Decrease of PCDD/F levels in human blood from Germany (1991 - 1996)

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Introduction

Routine analyses of PCDD/F in human blood with standardized methods started in Germany at the end of the Eighties and are practicised up to now only in a few laboratories. We assume, that over the last decade much more than 2000 German human blood samples have been analyzed for PCDD/F by scientific institutions and in particular by commercial laboratories, but only a small part of these data have been published.

In 1992 Fürst was the first who observed a decreasing trend of PCDD/F levels in human milk samples from Germany [1, 2] and similar observations were made by Alder et al. in 1994 [3]. The same trend was also observed for German blood samples [4, 5].

Material and Methods

In the period 1991 - 1996 we analyzed several hundred blood samples for their PCDD/F content. The blood measurements were carried out in part in the context of scientific studies, in part due to the fact that the subjects were suspected to have been exposed to assumed PCDD/F sources in connection with fire accidents, exposure with contaminated materials like dust, soil or degradation products of combustion processes.
The analytical method used by us has been described elsewhere [6] and will not be repeated here. The laboratory participated successfully at several national and international interlaboratory quality control programs.
In the present study only blood samples of subjects with known age (total range 9 - 82 years) were included for which the anamnestic data indicated, that no or only an unimportant (in most cases a brief) exposure towards supposed PCDD/F sources had taken place. The entire dataset evaluated in this study includes 507 blood samples and can be used for an estimation of the PCDD/F levels in general population human blood in Germany over the last 6 years.

Results and discussion

The dataset was divided into six subgroups, representing one-year sampling periods from 1991 to 1996. Basic statistical parameters with regard to age, fat content of the examined blood samples and the PCDD/F levels on a lipid basis are summarized in table 1 for the data subsets 1991 to 1996. Table 1 shows in addition the percentage of the difference of the mean and median values for the 1996 dataset in comparison to the 1991 dataset.

Although the groups examined in the different subsets from 1991 to 1996 do not have the same size and age structure, it is obvious, that the PCDD/F-levels have continously decreased over this time period. The PCDD/F-levels found in 1996 are approximately half the concentrations measured in 1991. This tendency can be found for all detectable congeners with 2,3,7,8-chlorosubstitution pattern, for the sum values and for the toxicity equivalents as well. The observed differences are statistically significant at $\alpha = 0.01$ for means and medians. Considerable congener specific differences can not be observed.

As shown in our pilot study in 1991 [6, 7] the blood levels of most PCDD/F congeners and the toxicity equivalents increase with age. In order to fit the data subsets and for statistical evaluation a multiplicative model was used: $CONCENTRATION\ IN\ BLOOD = A \cdot AGE^{B}$. Figure 1 shows the individual I-TEq values on a lipid basis in relation to age, the calculated regression lines and the 95%-prediction limits for all one-year subsets in individual plots.

The findings of the present study are more or less consistent with the results of our previous study: The correlation with age is significant for most of the 2,3,7,8-chlorosubstituted PCDD/F except for some of the higher chlorinated PCDF, both on lipid and on whole-weight basis. However, the correlation is stronger and more significant for the earlier datasets. Congeners increasing with age show an appreciable smaller range of variation for younger people when compared with older subjects in all datasets. The relative increases with age are not equal for all congeners, they show a decrease with the chlorination grade and, when comparing the one-year subsets among themselves, they decrease with the time of examination. The most pronounced age-related increase can be observed for the lower chlorinated PCDD and for 2,3,4,7,8-PCDF.

Although the specimens, except of the 1991 subgroup, were not selected in a systematic way to address representativity and the several one-year datasets do not consist of identical persons studied repeatedly, our data indicate, that a continous reduction of PCDD/F blood levels in the German general population has taken place over the last six years.

Conclusion

The data presented can be used for estimation of the German background exposure to PCDD/F during the last 6 years. The strong correlation with age and the rapid decline over time of the human blood levels indicate that only age-related reference values examined in the same period of time should be used to ascertain the internal exposure status with PCDD/F of individuals or groups.

References

[1] Fürst, P., Fürst, C., and Wilmers, K.; *Chemosphere* ***25***, 1029-1038 (1992)

[2] Fürst, P.; *Organohalogen Compounds* ***13***, 1-8 (1993)

[3] Alder, L., Beck, H., Mathar, W., and Palavinskas, R; *Organohalogen Compounds* ***21***, 39-44 (1994)

[4] Schecter, A., Päpke, O., and Fürst, P.; *Organohalogen Compounds* ***30***, 57-60 (1996)

[5] Schecter, A., Päpke, O., Fürst, P., and Ryan, J. J.; *Organohalogen Compounds* ***33***, 473-478 (1997)

[6] Schrey, P., Wittsiepe, J., Ewers, U., Exner, M., and Selenka, F.; *Organohalogen Compounds* ***9***, 261-267 (1992)

[7] Schrey, P., Wittsiepe, J., Ewers, U., Exner, M., and Selenka, F.; *Bundesgesundhbl.* ***36***, 455-464 (1993)

Table 1: Basic statistical data on PCDD/F levels [pg/g (lipid basis)] in German general population human blood (1991 - 1996)

	1991 (n = 95)		1992 (n = 157)		1993 (n = 17)		1994 (n = 74)		1995 (n = 69)		1996 (n = 95)		Change 1991 - 1996 based on	
	Mean	Median	Mean	Median	Mean	Median	Mean	Median	Mean	Median	Mean	Median	means	med.
Age [years]	44.7	47	42.4	43	40.5	42	46.5	49	45.2	42	37.7	35		
Fat content [mg/g]	5.701	5.67	5.72	5.53	6.02	6.04	5.95	5.82	5.74	5.44	5.14	5.01		
	Concentration in human blood [pg/g (lipid basis)]												[Δ%] *)	
2378-TCDD	4.62	4.4	4.14	3.8	3.18	2.8	5.13	4.9	2.81	2.6	2.34	2.2	-49	-50
12378-PCDD	18.0	17	16.1	15	12.8	11	12.6	10.5	9.93	9.5	7.85	7.5	-56	-56
123478-HxCDD	16.3	15	15.3	15	12.9	12	11.0	10	9.48	9.0	7.38	6.6	-55	-56
123678-HxCDD	45.9	46	42.9	42	34.4	33	19.4	16	24.6	23	24.4	23	-47	-50
123789-HxCDD	9.26	8.6	7.84	7.3	5.77	4.6	4.34	3.9	4.87	4.5	4.29	3.9	-54	-55
1234678-HpCDD	87.2	86	74.8	69	43.3	32	30.7	27	41.4	36	31.2	26	-64	-70
OCDD	446	420	462	400	340	300	231	200	293	250	257	220	-42	-48
2378-TCDF	1.37	1.1	1.16	0.78	1.59	1.0	1.41	1.2	1.42	1.2	--	--	--	--
12378-PeCDF	0.644	0.63	0.885	0.60	1.74	1.3	0.767	0.60	1.42	0.99	0.63	0.56	(-2)	(-10)
23478-PeCDF	34.3	32	30.4	26	21.2	18	21.9	20	18.5	15	17.1	16	-50	-50
123478-HxCDF	11.5	11	9.79	8.8	8.52	7.5	11.3	7.7	9.90	6.0	5.73	5.2	-50	-53
123678-HxCDF	16.5	16	13.9	13	12.4	11	11.9	10	9.70	7.8	6.53	6.2	-60	-61
234678-HxCDF	3.67	3.3	3.22	3.1	3.60	3.4	1.97	1.6	2.22	2.1	2.15	2.0	-41	-39
123789-HxCDF	0.44	0.44	0.956	0.51	--	--	--	--	--	--	--	--	--	--
1234678-HpCDF	15.35	14	14.9	12	18.6	12	7.39	6.2	7.71	7.15	6.55	5.8	-57	-59
1234789-HpCDF	0.48	0.43	0.651	0.56	1.87	1.1	0.644	0.55	0.702	0.62	0.55	0.50	(+15)	(+18)
OCDF	1.10	0.96	2.98	1.9	22.08	3.1	0.964	0.85	1.91	1.4	1.41	1.1	(+28)	(+15)
Sum P(4-8)CDD	633.5	621	627.1	551	454.5	368	317.7	281	382.1	340	334.4	287	-47	-54
Sum P(4-8)CDF	84.68	81.2	75.96	69.5	79.74	56.9	58.99	50.1	49.39	40.3	38.70	37.3	-54	-54
Sum P(4-8)CDD/F	718.4	706	703.2	625	534.5	446	376.7	340	431.6	379	373.1	321	-48	-55
BGA/UBA-TEq	21.81	21.65	19.61	18.94	15.59	13.04	15.39	14.89	12.74	12.14	10.97	10.23	-50	-53
NATO/CCMS-TEq	42.67	40.77	38.12	36.5	29.05	25.29	29.13	28.66	24.06	22.83	20.74	19.24	-51	-53

*) Values in parenthesis were not statistical significant at $\alpha = 0.01$. These congeners were only detectable in concentrations near to the detection limit.

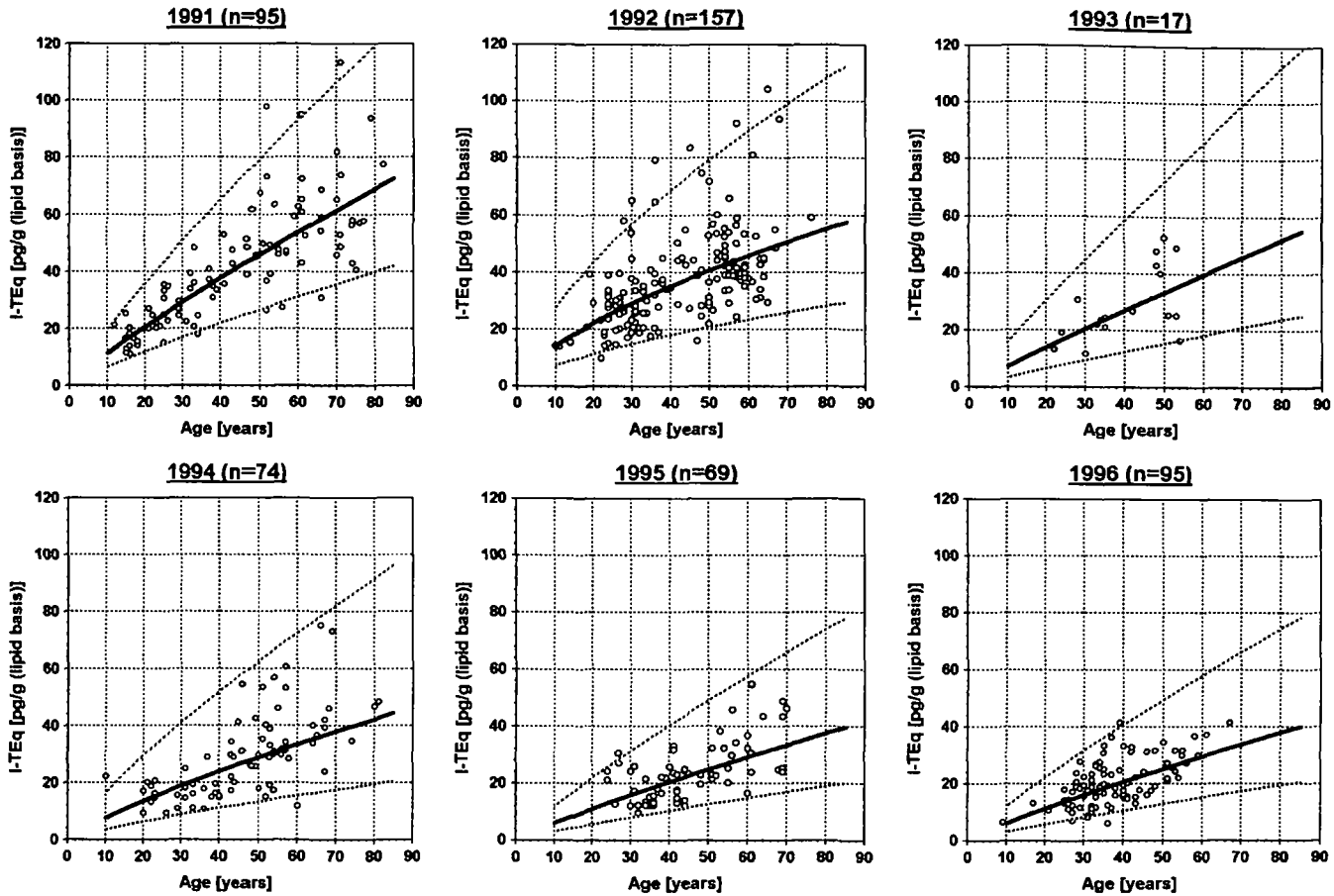

Figure 1: PCDD/F levels in German general population human blood (I-TEq [pg/g (lipid basis)]) in relation to age (1991 - 1996)

Parameters of regression using a multiplicative model $CONCENTRATION\ IN\ BLOOD = A \cdot AGE^{B}$: — = regression curve, --- = 95%-prediction limits.