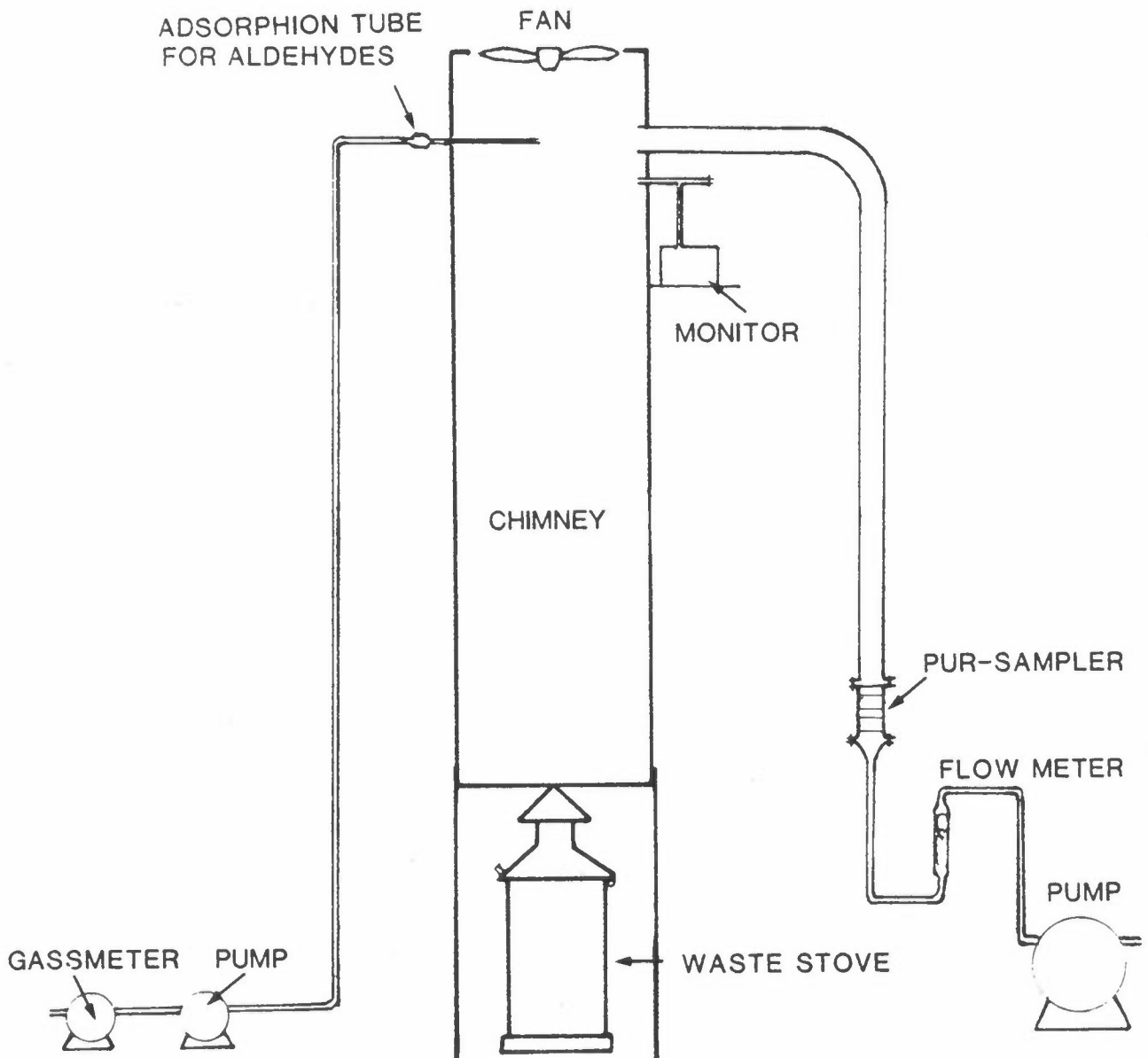


Figure 1: Experimental set-up for small stove incineration and flue gas sampling.

Grab samples of volatile hydrocarbons were taken during the most intensive burning period (about 10 min. after ignition) using aluminium coated polyethylene bags of 10 l volume. The sampling period was a few minutes.

Table 1 summarizes the conditions of the four incineration experiments which were carried out (3 with household waste, 1 with only packing materials).

Table 1: Summary of the experimental conditions for waste incineration in a small stove.

Run No.	Composition of waste	Amount burnt (kg)	Residue (kg)	Incineration time (min)	Air volume through sampling tube (m ³)
1	household ¹	2.4	1.0	35	2971
2	household ¹	3.0	1.0	50	4167
3	packing material ²	1.0	<0.1	15	1250
4	household ¹	2.9	1.4	35	2917

1 Paper, plastic, milk boxes, cardboard, food residues, glass, metal.

2 Cardboard, polyethylene, polystyrene, polyurethane foam

2.2 HOSPITAL WASTE INCINERATOR

The waste incinerator from a larger hospital in Oslo was chosen (Radiumhospitalet, Oslo, oven type Mustad L 402). The incineration capacity was about 3.5 t per batch (8-10 hours of incineration). The emitted flue gas volume was 1803 Nm³/h dry gas at 10% O₂) during the sampling period. Sampling was carried out as described earlier (1,2) by Center for Industrial Research over a 3 days period. Table 2 summarizes the sampling conditions. Particles were sampled on a preheated glass fiber filter (120^oC) and volatile compounds were collected by condensing the vapour phase at 0-10^oC. The remaining gaseous substances were trapped on XAD-2. All samples taken were collected together and analysed as one batch.

2.3 QUANTITATIVE ANALYSIS

Light hydrocarbons (C₁-C₁₂)

The grab samples collected in gas bags from the small stove experiments were analysed for C₁-C₅ compounds following the analysis

Table 2: Experimental conditions for flue gas sampling from a hospital waste incinerator (Radiumhospitalet, Oslo).

Date	Time from to	Sampled volume (dry, 10% O ₂) (Nm ³)	Collected dust (mg/Nm ³)	Oxygen content (%)
04.10.86	10.48 13.58	1.73	205	18.8
	14.05 19.17	1.99	275	19.2
05.10.86	10.45 16.32	2.79	241	18.8
	16.38 19.44	1.51	273	18.8
06.10.86	9.21 14.25	2.08	226	19.0

CO: average value <20 ppm, occasionally small spikes (~1 min) up to 120 ppm.

procedure FOG 1/79 (7). 2-3 ml were injected into a gas chromatograph equipped with a packed column. For C₆-C₁₂ compounds a sample aliquot of 100-500 ml air was trapped on a Tenax-adsorption tube. Its content was transferred by thermodesorption to a gas chromatograph equipped with a capillary column using the technique described in (8). C₆-C₁₂ compounds were identified and quantified by gas chromatography/mass spectrometry.

Aldehydes

The adsorption tubes coated with 2,4-dinitrophenylhydrazine were extracted with a small amount of acetonitrile and the formed aldehyde-hydrazone derivatives were separated and quantified by high performance liquid chromatography according to the NILU-method FOG 2/86 (6,9).

Polyhalogenated compounds Filters, PUR-foam and XAD-2 were precleaned as reported before (1,5). Toluene was used for soxhlet extraction (1,5). The sample extracts from filter and PUR-foam (small stoves) or from filter, condensate and XAD-2 (hospital waste incinerator) were collected together. Quantification of PCDD/PCDF and chlorobenzenes (CLB) was carried out as described before (1). The procedure for identification of polyhalogenated compounds has been reported in ref. 1. The sample was subfractionated by liquid chromatography and single

compounds identified by gas chromatography/mass spectrometry using electron impact and negative ion chemical ionization. Semiquantitative results were obtained using 1,3,5-tribromobenzene and octachloronaphthalene as internal standards assuming a response factor of 1 for all compounds.

3 RESULTS AND DISCUSSION

3.1 SMALL STOVE EXPERIMENTS

The averaged CO-concentration level in the collection and dilution tube was for all experiments in the order of 100 ppm (air flow 5200 m³/h). During the start-up period the upper measuring range of 300 ppm was exceeded for shorter periods. The real values are therefore somewhat higher. This indicates a rather poor incineration efficiency in the small stove. The CO-emission factors per kg waste were as high as 200-250 (!) g CO/kg waste. Figure 2 shows the CO-concentration in the flue gases during experiment No. 4 (see Table 1).

The poor incineration efficiency was confirmed by the high emission factors for aldehydes and C₁-C₁₂ hydrocarbons (see Tables 4, 5 and 6). Most of the determined values were in the g/kg waste level.

In all samples the presence of halogenated compounds was neglectable. The level of PCDD/PCDF was comparable to that found in ambient air in industrialized areas (Σ PCDD/PCDF: 10-100 pg/m³). Only in one case, traces of chlorobenzenes could be detected (run No. 2) which were above the level found in ambient air (see Table 6). No other halogenated compounds or compound classes could be detected.

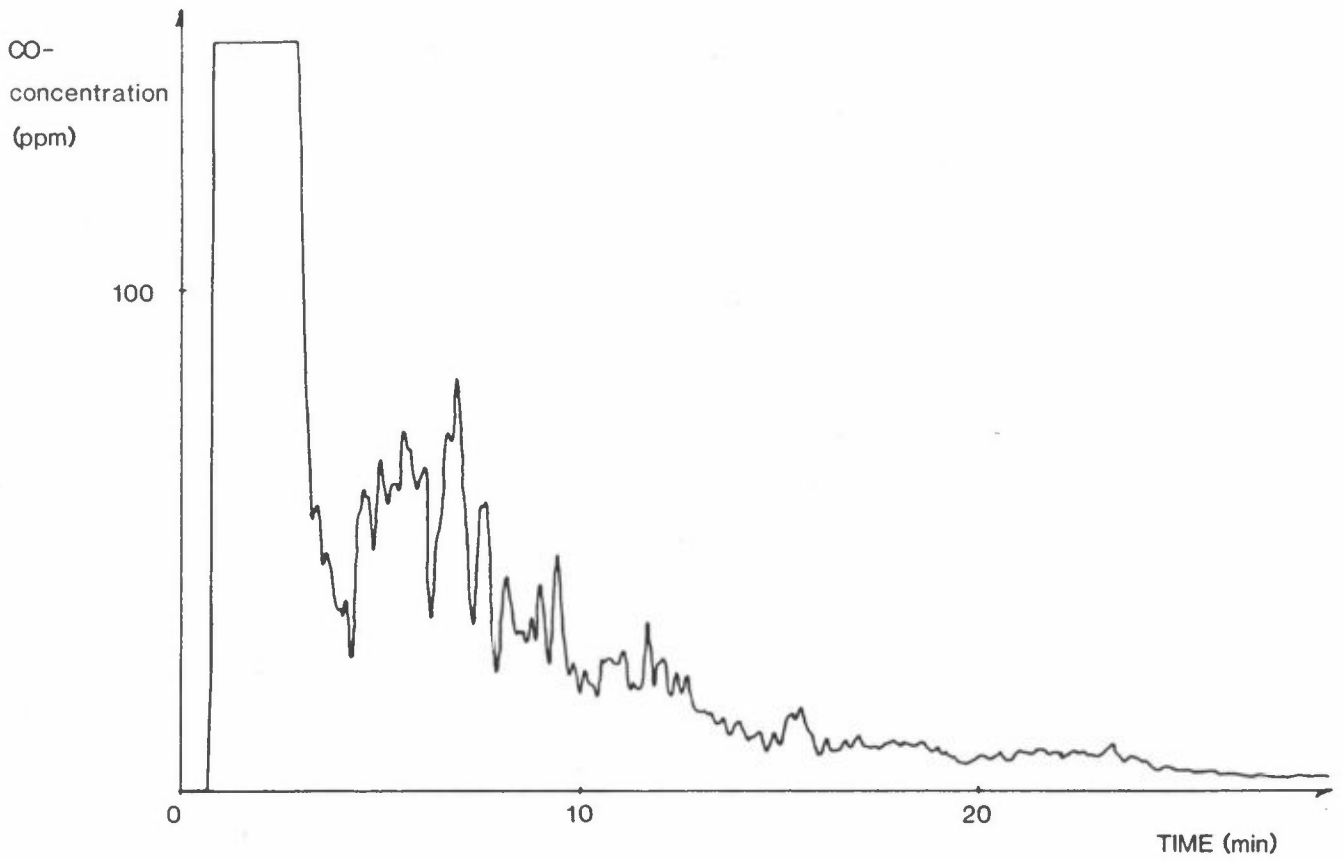


Figure 2: CO-concentration level during incineration experiment No. 4 using a small stove.

Table 3: Emission factors and concentration levels in the collection tube for different aldehydes and particulate matter from waste incineration in small stoves (see also Table 1). The values are given in g/kg burned waste or mg/m³.

Run No.	Particulate matter g/kg	Formaldehyde		Acetaldehyde		Acrolein		Aceton + Propane		Crotonaldehyde + i-butanal	
		g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³
1	22.5	1.1	0.90	0.64	0.52	0.25	0.20	0.34	0.28	<0.05	<0.04
2	32.1	2.2	1.58	1.6	1.15	0.70	0.50	0.42	0.30	0.07	0.050
3	38.3	1.2	0.96	1.0	0.80	0.20	0.16	0.25	0.20	<0.05	<0.04
4	16.1	1.6	1.59	1.1	1.09	0.15	0.15	0.17	0.17	0.05	0.05

Table 4: Emission factors and concentration levels in the collection tube for different C₁-C₅ hydrocarbons from waste incineration in small stoves (see also Table 1). The values are given in g/kg burned waste or mg/m³.

Run No.	Methane		Ethane		Ethylene		Propane		i-Butane		n-Pentane	
	g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³	g/kg	mg/m ³
1	4.7	3.9	1.7	1.34	0.12	0.1	1.9	1.55	1.6	1.35	0.6	0.51
2	5.0	3.6	0.7	0.53	0.3	0.23	2.3	1.71	0.95	0.69	0.6	0.45
3	2.2	1.8	n.d.	n.d.	n.d.	n.d.	0.17	0.14	0.4	1.89	n.d.	n.d.
4	1.91	1.90	0.14	0.14	0.04	0.04	0.38	0.38	0.92	0.92	0.15	0.15

Table 5: Emission factors and concentration levels in the collection tube for the most important C₆-C₁₂ compounds from run No. 2 (Table 1).

Compound	Concentration	
	g/kg waste	mg/m ³
Benzene	6.7	4.8
Toluene	1.4	1.0
Silicone	0.38	0.27
o-xylene	0.38	0.27
m,p-xylene	0.32	0.23
Methylpyridine	0.33	0.24
Trimethylbenzene	0.13	0.10
Silicone	0.47	0.34
Aliphatic aldehyde	0.18	0.13

Table 6: Emission factors and concentration levels in the collection tube for chlorinated benzenes from waste incineration in small stoves (run No. 2, household waste).

Compound name	Concentration	
	µg/kg waste	ng/m ³
1,4-Dichlorobenzene	n.d.	n.d.
1,3,5-Trichlorobenzene	8.3	4.0
1,2,4- "	9.8	4.7
1,2,3- "	5.2	2.5
1,2,3,5 + 1,2,4,5-Tetra-chlorobenzene	5.6	2.7
1,2,3,4-Tetrachlorobenzene	3.9	1.9
Pentachlorobenzene	n.d.	n.d.
Hexachlorobenzene	1.3	0.63

n.d. not detected

The results can be summarized as follows:

- Poor incineration efficiency (low temperature, high CO concentrations, reductive combustion conditions results) in very

high emission factors for C_1 - C_8 hydrocarbons, aldehydes, CO and particulate matter. Incineration of household and other waste in small and rather simple stoves is a considerable sources for such type of air pollutants and should therefore be avoided.

- Burning of waste on landfills undergoes at even poorer incinerations conditions, which will lead to higher emission factors for particulate matter and C_1 - C_{12} compounds than reported for small stoves. At least locally burning on landfills can therefore be a considerable source for such substances.
- Under these incineration conditions only occasionally neglectable amounts of polyhalogenated compounds are formed. This type of waste incineration is normally not a source for toxic polyhalogenated compounds such as PCDD/PCDF. Packaging material consists today mainly of cardboard, polyethylene, polystyrene and polyurethane foam. PVC is only used occasionally. No polychlorinated compounds could be detected when that type of waste was burned.

3.2 HOSPITAL WASTE INCINERATOR

Recently, relative large emissions of PCDD/PCDF have been reported when hospital waste is burned in small incinerators (4). It was therefore of interest to study the concentration level of other polyhalogenated compounds present in the emissions of a small hospital incinerator operated in a batch mode (8-10 hours during working days only). The same sampling and analysis methods as for earlier investigations were used (1,2).

In a first step chlorinated benzenes (CLB) and PCDD/PCDF were determined quantitatively. The results are summarized in Table 7 and 8. Extremely high concentration levels expressed as 2,3,7,8-TCDD equivalents according to Eadon (10) were found in the flue gases exceeding the average values for well-operated incinerators by a factor of 100. Nevertheless, the total emitted amount per year was not higher than for medium scale municipal incinerators (about 0.9 g 2,3,7,8-TCDD equivalents per year). However, a stack height of only about 10 m

(placed on the roof of a building) leads to a insufficient disperison of the emissions and most of the emitted PCDD/PCDF will be deposited within a few hundreds of meters. The concentrations for CLB were also several orders of magnitude higher than for any other incinerator studied in this project (1). An annual emission rate of about 780 g was calculated for a 200 days per year operation period.

As a next step it was tried to identify and quantify as many polyhalogenated compounds as possible in the sample extract. In opposite to other incinerators where both non-polar and very polar polyhalogenated substances could be found, the emissions of the hospital waste incinerator contained mainly non-polar polyhalogenated compounds. The more polar fractions were completely free for halogenated compounds. An exception were the very large amounts of polychlorinated phenols present in the medium-polar HPLC-fractions, which are in addition to CLB precursors for the formation of PCDD/PCDF. The concentration level in the emissions of the other investigated incinerators was at least two orders of magnitude lower (only semiquantitatively determined). A survey about identified substance classes is given in Table 9.

The results can be summarized as follows:

- The composition of hospital waste (high amount of plastic materials), occasional overloading of the oven and an upper incineration temperature limit of about 800⁰C (to avoid melting of glass residues which sinter the slag together) might lead to high concentrations of mainly non-polar polyhalogenated compounds such as PCDD/PCDF, CLB, polychlorinated biphenyls and naphthalenes as well as the more polar chlorophenols in the flue gases.

Table 7: Concentration levels for PCDD/PCDF in the emission of the hospital waste incinerator. The PCDD/PCDF-values were not corrected for recovery of ^{13}C -marked isomers added to the filter surface before sampling since the concentration range was about a factor of 100 higher than expected.

Compound	ng/Nm ³	µg/kg waste
2378-tetra-CDF	82	0.38
Σ tetra-CDF	2270	10.5
12376/12348-penta-CDF	214	0.98
23478-penta-CDF	172	0.80
Σ penta-CDF	2940	13.6
123478/123479-hexa-CDF	194	0.90
123678-hexa-CDF	456	2.1
123789-hexa-CDF	27	0.12
234678-hexa-CDF	163	0.75
Σ hexa-CDF	4300	20.0
Σ hepta-CDF	895	4.1
Octa-CDF	395	1.8
Σ totally	10'800	50.0
2378-tetra-CDD	15.4	0.071
Σ tetra-CDD	400	1.9
12378-penta-CDD	54	0.25
Σ penta-CDD	490	2.3
1234678-hexa-CDD	37.5	0.17
123678-hexa-CDD	48	0.22
123789-hexa-CDD	64	0.30
Σ hexa-CDD	640	3.0
Σ hepta-CDD	800	3.70
Octa-CDD	2970	13.8
Σ totally	5300	24.5
2378-TCDD Equivalents	280	1.3

Total annual emission assuming 200 days operation (3.5 tons, 9 hours operation per day):

Σ PCDD/PCDF: ~ 52 g/year

2,3,7,8-TCDD-equivalents (): 0.9 g/year.

Table 8: Concentration levels for chlorobenzenes in the emissions of the hospital waste incinerator.

Compound name	Concentration	
	µg/kg waste*	µg/m ³
Dichlorobenzenes	n.d.	n.d.
1,3,5-Trichlorobenzene	28.7	6.2
1,2,4- "	289	62.4
1,2,3- "	126	27.2
1,2,3,5- and 1,2,4,5-Tetra-chlorobenzene	196	42.4
Pentachlorobenzene	293	63.2
Hexachlorobenzene	183	39.6

*Assuming 3.5 t waste per day, 9 hours operation per day.

Total annual emissions rate (200 days operation):

Σ Chlorobenzenes: 780 g/year.

- The CO-level in the flue gases was rather low. In some investigations of the efficiency of municipal incinerators it has been indicated that low CO-levels are necessary to suppress the formation of PCDD/PCDF and other halogenated compounds (11). Recent findings (12) and the present study indicate that considerable amounts of PCDD/PCDF can be formed even at low CO-concentrations (< 100 ppm) and that also the O₂-concent of the flue gases influences the formation process. High O₂-levels favors the formation of PCDD/PCDF.
- The same non-polar compound classes which could be identified in a previous study (1) were also present in the emissions of the hospital waste incinerator.
- The concentration levels were at least one to two orders of magnitude higher than for small household waste incinerators. However, due to the lower amount of waste burned in the hospital incinerator the total emissions of PCDD/PCDF per year are not higher than for a medium-scale municipal waste incinerator. Nevertheless, large emissions of chlorinated benzenes and phenols (kg/year level) and an insufficient dispersion of the flue gases due to a very low stack recommends highly a redesign of the existing incinerator.

Table 9: Concentration levels for identified polyhalogenated compounds in the emission of the hospital waste incinerator. A semiquantitative determination was carried out assuming response factors of 1 for all compounds. Values for dry gas at 10% O₂.

Compound name	Molecular weight	Concentration	
		µg/kg waste*	µg/Nm ³
Pentachlorobenzene	248		See Table 7
Monobromotetrachlorobenzene	292	166	36
Monobromotrichlorobenzofurane	298	46	10
Hexachlorobenzene	282		See Table 7
Pentachlorostyrene	274	19	4
Monobromotetrachloro compound	310	14	3
Tetrachlorobenzodioxin	270	14	3
Tetrachlorohapthalenes	264	565	122
Pentachloronaphthalenes	298	815	176
Hexachloronaphthalenes	332	865	187
Heptachloronaphthalenes	366	115	25
Hexachlorobiphenylether	374	278	60
Cl ₄ Br compound	298	750	162
Pentachlorobiphenyl	324	56	12
Hexachloropiphenyl	358	90	19.5
Heptachlorobiphenyls	392	170	37
Octachlorobiphenyls	426	36	7.8
Trichlorophenols	196	1150	250
Tetrachlorophenols	230	15300	3310
Pentachlorophenol	264	9950	2150

*Assuming 3.5 t waste per day, 9 hours operation per day.
 Total annual emission rate (200 days operation):
 Σ Chlorophenols: 18.5 kg/year.

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TITTEL Concentration levels of C ₁ -C ₁₀ hydrocarbons and polyhalogenated compounds in the emissions of small household waste stoves and a hospital waste incinerator		PROSJEKTLEDER M. Oehme	NILU PROSJEKT NR. N-8327
		TILGJENGELIGHET A	OPPDRAGSGIVERS REF.
FORFATTER(E) M. Oehme, S. Manø, H. Stray, A. Mikalsen			
OPPDRAGSGIVER (NAVN OG ADRESSE) Utvalg for miljøgifter NTNF, Sognsveien 72 0801 OSLO 8			
3 STIKKORD (å maks. 20 anslag) Dioksiner Avfallsforbr. Utslipp			
REFERAT (maks. 300 anslag, 7 linjer) Utslippet fra små søppelforbrenningsovner (~100 l) og fra et sykehus-avfallsforbrenningsanlegg ble undersøkt på forekomst av C ₁ -C ₁₀ -hydrokarboner, aldehyder, polyklorerte dioksiner og andre polyhalogenerte komponenter. Utslippsfaktorene pr kg søppel var flere størrelsesordener høyere enn for vanlige forbrenningsanlegg.			

TITLE
ABSTRACT (max. 300 characters, 7 lines) The presence of C ₁ -C ₁₀ -hydrocarbons, aldehydes, polychlorinated dioxins and other polyhalogenated compounds was studied in the emissions from a small household waste stove and a hospital waste incinerator. The emissions factors per kg waste were several orders of magnitude higher than for conventional municipal waste incinerators.

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