PCDD/PCDF levels in human blood of people living in a highly PCDD/PCDF contaminated area next to a metal reclamation plant.

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#### Abstract

During the last years increased PCDD/PCDF levels were found in soil, dust and vegetables in an area near to a metal reclamation plant in Southern Germany. In blood samples of 22 volunteers of this area some specific congeners of PCDF were elevated compared to a group not specifically exposed. Comparison of PCDD/PCDF levels between soil, dust, vegetables and human blood will be presented.

### Keywords

Metal reclamation plant, PCDD, PCDF, environmental levels, human blood.

### Introduction

In recent years metallurgical processes were identified as an important source of polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF). In the surrounding of a metal reclamation plant in Baden-Württemberg (West Germany) the contamination of soil, dust from homes, indoor air and vegetables was investigated over a period of 3 years (1987 - 1990). PCDD/PCDF concentrations in soil range between <1 and 100800 ng TE/kg. The PCDD/PCDF concentrations of vegetables were found in the range of not detected (detection limit per isomer 0,1 ng/kg) to 134 ng TE/kg dry weight. These results (Hagenmaier 1988) were demonstrated earlier. Earlier investigations in this industrial zone showed a contamination of soil with heavy metals. In 1986 the plant was closed. People live in the direct neighbourhood to the plant. Vegetables were produced by some of the inhabitants for private consumption.

Little is known about PCDD/PCDF uptake by humans exposed chronically due to a local emission source. Therefore, the concentrations of PCDD/PCDF were investigated in blood samples of 22 volunteers of this area.

## Materials and Methods

In 22 randomized persons (11 males and 11 females) 80 ml of blood were drawn for PCDD/PCDF analysis. To get an idea about the exposure, the participants were interviewed with the aid of a questionnaire. The analysis of PCDD/PCDF in human blood was performed by "ergo laboratories" in Hamburg (Päpke et al.1989). Dust samples were analysed by "Biocontrol Laboratories", Mainz (West Germany). The indoor air analysis was carried out by Gesellschaft für Arbeitsplatz und Umweltanalytik (GFA), Münster-Roxel (West Germany) and by the Institute of Organic Chemistry, University of Tübingen.

Toxic equivalent (TE) factors proposed by the German Federal Health Office (Appel et al. 1985) were used.

# Results and Discussion

Figure 1 shows the PCDD/PCDF concentration in soil of the area concerned, expressed in ng TE/kg soil. Highest soil concentrations were found next to the plant. The maximum concentration up to 100800 ppt TE was found next to a plant air vent.

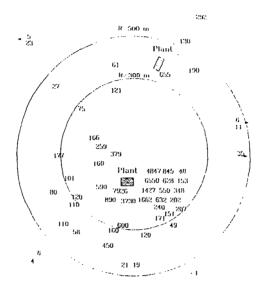


Fig. 1: Location of the reclamation plant and soil concentrations in the surrounding area (in ng TE/kg).

Table 1 describes the PCDD/PCDF contamination of some houses in the area. Corresponding soil concentrations are shown in the table. The indoor air PCDD/PCDF

Tab. 1: PCDD/PCDF levels of dust, indoor-air, interior walls and corresponding soil concentrations.

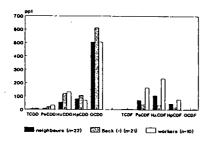
dust (room)	dust (loft)	interior	indoor mir	soil(0-7 cm)	ഒലു1(0-20 ലഅ)
ng TE /kg	ng TE /kg	ng TE /ky	py TE/cbm loft room	ng 1	tE /kg
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1034,00		100,00			· · · · · · · · · · · · · · · · · · ·
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437,00	52168,00				629,00
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]	67535,00		0,10 0,18	671,00	12628,00

concentration was 0,18 - 0,3 pg TE/cbm. Extremely high contamination of dust was observed. Up to 578444 ppt TE could be detected in dust particles on one loft. Table 2 shows the PCDD/PCDF levels in (blood-) fat of the 22 persons. (Blood-) fat concentrations of 10 workers\* of the reclamation plant are shown for comparison in this table. The 22 persons, 10 - 70 years old, had been living in the contaminated area at least for a 10 years period, one person even up to 58 years. Eleven persons declared consumption or earlier consumption of vegetables from the kitchen garden in this area. Three persons worked in the metal plant, for a period of more than 10 years.

Tab. 2: PCDD/PCDF levels in blood from neighbours and plant workers (means and ranges) in ng/kg fat.

Congeners		Neighbours	eighbours (n=22)		Plant Workers (n=10)		
	Mean	Range		Mean	Range		
2.3.7.8 TCDD	3,4	1,3	6,2	9,2	2,6	33,0	
PeCDD	12,4	2,9	21,0	34,4	12,0	97,0	
ΣHxCDD	59,2	29,7	118,0	133.0	61,0	308.0	
HpCDD	83,5	23,0	238,0	70.8	26,0	145.0	
OCDD	505,9	176,0	2126.0	506.4	139.0	1098.0	
ΣPCDD	664,1	249,7	2506,9	753,8	277,0	1249,0	
2.3.7.8 TCDF	4,9	1,4	9,8	2,7	1,9	4,3	
ΣPeCDF	72,9	28,0	228,0	166,0	39,7	385,6	
ΣHxCDF	108,6	43,7	410,5	236,6	48,0	605,7	
ZHpCDF	48,3	16,0	190,4	77,8	28.6	211,3	
OCDF	4,3	1,2	8,8	7.2	2,8	16,0	
ΣPCDF	238,9	116,2	746,1	490,3	125,1	1205,0	
ΣPCDD/F	903,0	369,1	2772,1	1244,1	665,2	2454,0	
EPCDD/F(TE)	31,0	16,1	80.4	68,5	21,5	175,0	

When comparing the results with the fat concentrations of a group not specifically exposed (Beck et al.1989) and with the (blood-) fat concentrations of the plant workers a typical pattern of penta-, hexa- and hepta-furans was found to be characteristic. This becomes even more obvious when mean values are compared (Fig. 2).



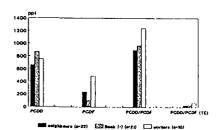


Fig. 2: Comparison of PCDD/PCDF levels (means) of neighbours, plant workers and a not specifically exposed group (in ng/kg fat).

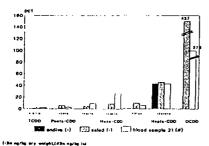
\*This investigation was carried out by order of the Süddeutsche Edel- und Unedelmetall-Berufsgenossenschaft.

The results of TE calculations are shown in Table 2. The maximum of 80 ppt TE belongs to one reclamation plant worker. In 5 persons (sample no. 2, 6, 13, 15, 22) pentar, hexa- and/or hepta-CDF levels were above the 'background range'. In one of these persons (sample no. 13) additionally the octa-CDD level was above the 'background range' (Tab. 3). Two of these persons (no. 2 and 6) worked in the plant. One person (no. 13) lived on a diet. Two of them (no. 6 and 13) declared regular or occasional vegetable consumption.

Tab. 3: (Blood-) fat concentrations of participants with elevated penta-, hexa-, hepta-CDF and/or octa-CDD levels (in ng/kg fat).

Congeners Sample No	2 .	6	13	15	22
2.3.7.8 TCDD	5,7	6,2	3,9	3,9	3,4
PeCDD	20,0	21,0	21,0	19.0	8,9
ΣHxCDD	66,0	88,0	118,0	80,0	38,0
HpCDD	67,0	127,0	238,0	92,0	37,0
OCDD	241,0	393,0	2126,0	621,0	185,0
EPCDD	399,7	635,2	2506,9	815,9	272,3
2.3.7.8 TCDF	2,6	6,4	1,4	9,0	4,6
ΣPeCDF	228,0	143,8	103,5	186,0	135.6
EHXCDF	410,5	255,6	91,0	404,4	243,9
ΣHpCDF	102,5	102,6	62,4	89,1	190.4
OCDF	2,5	4,2	6,9	8,8	3,6
ΣPCDF	746,1	512,6	265,2	697,3	578,1
EPCDD/F	1145,8	1147,8	2772,1	1513,2	850,4
ΣPCDD/F(TE)	80,4	60,4	42,5	76,2	49,0

With regard to the food chain some vegetables from the kitchen garden of one investigated person and the blood of this person were compared (Fig. 3). There are slight indications of a correlation.



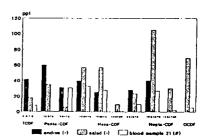


Fig. 3: Comparison of 2. 3. 7. 8-substituted PCDD/PCDF in corresponding vegetables and a human blood sample.

This correlation becomes evident by comparing the persons who declared consumption of vegetables from the kitchen garden with those who did not (Tab. 4). An increase in penta- and hexa-GDF is obvious. Persons who worked in the plant, persons with only occasional consumption and one child, which is described later, were not considered.

Tab. 4: PCDF levels in blood of participants with and without vegetable consumption from a kitchen garden (means and ranges) in ng/kg fat.

BLOOD SAMPLE NO.	VEGETABLE CONSUMPTION FROM A KITCHEN GARDEN ( n=						
	TCDF (2.3.7.8-)	P5CDF	H6CDF	H7CDF	OCDF	EPCDF	
4	4,3	45,0	50,8	34,0	4.5	138,6	
8	4,2	117,1	105,0	34,0	5,0	265,3	
14	9,8	65,0	67,0	27,2	4,1	173,1	
15	9,0	186,0	404,4	89,1	8,8	697.3	
16	2,9	42,0	51,8	19,6	3,8	120,1	
18	2,6	71,5	83,0	16,0	2,0	175,1	
19	5,3	72,2	43,7	21,2	4,2	146.6	
21	8,5	31,0	70,8	28,0	5,6	143,9	
MEAN	5,8	78,7	109,6	33,6	4,8	232,5	
MIN	2,6	31,0	43,7	16,0	2,0	120,1	
MAX	9,8	186,0	404.4	89,1	8.8	697.3	

NO VEGETABLE CONSUMPTION FROM A KITCHEN GARDEN (n=8)

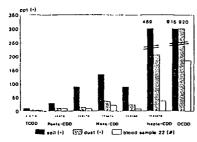
BLOOD SAMPLE No.	TCDF (2.3.7.8-)	P5CDF	H6CDF	H7CDF	OCDF	EPCDP
1	8,9	29,8	49,4	27,0	6,9	122,0
5	4,3	32,0	63,5	43,0	4,7	147,5
7	6,7	28,0	44,6	36,0	4,1	119,4
9	5,4	31,0	46,0	29,0	4,8	116,2
10	3,3	38,3	54,0	35,0	5,0	133,1
11	3,5	34,7	55,4	33,6	2,4	129,6
12	4,5	28,0	47,0	36,0	3,9	119,4
_17	3,4	28,8	55,8	39,4	2,4	129,8
HEAN	5,0	31,3	52,0	34,9	4.3	127.1
MIN	3,3	28,0	44,6	27.0	2.4	116,2
MAX	8.9	38,3	63,5	43,0	6,9	147,5

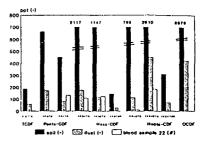
The results of the (blood-) fat analysis of the children are shown in Table 5.

Tab. 5: PCDD/PCDF levels in blood of children from the investigated group (in ng/kg fat).

Congeners Sample No.	22	21	12	11
2.3.7.8 TCDD	3,4	2,5	2,0	2,5
PeCDD	8,9	2.9	11,0	14,0
ΣHxCDD	38,0	43,4	80.9	45.4
HpCDD	37,0	43,0	138,0	58,0
OCDD	185,0	276,0	229,0	257.0
ΣPCDD	272,3	367,8	460,9	376,9
2.3.7.8 TCDF	4,6	8,5	4,5	3,5
ΣPeCDF	135,6	31,0	28,0	34,7
ΣHxCDF	243,9	70,8	47,0	55,4
ΣHpCDF	190,4	28,0	36,0	33,6
OCDF	3,6	5,6	3,9	2,4
ΣPCDF	578,1	143,9	119,4	129,6
ΣPCDD/F	850,4	511,7	580,3	506,5
ΣPCDD/F(TE)	49.0	19,2	21,1	19,0

The children are in the age of 10 - 14 years. Only one child (sample no. 21) declared consumption from the kitchen garden. One child (sample no. 22) lived opposite to an air vent belonging to the plant. The comparison of PCDD/PCDF levels from soil, dust and the blood sample from this child is shown in Figure 4. Taking the results of the other 3 children into account there is an increase in penta—, hexa— and hepta-CDF levels. This may be due to dust and/or soil ingestion by the child.





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Fig. 4: Comparison of 2. 3. 7. 8-substituted PCDD/PCDF levels in corresponding soil, dust and human blood.

#### Conclusions

- Regarding the total amount of PCDD/PCDF (blood-) fat levels one person is out of the range of a not specifically exposed group. Taking the toxicity equivalents into consideration two persons are out of this range.
- Specific isomers (penta-, hexa- and hepta-CDF) are increased in the blood samples. This corresponds to the pattern found throughout the contaminated area.
- Increased PCDD/PCDF levels in blood can be traced to occupational exposure and/or food intake.
- For children soil and/or dust ingestion may be a pathway of special importance.
- The rather high local contamination did not lead to a general increase in PCDD/PCDF levels in blood.

# References

Appel K., H. Beck, A.G. Hildebrandt and W. Lingk (1985) Ansatz einer gesundheitlichen Bewertung von Dioxinen und Furanen.
In: Sachstand Dioxine - Stand November 1984 (Umweltbundesamt ed.), Berichte 5/85 pp 237 - 266. Erich Schmidt Verlag, Berlin.

Beck H., Eckart K., Mathar W., Wittkowski I. (1989) Levels of PCDD and PCDF in Adipose Tissue of Occupationally Exposed Workers. Chemosphere, 18 pp 507 ~ 516.

Hagenmaier, H. (1988) 8-th International Dioxin Symposium, Umea

Päpke O., Ball M., Lis Z.A., Scheunert K. (1989) PCDD/PCDF in Whole Blood Samples of Unexposed Persons. Chemosphere 19, pp 1-6 and 941-948.