Dioxin Mass Balance for the City of Hamburg, Germany. Part 4: Follow up Study - Trends of PCDD/PCDF Fluxes

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Abstract

The trends of PCDD/PCDF emissions in Hamburg from 1988 to 1995 are evaluated with respect to resulting impacts on man and environment. Emission reductions of about 90 % from the major sources together with product regulations have substantially reduced dioxin fluxes. Effects are measurable in the environment. As nationwide analyses also show decreasing human body burdens, the dioxin problem has lost much in importance.

1. INTRODUCTION

The Dioxin-Balance of Hamburg (FHH 1995) was intended to identify sources of polychlorinated dibenzo-p-dioxins and dibenzofurans relevant for human exposure and environmental effects in Hamburg. Methodology of the balance, results concerning wastes and dietary exposure are reported elsewhere in this volume (LAU et al. 1996 a and b, WESP et al. 1996). This contribution will evaluate results with respect to measures taken in the past and possible future efforts. Beside the investigation of FHH 1995 data the dioxin emission inventory of Hamburg of 1988/90 (FHH 1991) were used to investigate the development of dioxin emissions.

2. WASTE INCINERATION

Incineration of household wastes and hazardous wastes for a long time has been considered a major source of dioxins to the environment and the dominant origin of airborn dioxins. This also was true for Hamburg until a few years ago. Sanitary measures at one municipal solid waste incineration plant and the substitution of the other as well as the reconstruction of the hazardous waste plant reduced emissions by 97.3 %. In Table 1 the reduction of the emissions of the local waste incineration plants is presented.

Table 1: PCDD/PCDF emissions of the local municipal solid waste incineration plants (MSWI) and the hazardous waste incineration (HWI) (g/a I-TEQ)

Year	MSWI I	MSWI B	MSWI II	HWI	Total
1988/90	1.6		4.4	1.0	7
1992	0.95		3.0	0.56	4.5
1995		0.07	0.07	0.05	0.19

Whether waste incineration generates dioxins or in effect is an environmental sink has been a lasting debate not only among engineers and scientists. A balance for the old MSWI I that went out of operation in 1993 and its successor, the MSWI B, working since 1994 was established. The parameters and assumptions of the calculation are given in Table 2.

Table 2: Premises of the dioxin balance for the old (MSWI I) and the new (MSWI B) municipal solid waste incineration plant

	MSWI I	MSWI B
Flue gas (m^3/t)	5000	4000
PCDD/PCDF concentration in flue gas (ng/m^3 I-TEQ)	1.2	0.05
Slag (t/t)	0.3	0.3
PCDD/PCDF concentration in slag (µg/t I-TEQ)	19	10
Filter dust (t/t)	0.05	0.023
PCDD/PCDF concentration in filter dust (µg/t I-TEQ)	1100	1100

Based on the assumption of an annual total waste input of 157,900 t with an average content of PCDD/PCDF of 50 µg/t I-TEQ, into each of the MSWI considered an overall dioxin balance can be calculated. Results are shown in Table 3.

Table 3: Overall dioxin balance for the old and the new municipal solid waste incineration plant (g/a I-TEQ)

Plant	Output flue gas	Output slag	Output filter dust	**Output total**	**Input total**
MSWI I	0.95	0.90	8.7	**10.5**	**7.9**
MSWI B	0.03*	0.47	4.0	**4.5**	**7.9**

*Actual output is higher due to additional waste input (see Table 1).

It can be concluded that the old MSWI I "produced" dioxins while in the new MSWI B only 57 % of the dioxins that entered the plant were existent after incineration (if processes like de novo synthesis are not regarded). However, the dioxin balance of a MSWI must be assessed with respect to the potential effects of the dioxins that remain after the incineration process: Some 90 % are contained in filter dust which will be deposited in controlled hazardous waste disposal sites and thus is withdrawn from the natural environment, another 10 % will be contained in slag/road building materials at low concentration levels, and only 0.4 % enter the environment via emission into the atmosphere. This part was reduced to 3 % of the flux originating from the old MSWI.

An open question so far is the reason of the comparatively high flux of PCDD/PCDF into the incineration, i. e. what are the main reasons for a level of 50 µg/t I-TEQ in municipal solid wastes.

In the Federal Republic of Germany (FRG) estimated PCP/Na-PCP consumptions were 750 t in 1979 and 230 t in 1984 (BUA 1985). Assuming an average PCDD/PCDF content of 2 mg/kg I-TEQ (HAGENMAIER et al. 1987) this would lead to dioxin loads of 1500 g and 460 g in the respective years. Considering that 3 % of the FRG-consumption may have occurred in Hamburg up to 45 g seem a plausible annual input of PCP-born dioxins into Hamburg at the time of intensive use of PCP. This is at least in the same order of magnitude as the annual flux of dioxins calculated for the total of municipal and commercial solid wastes in Hamburg of 49 g I-TEQ for the year 1992 (FHH 1995). The assumption that some ten years after purchase wood products, heavy textiles, etc. show up as wastes does seem reasonable. If PCP actually is a/the major source of dioxins in household wastes it is to be expected that in the foreseeable future the German PCP-ban of 1989 will effect a decline in waste dioxin contents.

3. INDUSTRIAL SOURCES

In Hamburg ferrous and non-ferrous metal production is an important part of the local industry. Especially recycling of copper introduced chlorine and organic material into the metallurgical processes and caused dioxin formation. Since 1992 sanitation and reconstruction of the blast-furnace and the refinery at the copper smelter lead to a reduction of the emissions of estimated 60 %. Figures of industrial emissions are given in Table 4.

Table 4: Emissions of industrial sources (g/a I-TEQ)

Emittent	1988/90	1992	1995
Copper industry	1.44	1.2	0.5
Aluminium industry	0.15	0.008	
Steel production	0.2	0.166	
Petrochemical industry		<0.001	
Total	1.79	1.374	<0.6

4. AUTOMOBILE TRAFFIC

Application of 1,2-dichlororethane as scavenger in leaded gasoline caused PCDD/PCDF-formation during combustion and thus led to considerable dioxin contents in the exhaust gas of gasoline fuelled motor vehicles (SCHWIND et al. 1991). Since the regulation of gasoline lead contents (Gasoline Lead Law of 1971/1994) and the banning of halogenated scavengers (Scavenger Act of 1992), both allowing certain transition periods, dioxins originating from combustion engines are rapidly declining in the FRG. In addition catalysts further reduce emissions of dioxin.

Calculated fluxes for the city of Hamburg in Table 5 show a reduction of more than 90 % for total automobile traffic emissions since 1988.

Table 5: Emissions from automobile traffic (g/a I-TEQ)

	1988/90	1992	> 1995*
Gasoline engines		0.23	0.01
Traffic total	0.53	0.25	0.04

*after equipment of all gasoline fuelled cars with catalysts.

5. DISCUSSION

In Table 6 a synopsis is given for the main emission sources and resulting dioxin emissions of the respective years of reference. The dioxin source next in importance to those listed is supposed to be residential heating by coal fired stoves, estimated to be in the range of 0.07 - 0.34 g/a I-TEQ in 1992. Since modernising of houses and expanding district heating cause a steady decrease of coal fired stoves dioxin emissions from this source will be less, too.

Table 6: Emissions by main sources (g/a I-TEQ)

source	1988/90	1992	1995	1988 - 1995 reduction in %
Waste incineration	7	4.5	0.19	>95
Industry	1.79	1.37	<0.6	>65
Traffic	0.53	0.25	0.04	>90
Total	9.32	6.12	<0.9	>90

To conclude, emissions into the air have been effectively reduced, the estimate is to less than one tenth in 1995 as compared to 1988/90.

Dioxin monitoring in ambient air and deposition by the State Environmental Office of Hamburg demonstrates a decrease of about 70 % and 20 %, respectively (Table7). The fact that the reduction in ambient air is much less than emission reduction may be attributed to long range transport and to smaller diffuse sources not identified yet.

Table 7: PCDD/PCDF concentration in air (fg/m^3) and deposition flux [pg/(m^{2}d)] (I-TEQ)

	1990	1993	1995
Air	108	37	36
Deposition	16	11	13

Fluxes from air to plants, water, soil, composts, etc. via deposition are expected to become smaller steadily. A reduced flux of dioxin from the air to plants, especially fodder plants, lowered PCDD/PCDF-contaminations in milk and meat (WESP et al. 1996). It is believed that emission reduction is the major reason for a reduced input into human food which has led to reduced body burdens in the general population of the FRG. This has been proven by recent analyses of blood fat and human breast milk (PÄPKE et al. 1996, Niedersachsen 1995, FÜRST 1993).

The rapid decrease of human body burden, about 50 % within six years is contradictory to generally accepted theories of human dioxin uptake and/or kinetics of elimination.

If half life times are between three and twenty years for elimination of individual congeners (FLESCH-JANYS et al. 1994) and if uptake is about 100 pg/d I-TEQ, 30 % of which are stored in body fat, a simple calculation can show that typical human body fat concentration of > 50 ng/kg I-TEQ fat at the age of 75 can not be reached. The only plausible explanation for this as well as for the rapidness of decline of measured body burdens is that at the time when blood fat levels increased by about 0.6 ng/(kg·a) I-TEQ (SAGUNSKI et al. 1992) uptake actually was much higher than today, model balances yield results up to 400 pg/d I-TEQ. So the progress reached in dioxin reduction with respect to human nutrition probably is by far higher than generally thought.

6. CONCLUSION

The emission reductions described have reduced dioxin risks to a very large degree. Furthermore remedial measures have minimized dioxin exposures due to contaminated sites. Since also dioxin input into the environment by organochlorine compounds (PCP, PCB, pesticides) has been minimized a continuous further decline of dioxin reaching man or natural environment can be expected.

The "Dioxin Mass Balance for the City of Hamburg" a. o. was to investigate whether in Hamburg further measures on the local level are necessary to reduce dioxin risks. So far reduction has been effective, the major local contribution being emission reduction from waste incineration and industrial sources as well as cleaning up of contaminated sites. Both are continuously going on at high costs. A third MSWI is being planned and remedial action especially at a former pesticide plant cleans up burdens of the past.

A point of high concern in Hamburg is the dioxin load carried by the River Elbe and sedimented in the harbours of Hamburg. Originating mainly from (former) industrial sources upstream of Hamburg some 70 g/a I-TEQ reach the city by suspended particle transport. Approximately 23 g/a of these are contained in sediment dredged from the river and harbours in Hamburg to maintain shipability. They have to be disposed of in Hamburg. Although Hamburg has taken steps to help cleaning measures at the place of origin this problem can only be solved by national and international efforts.

Federal legal measures have been effective, too. Beside the Federal Clean Air Act regulating dioxin in flue gas of MSWI, especially the ban of PCP, PCB, and scavengers, substantially reduced dioxin fluxes. However, PCP may still be a problem in imported goods, so international regulations and control are necessary. Other products also may contain dioxins at unacceptable levels, e. g. resulting from dyes or biocide treatment of textiles. Thorough investigations for possible sources should be a task of Federal institutions.

In general it may be concluded that combined efforts of science and politics have brought great progress towards solution of the dioxin problem but have not completely solved it yet.

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