

Co-Combustion of E+E Waste Plastics in the TAMARA Test Plant

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ABSTRACT

The co-combustion of different amounts of various plastic fractions of electrical and electronic (E+E) waste together with municipal solid waste has been tested in the Karlsruhe test incinerator TAMARA. The tests revealed no negative influences upon the combustion process. In general the increased heating value of the fuel causes an improved burnout in all residue streams. The halogens Cl and Br added with the plastics are mainly transferred as HCl or HBr into the flue gas. An influence upon the formation of chlorinated dioxins and furans could not be observed. With increasing Br feed bromine containing homologues were detected in the raw gas. The furans formed easier than the dioxins and those homologues carrying one Br atom were by far prevailing. Even at high Br input the total amount of mixed halogenated species was limited to approximately 30 % of the total load of such compounds which did not leave the typical operation window for PCDD/PCDF in TAMARA. The co-combustion tests demonstrated that MSW combustion is an ecologically acceptable and economically sound disposal route for limited amounts of specific E+E waste.

INTRODUCTION

The plastic fraction of electrical and electronic (E+E) waste is a material the disposal of which is in discussion at various forums. Different waste management routes including material recycling, chemical recycling and energy recovery are under evaluation. Municipal solid waste combustion (MSWC) as a means for recovery of the energy stored in the plastics seems to be a promising option. Existing research of various groups^{1, 2}, however, has mostly focused on assumed negative effects of heavy metals like Cd, Cu, Pb and Sb and the halogens bromine and chlorine. The effect flame retardancy related compounds or elements may have upon the formation of organic micropollutants such as chlorinated, brominated or mixed halogenated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) is controversially discussed, too.

Due to the lack of documented operational experience and to the limited awareness about bromine containing materials a research program for co-combustion experiments in a test incinerator was launched by the Forschungszentrum Karlsruhe (FZK) and a project group under the umbrella of APME (Association of Plastics Manufacturers in Europe). The goals of the project were to study the effects of

- higher E+E waste concentrations in order to simulate real life worst case conditions,
- higher concentrations of volatile heavy metals in the feed due to E+E products,
- flame retardancy related organic compounds like polybrominated diphenylethers (PBDPE), biphenyls (PBB), and tetra-bromo-bisphenol A (TBBA) as well as the active elements like bromine, phosphorous or chlorine.

The major results of the tests carried out in two test campaigns are compiled in this paper.

THE TEST INCINERATOR TAMARA

The Karlsruhe test incinerator TAMARA³ is a mass burner with a throughput of 250 kg/h of precondi-

tioned, mainly shredded waste. TAMARA has installed a reciprocating grate and the design of the combustion chamber can be changed from the original counter current configuration into a parallel flow geometry by the installation of variable roof elements.

The dedusting is performed by means of a fabric filter which is operated at approx. 200 °C. For flue gas cleaning a two-stage wet scrubbing system is implemented. The E+E material was added by a separate feeding system directly into the feeding chute. An isokinetic sampling train is installed upstream of the fabric filter in order to get representative samples of the gas phase as well as of the particulate matter in the raw gas. All other solid streams were sampled caring for a representative sampling, subsampling and sample pretreatment. The sampling and analysis of chlorinated dioxins and furans was done by FZK and simultaneously by two German commercial analytical laboratories, GfA (Münster) and ITU (Berlin).

TAMARA is equipped with a variety of monitoring systems to document all mass streams, temperatures, pressures etc. The major gas components in the raw gas are monitored on line, e.g. O₂ by an ZrO₂ electrode, CO₂, CO, HCl, SO₂, NO, N₂O, H₂O by means of spectroscopic or electrochemical methods.

MATERIALS AND TEST PROGRAM

Material Categories. Since the information on E+E waste categories is rather scarce and very little is known about the article type and the respective amounts of waste the E+E waste sourcing was from larger scale commercial operations to get representative testing materials. The pretreatment was part of the commercial operation today practiced by the following companies:

IUTA	consumer type (small appliances, kitchen machines, PC, TV, ...)
EGR	commercial / small industrial type (medium size parts, PC, fax machines, ...)
Schleswag	industrial (electrical housings, switches, main frames, ...)

The composition of the actual European average polymer mixture has been simulated by blending materials today sold on the market. Members of the APME have supplied these market typical qualities. The overall composition assumed for today's and future European market agrees very closely with the SOFRES report⁴ (PS 29%, ABS 16%, PP 11%, PA 8%, PVC 8%, PC 7%, PET/PBT 6%, PU 6%, PPO 3%, PMMA 2%, Epoxies 2%, POM 1%, Silicones 1%).

Inorganic Composition. Table 1 compiles the concentrations of some elements in the different feed streams which are characterized by a substantial additional input during the co-combustion tests. An increase in load has in particular been expected for the flame retardancy related elements Cl, Br and Sb. Especially for the industrial material a high amount of metallic species, e.g. Cu and Pb had been anticipated and were indeed found.

Organic Composition. To analyse for the organic compounds used for flame retardancy in the E+E materials is a difficult task since there is only limited information about the types of used for that purpose in plastics for the E+E market and there is no standardized method for the isolation and analytical procedure in mixed plastics. Furthermore some of the originally used materials are polymerized during the blending procedure and hence there is no way to analyze for them in the finished product.

Therefore an interlaboratory comparison of the analysis of the consumer, commercial and industrial material was initiated between two well known certified German laboratories, GfA and ITU. The analytical review was limited to three classes of compounds, that is PBB, PBDPE and TBBA.

The accuracy of these quantitative results is satisfactory considering the fact that both laboratories had different methods for sample pretreatment, e.g. solvent extraction techniques. The calibration was done with solutions of available native congeners of the compound classes to be analyzed. The PBB levels were below detection limits in all samples but in the industrial material. Here a typical concentration of

50 mg/kg was found.. PBDPE were detected in all three samples with levels ranging from 100 to 20 000 mg/kg. The amount of TBBA found in all samples ranged from 100 to 6 000 mg/kg.

The analysed flame retardants contributed only 5 – 50 % to the directly analysed Br content in the various E+E plastics. One explanation is that other compounds have been used for flame retardancy, too, the other one is that some flame retardants have been compounded into the plastic matrix and cannot be recovered any more.

Another class of organic micropollutants regulated in German ordinances for the transport and handling (German Regulations for Hazardous Materials) and for the marketing of materials (German Chemicals Banning Ordinance) are chlorinated and brominated dioxins and furans. Both ordinances set limits for single isomers like the 2,3,7,8-TCDD and for the sum of 17 chlorinated and 8 brominated isomers. Limits for mixed halogenated congeners are not specified.

In a special program APME organised the analysis not only of the TAMARA samples but of a large number of market products. In summary the concentration of halogenated dioxins and furans varies substantially ranging from < 1 to several hundred µg/kg. The average value is around 20 µg/kg and meets the German regulations. Another finding of the program was that the amount of brominated congeners belonging to the group of 8 selected PBDD/PBDF is larger than the equivalent group of 17 chlorinated congeners.

The total level of halogenated dioxins and furans in existing end of life E+E equipment suggests that material recycling of plastics from these articles should be looked at with great care. Disposal by other methods such as combustion, gasification, or thermolysis which guarantee a total destruction of these compounds is to be preferred.

Test Program. The test program comprised about two weeks of TAMARA operation. Two test runs were conducted per day, each lasting four hours. On the first and on the last day of a week two reference tests were performed burning the basic fuel only. During the tests in the first week the combustion chamber was in a parallel flow geometry, in the second week a middle flow was adjusted. The combustion temperature was kept at approx. 900 °C in the first and approx. 930 °C in the second week. The co-feeding was 3, 6 and 12 wt % in the case of the commercial and the industrial material, 6 and 12 wt % for the new and 3 wt % for the consumer material.

DISCUSSION OF RESULTS

Combustion Efficiency

Some important data for the evaluation of the experiments are compiled in Table 2. The data show the increase in heating value of the feed stream with increasing addition of E+E waste plastics. The combustion temperatures, the CO levels and the raw gas concentrations of HCl and HBr are given, too.

The combustion quality was not negatively influenced by the addition of the E+E materials. All parameters and raw gas concentrations showed rather constant numbers during the different trials. As had already experienced from the co-combustion of mixed post-consumer plastic waste the increased heating value of the plastics improved the burnout in all residue streams⁵, especially in the grate ashes as can be seen in Fig. 1. Due to the higher temperature on the back end of the grate the effect is more pronounced in the parallel flow tests. The burnout in the raw gas was extremely low in all test runs (CO < 5 mg/m³ and TOC in the fly ashes < 700 ppm) and a major difference could not be detected between the two configurations.

No traces of the brominated flame retardants analysed in the E+E materials could be detected in raw gas samples. In view of the well known high destruction potential of waste combustion⁶ this result had been

expected. Similar statements can be made for the brominated dioxins and furans. Only trace amounts of furans were detected in the raw gas of one single reference test and during the addition of 3 wt. % of commercial material. Due to the low input numbers and the high analytical error no quantitative destruction yields were calculated. The same stays for the PCDD/PCDF since the major fraction found in the raw gas has been originated in the boiler and a distinction between compounds having survived the combustion process and those having been synthesized post combustion cannot be made.

During the co-combustion of the industrial material which contained visible amounts of Cu fibres and metallic residues from the circuit boards, pure metals were found in the grate siftings. To avoid his unwanted effect a thorough metal removal of the E+E waste material is recommended.

Element Partitioning

The addition of E+E waste caused – depending upon the material – a more or less substantial increase of the halogens Cl and Br as well as of some heavy metal inventories (Sb, Cu, Pb) in the feed stream. Out of the total number the flame retardancy born elements Cl, Br and Sb will be discussed in short terms.

The analysed Cl varied between approx. 6 000 and 12 000 ppm. The comparison of this input numbers to the outputs analysed in the different compartments is shown as a percent partitioning in Fig. 2. The Cl data show a substantial scattering and the averaged output is 10 – 20 % higher than the input which is a satisfactory result for this kind of balancing. About 70 – 80 % of the total Cl inventory is found in the gas phase. The respective HCl concentrations in the raw gas cover the range of 670 to 1 000 mg/m³ (compare Table 2). A more scrutine investigation reveals a significant difference of the partitioning depending upon the geometry of the combustion chamber. During the first test campaign the fly ashes carry a significantly higher fraction of the Cl inventory than they do in the second campaign. The reason is the higher residence time of the acid HCl and the alkaline fly ashes at high temperatures in the parallel flow configuration.

This effect is more easily seen in the behaviour of Br. Fig. 3 and Fig. 4 show the sum of Br in the analysed output streams plotted against the respective number of the analysed input. Again the match of both of the Br balances can be called satisfactory. The graphs indicate an increasing incorporation of Br into the fly ashes with increasing Br input in the parallel flow co-combustion tests. At high input the curve seems to level off. In the middle flow tests when even higher amounts of Br were added along with the E+E waste only smaller fractions of Br were found in the fly ashes. This distinction is hard to be explained by the change in combustion geometry only. The analysed K and Ca concentrations suggest a higher alkalinity of the fly ashes in the first campaign.

As an example for the typical results of heavy metal balancing the percent partitioning of Zn is shown in Fig. 5. Although the variability of the data is higher than found in the case of the halogens the match of this balance can be called satisfactory, too. The scattering is more distinct for metals like Cu and Pb which are often found as pure metals in the E+E waste.

An important element involved in the flame retardancy of E+E materials is Sb. Fig. 6 compiles the percent balances for Sb as found during all of the test trials. Except for two tests, where the input was most likely underestimated, the balances close very well.

PCDD / PCDF in the Raw Gas

Former experiments in TAMARA did not show an effect even if the Cl load was elevated by a factor of five⁵. This has been supported by similar tests in a German full scale MSW incinerator⁷ and by two recently published studies upon the relationship of PVC and PCDD/F in waste combustion⁸. There is one paper claiming an increase in PCDD/F if electronic waste flame retarded by polybrominated diphenylethers is co-combusted in a MSWC plant⁹. This could not be confirmed by TAMARA tests on the co-combustion of flame retarded XPS and PUR foams¹⁰.

The levels of PCDD and PCDF in the raw gas of TAMARA tend to decrease with increasing time of operation. Hence the matching reference level of PCDD/F for a certain co-combustion test is somewhere between the respective initial and final reference test results. The PCDD and PCDF concentrations analysed by FZK, GfA and ITU were in most cases very close to each other. The raw gas concentrations of PCDD and PCDF are compiled in Table 3. Fig. 7 depicts the sum of homologues as a function of the Br level in the feed stream. The data show the typical scattering. A significant increase or homologue change caused by the co-combustion of the flame retarded E+E materials is not revealed.

Br Containing Dioxins and Furans in the Raw Gas

This situation changes as far as bromine containing homologues are concerned. According to literature and to the above cited TAMARA tests brominated and mixed halogenated compounds add only a small percentage to the PCDD/PCDF levels in MSWC^{10, 11, 12}. The sum concentrations of bromine containing dioxins and furans are compiled in Table 3 which also contains the Br input data. Fig. 8 and Fig. 9 show the sum of Br containing homologues analysed in the raw gas of TAMARA. There were next to none purely brominated homologues found. The prevailing homologues analysed were those containing one Br atom. Homologues containing two Br atoms were only detected if very high amounts of Br were added and those containing more than two Br atoms could not be detected. The formation of bromine containing furans exceeded that of dioxins.

The formation of bromine containing furans increases with increasing Br input and seems to level off at high Br loads (compare Fig. 10). The effect is less pronounced for the dioxins the concentrations of which, however, are very low. These findings confirm the results of the above cited foam co-combustion tests.

The graph in Fig. 11 demonstrates that the Br containing furans in these tests sometimes reached the same order of magnitude than the purely chlorinated ones. This has been observed in the two tests when 12 wt. % of commercial or industrial E+E material were co-combusted. The dioxins were always lower in concentration by a factor of 3 – 4 than the furans and did not add substantially to the total sum of the halogenated compounds in question.

In all tests the total dioxin/furan load in the raw did not exceed the typical operation window of TAMARA which is shown in the background of Fig. 11 and covers the range of 20 – 120 ng/m³. Samples taken after the flue gas had passed a charcoal filter resulted in residual PCDD/F concentrations of <0.001 ng/m³. Bromine containing homologues could not be detected. This means that the contribution of brominated species is of no concern for the emission of MSW incineration plants as far as a modern air pollution control system is installed.

CONCLUSIONS

Co-combustion tests of different types E+E waste and MSW in the Karlsruhe test incinerator TAMARA were performed looking for real effects in a modern MSW combustor. Up to 12 wt. % of E+E material was added to the basic fuel. The following most important results were obtained:

- The combustion process was not negatively influenced even if high amounts (up to 12 wt. %) of E+E material were added to the basic fuel.
- The increase in heating value improved the burnout of the bottom ashes.
- Residual organic compounds indicating unburnt flame retardants could not be detected in the flue gas.
- The levels of Cl in the feed were moderately, those of the flame retardancy related elements Br and Sb as well as those of a number of heavy metals were substantially increased by the addition of the E+E wastes. Some metals, especially Cu and Pb may be present in the metallic state and tend to sift through the grate. Hence a thorough metal separation from the E+E plastic waste prior to combustion is strongly recommended.
- The increased Cl and Br levels caused no significant increase of the concentrations of PCDD or

PCDF in the raw gas.

- The formation of limited amounts of brominated and chlorinated dioxins and furans was observed with the furans exceeding the dioxins by a factor of 3 – 4. For both classes of compounds mainly homologues carrying one Br atom were detected.
- At high input of E+E waste Br containing homologues added up to 50 % to the total load of dioxins and furans. The data obtained did not leave the typical operation window of TAMARA.
- Air emission sampling during some co-combustion tests resulted in PCDD/PCDF concentrations of $<0.001 \text{ ng/m}^3$ showing that activated carbon is effectively removing the moderate to low PXDD/PXDF concentrations in the raw gas to almost zero levels.

The co-combustion experiments did not show severe influences upon the quality of waste incineration. The expected promoting effect upon the production of Br containing dibenzo-p-dioxins and dibenzofurans stayed moderate even if high amounts of E+E material were added. In full scale incineration plants the co-feeding of high calorific waste fractions will be limited to 3 – 5 wt % as a worst case. In those cases the increase in dioxin and furan levels will stay within the window of the PCDD/F levels typically found in such incineration plants.

On the basis of the results presented here it seems a fair statement:

- MSW combustion is an ecologically acceptable and economically sound disposal route for limited amounts of specific E+E waste. The metal content, however, should be reduced as is done today in most recycling operations in order to reduce the maintenance of the grate and to minimise the amount of metallic species in the grate siftings.

ACKNOWLEDGMENTS

This project has been supported by the following associations: APME (Association of Plastics Manufacturers in Europe), BFRIP (Brominated Flame Retardant Industry Panel), EBFRIIP (their European equivalent), EFRA (European Flame Retardant association), FIEE (Fédération Française des Industries Electriques et Electronics), FZK (Forschungszentrum Karlsruhe), ZVEI (Zentralverband der Elektro- und Elektronikindustrie).

A number of people from different companies have contributed to the performance of the project: H. Geisert, H. Gramling, R. Härtel, H. Reis, R. Siegel (FZK), J.M. Falguière (Du Pont), R. Martin (APME), W. Pauli (ABB), M. de Poortere (EBFRIIP/BFRIP/EFRA), M. Rohr (Bayer). The authors are very grateful for all of these contributions.

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Compound	Concentration (µg/m³)	Reference	Year	Location	Notes	Other
PCB	0.001	1	1985	Germany		
PCB	0.002	2	1986	Germany		
PCB	0.003	3	1987	Germany		
PCB	0.004	4	1988	Germany		
PCB	0.005	5	1989	Germany		
PCB	0.006	6	1990	Germany		
PCB	0.007	7	1991	Germany		
PCB	0.008	8	1992	Germany		
PCB	0.009	9	1993	Germany		
PCB	0.010	10	1994	Germany		
PCB	0.011	11	1995	Germany		
PCB	0.012	12	1996	Germany		
PCB	0.013	13	1997	Germany		
PCB	0.014	14	1998	Germany		
PCB	0.015	15	1999	Germany		
PCB	0.016	16	2000	Germany		
PCB	0.017	17	2001	Germany		
PCB	0.018	18	2002	Germany		
PCB	0.019	19	2003	Germany		
PCB	0.020	20	2004	Germany		
PCB	0.021	21	2005	Germany		
PCB	0.022	22	2006	Germany		
PCB	0.023	23	2007	Germany		
PCB	0.024	24	2008	Germany		
PCB	0.025	25	2009	Germany		
PCB	0.026	26	2010	Germany		
PCB	0.027	27	2011	Germany		
PCB	0.028	28	2012	Germany		
PCB	0.029	29	2013	Germany		
PCB	0.030	30	2014	Germany		
PCB	0.031	31	2015	Germany		
PCB	0.032	32	2016	Germany		
PCB	0.033	33	2017	Germany		
PCB	0.034	34	2018	Germany		
PCB	0.035	35	2019	Germany		
PCB	0.036	36	2020	Germany		
PCB	0.037	37	2021	Germany		
PCB	0.038	38	2022	Germany		
PCB	0.039	39	2023	Germany		

Table 1 Concentrations of selected elements in the materials of the 2nd campaign (data in mg/kg, consumer data of 1st campaign)

	basic fuel	consumer	commercial	industrial	new material
P	2080	250	2600	8100	1200
Cl	6150	3895	10990	1865	38550
Cu	112	1900	82	77500	137
Zn	430	620	1540	4720	292
Br	34	4290	15000	5750	8600
Cd	9	240	123	29	41
Sn	24	85	63	1230	18
Sb	26	2000	1950	2200	2630
Pb	600	146	96	1890	1890

Table 2 Lower heating value of feed, temperature in the combustion chamber, oxygen, HCl and HBr in the raw gas for all test trials

	H _l kJ/kg	T °C	O ₂ vol %	CO mg/m ³	HCl mg/m ³	HBr mg/m ³
ref.1.2	6960	894	9.7	1.2	730	3
cons.3%	7950	900	9.9	1.7	680	9
comm.3%	7910	912	9.9	2.1	900	204
ind.3%	7960	901	10.0	1.7	840	72
ind.6%	8480	893	10.4	3.6	850	131
ref.2.1	7440	892	9.9	1.2	750	3
ref.3.1	8880	930	10.0	0.0	790	4
ref.3.2	8770	912	10.3	0.1	760	3
n.mat.6%	10580	923	10.5	1.8	870	63
n.mat.12%	12440	949	10.5	0.0	1010	119
comm.6%	10260	926	10.5	0.1	860	157
comm.12%	12760	937	10.9	0.2	940	292
ind.12%	10840	932	10.6	0.3	800	141
ref.4.1	8840	939	9.9	0.3	670	4

Table 3 Br input and raw gas concentrations of chlorinated and mixed halogenated dioxins and furans

test	Br input mg/kg	PCDD ng/m ³	PCDF ng/m ³	Br _x Cl _y DD ng/m ³	Br _x Cl _y DF ng/m ³
ref.1.2	67	15.0	20.2	0	0
cons.3%	270	9.8	12.1	0	0
comm.3%	1979	19.8	23.6	3.13	12.4
ind.3%	606	12.6	14.2	1.2	5.0
ind.6%	1193	14.4	16.2	2.0	13.5
ref.2.1	68	8.4	10.1	0	0
ref.3.1	57	16.0	27.2	0.5	2.5
ref.3.2	45	18.2	28.2	0.5	2.5
n.mat.6%	757	16.3	27.8	1.1	7.1
n.mat.12%	1650	20.1	31.0	2.5	20.9
comm.6%	1297	8.8	11.7	1.9	11.4
comm.12%	2735	14.0	15.5	5.8	20.6
ind.12%	1000	14.9	16.9	6.1	15.6
ref.4.1	38	9.6	13.6	1.2	4.3

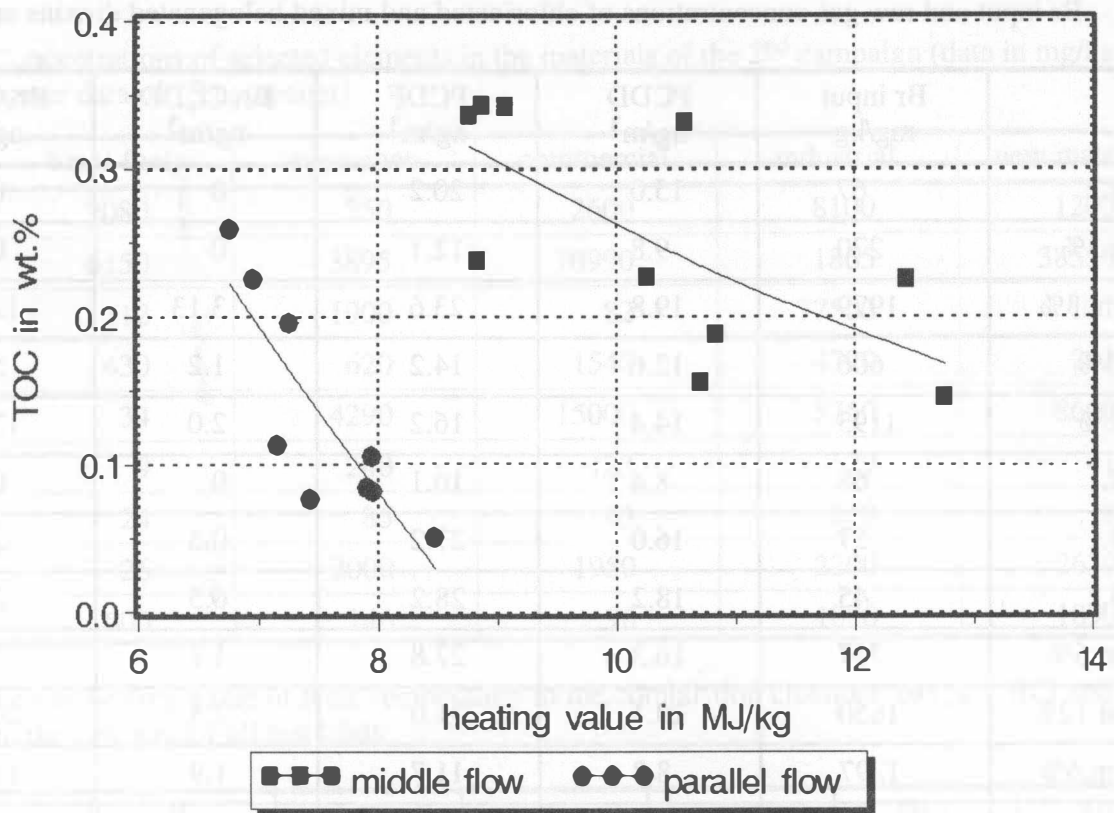


Fig. 1: TOC in the grate ashes vs. heating value of the feed

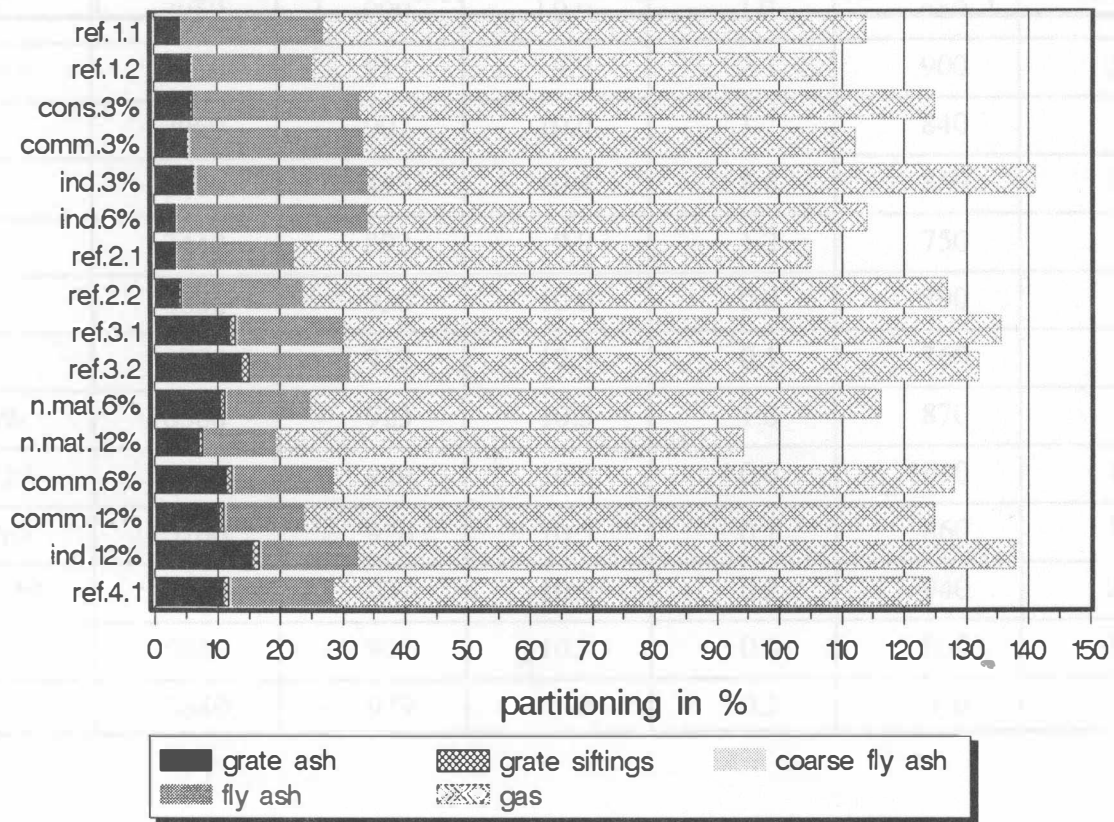


Fig. 2: Percent partitioning of Cl

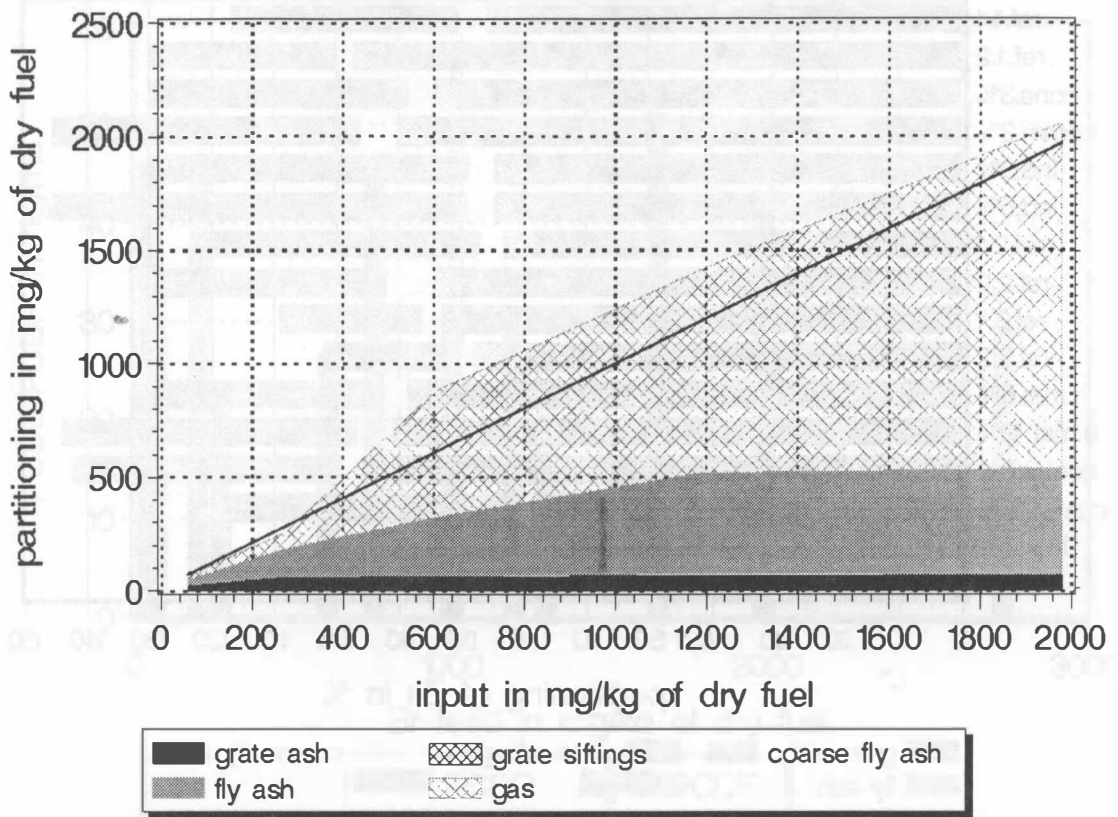


Fig. 3: Partitioning of Br in the parallel flow combustion geometry

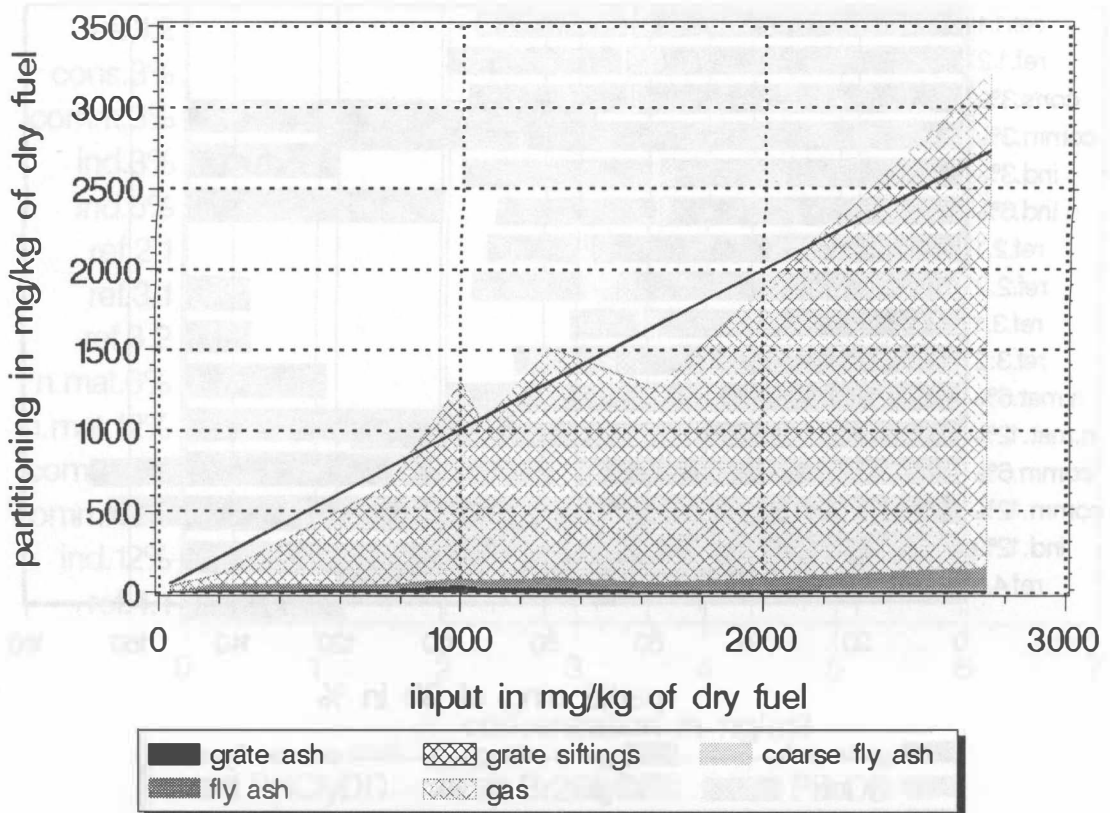


Fig. 4: Partitioning of Br in the middle flow combustion geometry

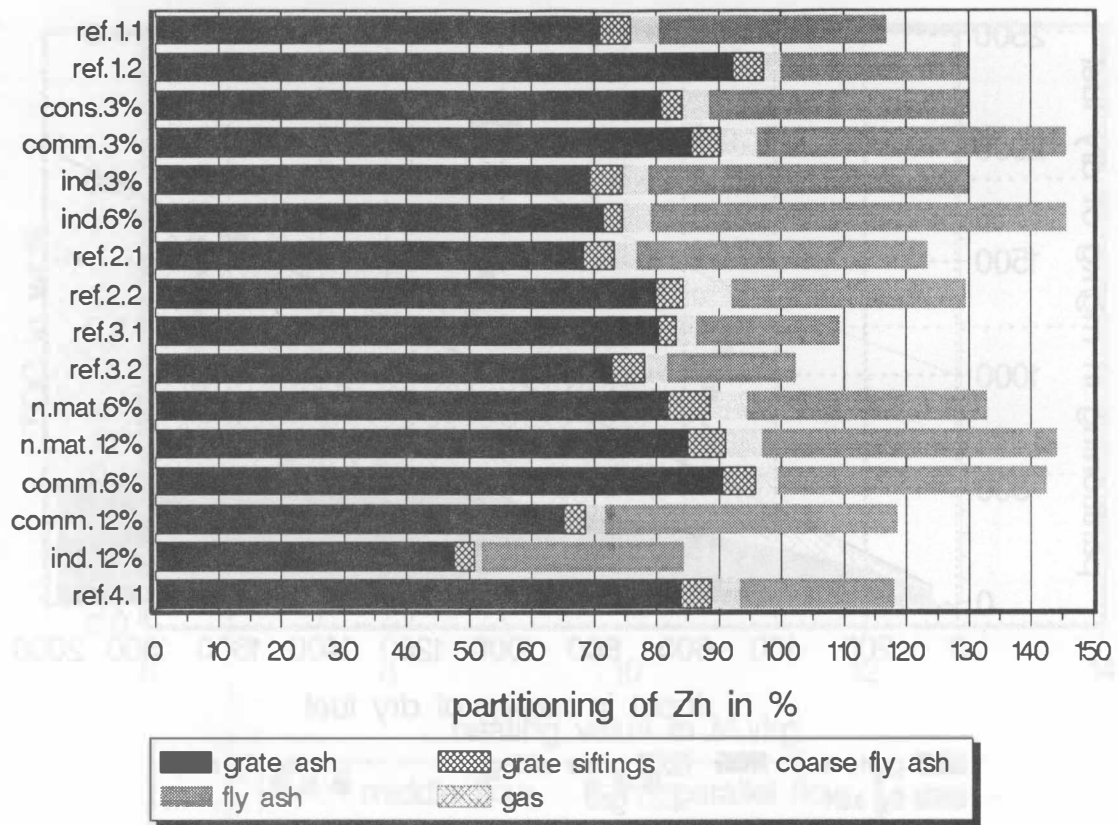


Fig. 5: Percent partitioning of Zn

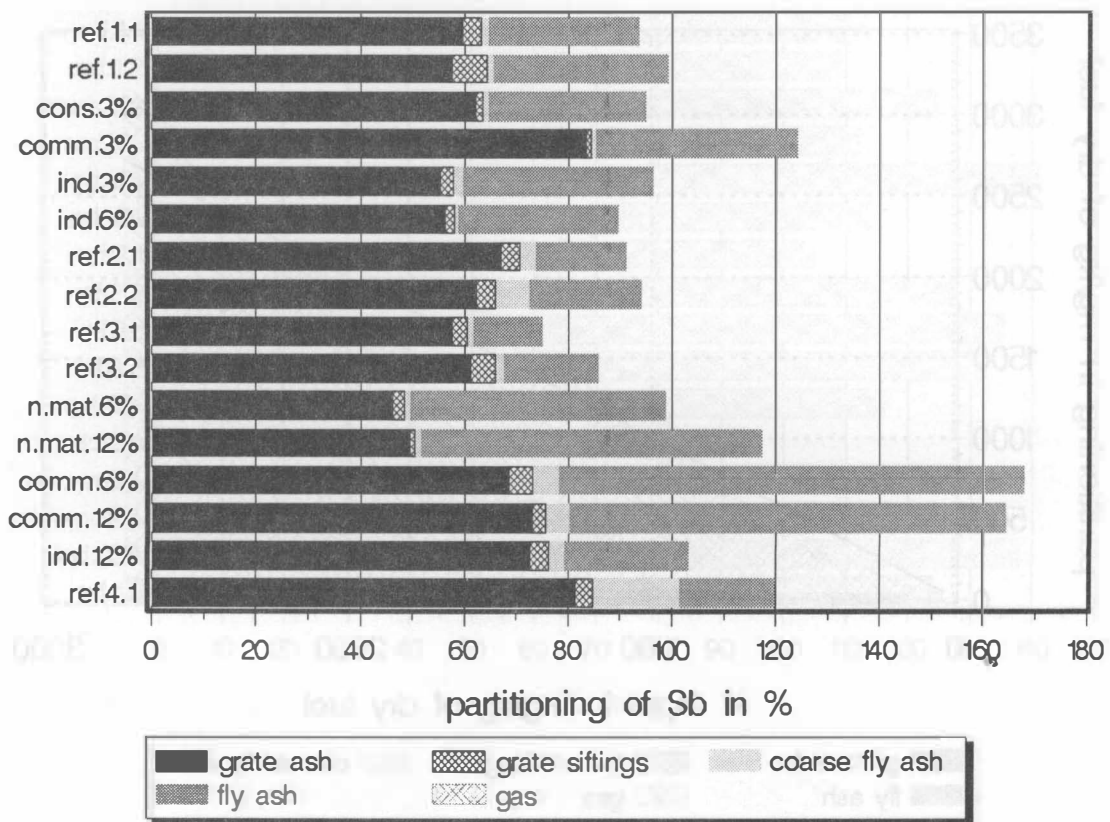


Fig. 6: Percent partitioning of Sb

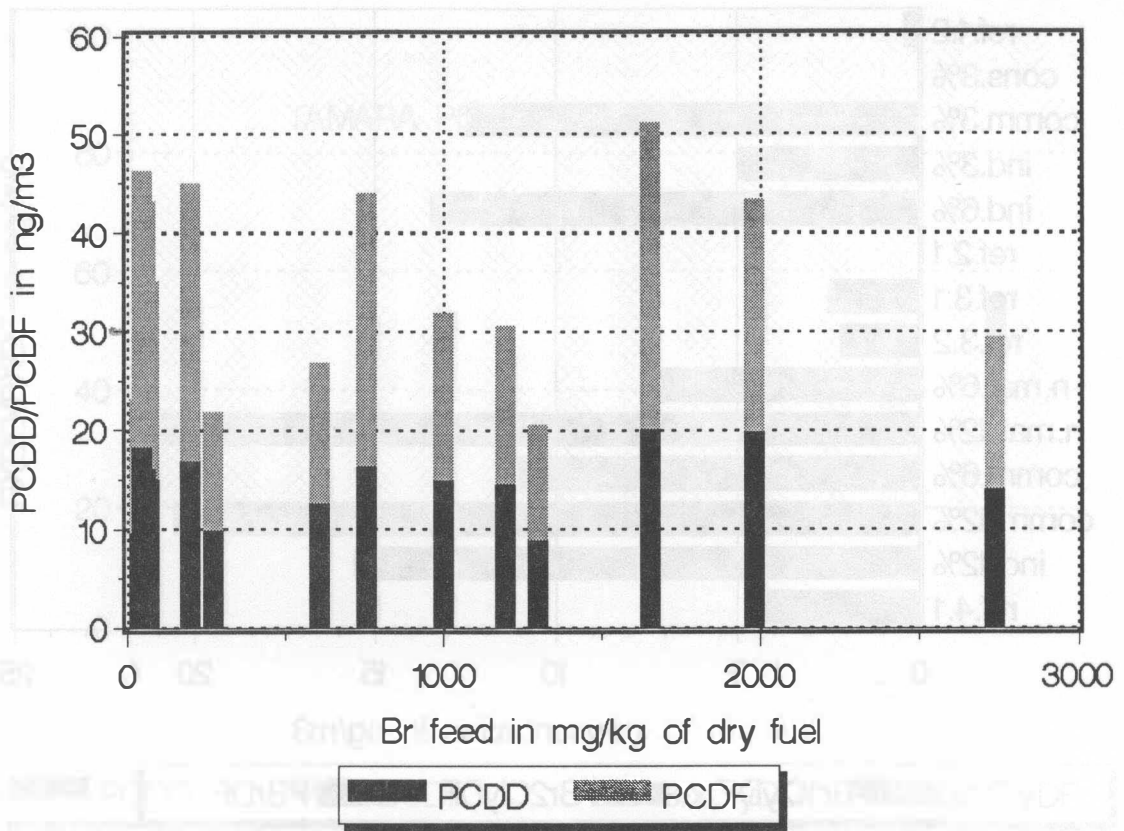


Fig. 7: Sum of PCDD/PCDF levels in the raw gas

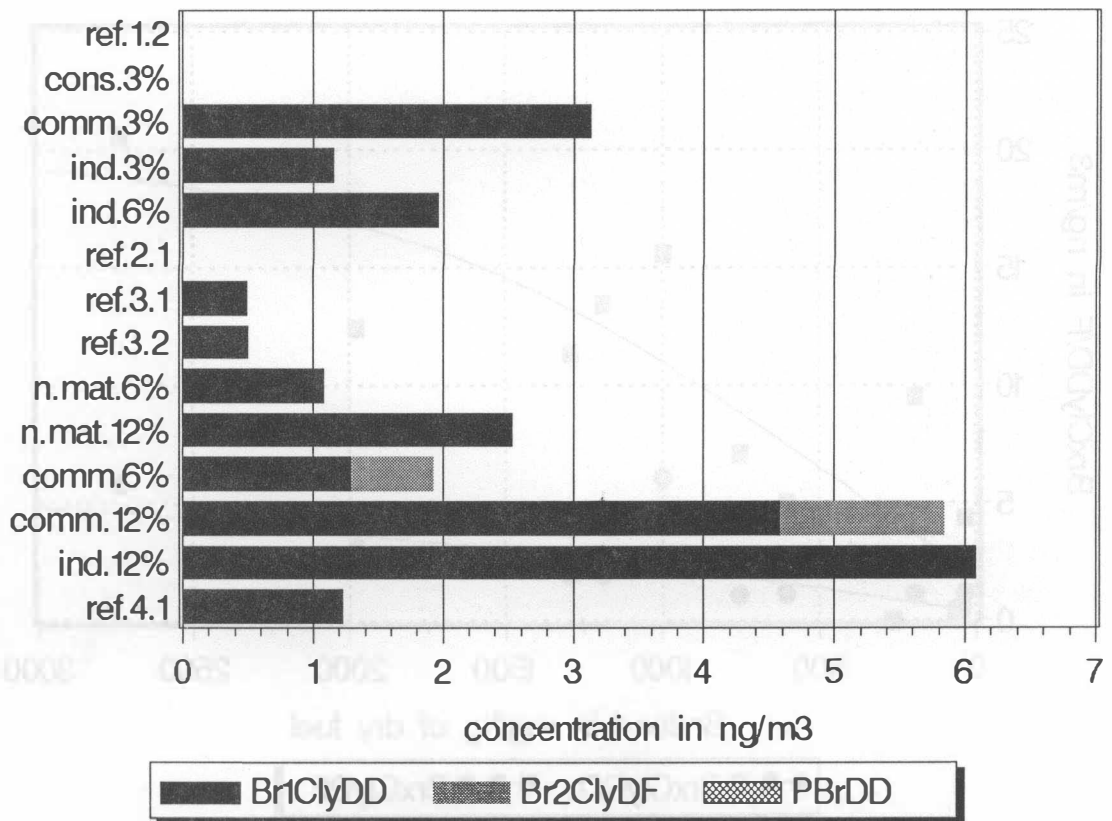


Fig. 8: Br containing dioxins in the raw gas

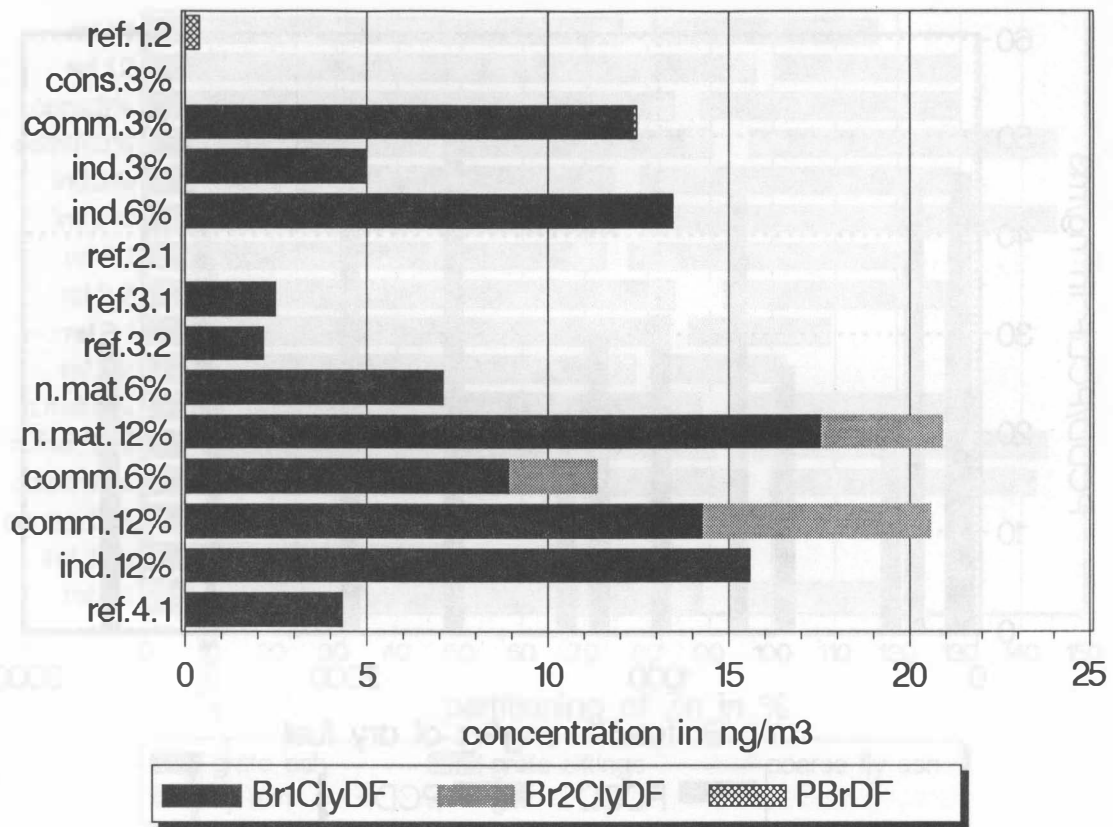


Fig. 9: Br containing furans in the raw gas

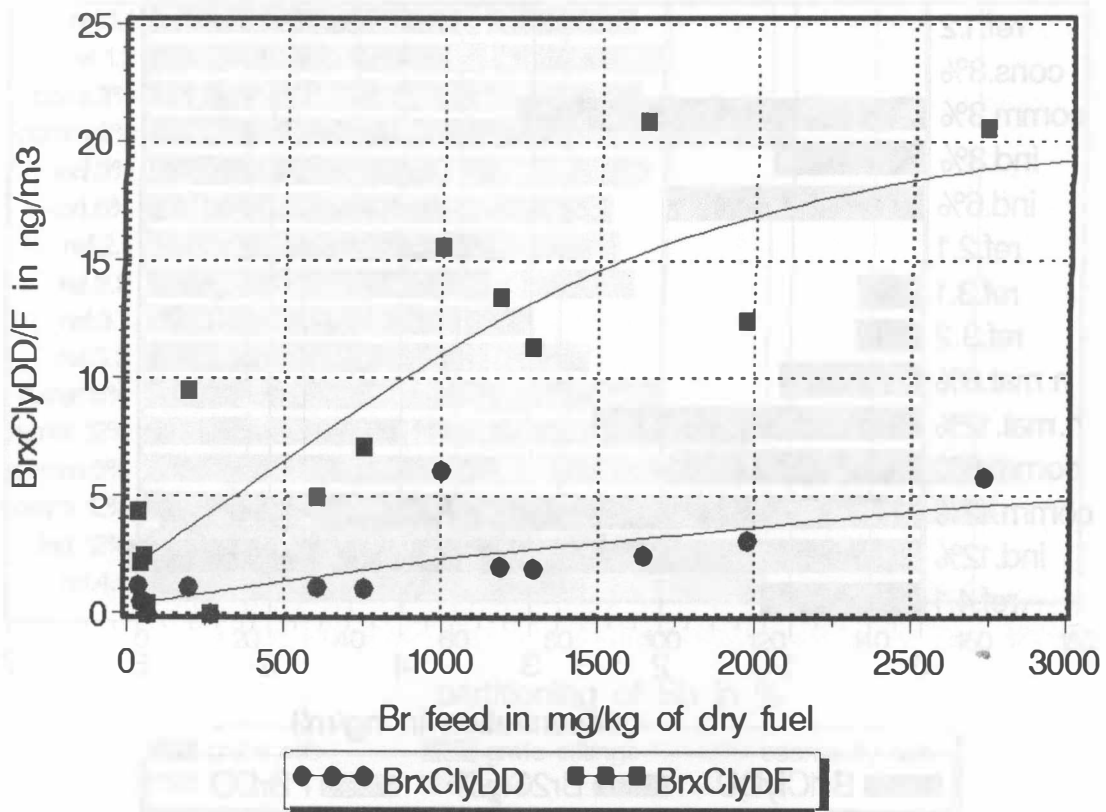


Fig. 10: Bromine containing dioxins and furans versus Br feed

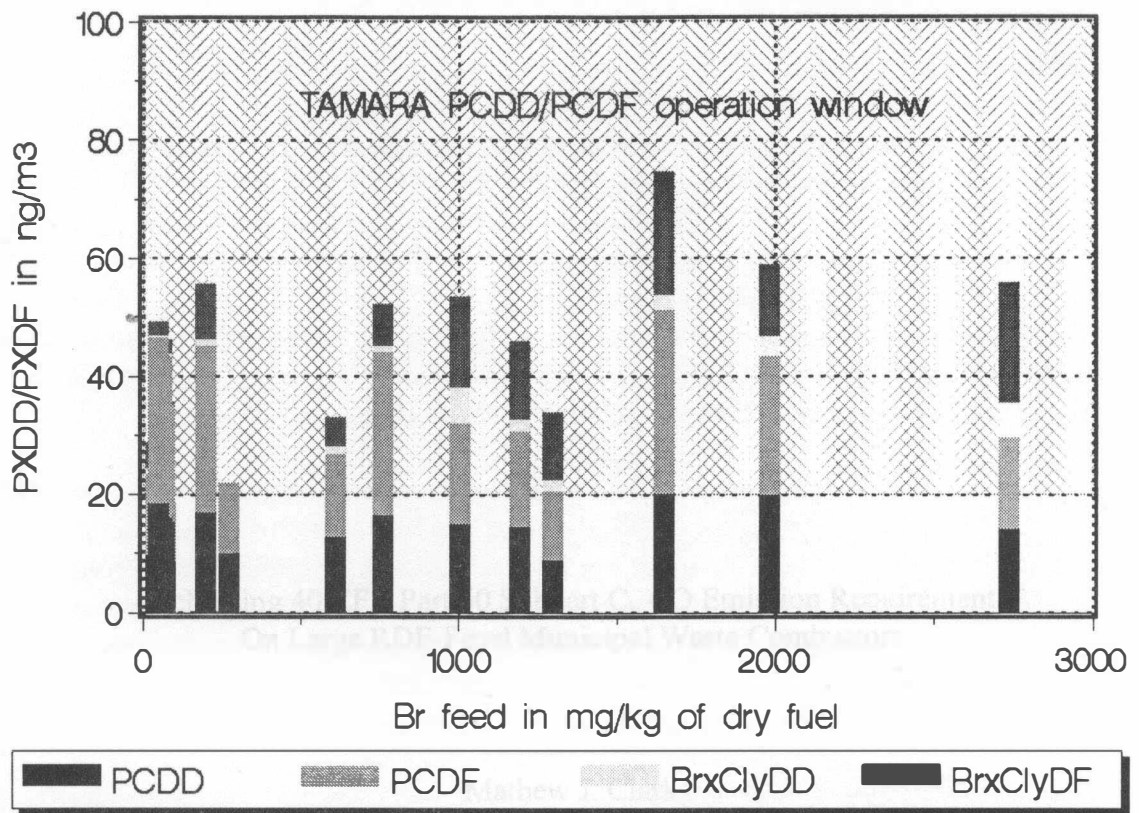


Fig. 11: Sum of halogenated dioxins and furans

