

Serum Levels of Polychlorinated Dibenzo-*p*-dioxins and Polychlorinated Dibenzofurans in Pulp and Paper Mill Workers

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1. Introduction

In this report, we describe the results of a study in which serum levels of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in long-term pulp and paper mill workers were compared to levels in community residents who had never worked at the mill.

2. Methods

We classified jobs according to the potential for exposure to bleached pulp or paper dust and process water or effluent mists. Areas of the mill considered having a high exposure potential were the bleach plant, dry end of the paper machines, rewind areas, finishing areas, and effluent treatment plant. Areas of the mill considered having a low exposure potential were the groundwood mill/long log area, wood yard, and Kraft mill. Two worker groups were defined, those with 10 or more years in high exposure potential areas and those with 10 or more years in low exposure potential areas and less than one year in high exposure potential areas. Each participating worker was asked to identify a friend who lived in the community but never worked in the mill.

Study participants provided a 250-milliliter, nonfasting blood sample. Serum was analyzed for PCDDs and PCDFs using high-resolution gas chromatography/high-resolution mass spectrometry.^{1,2)} All measurements were corrected for total serum lipid.³⁾ When serum results were reported as below the limit of detection, we calculated an imputed value.⁴⁾ To assess whether the imputation method affected our findings, analyses were repeated by replacing nondetectable values with zero. We examined differences between exposure groups in individual PCDD and PCDF congeners and in the toxic equivalents (I-TEQ) based on the international toxic equivalence factors.⁵⁾

3. Results

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Based on seniority lists obtained from the mill, 76 workers were identified as possible study subjects. We were successful in contacting 68 of these workers by phone; 10 refused to participate in the study. Forty six of the 58 workers with the longest seniority were selected for the study. Thirty-two workers worked for 10 or more years in high exposure potential areas. Seven of these no longer worked in these areas at the time of the study. Fourteen workers worked 10 or more years in low exposure potential areas. Seven of these workers also had been in high exposure potential areas for between one and eight years. [These seven workers were considered to have mixed exposure potential.] Twenty-three community residents identified by participating workers were contacted. Twelve agreed and seven refused to participate, and four were excluded because of prior work at the mill. Workers and the union identified four replacements; all participated.

Overall, there were no appreciable differences among the three exposure groups in the median values for specific PCDDs or PCDFs (Tables 1 and 2). The median value in high-exposure-potential workers was greater than in community residents for three of the eight PCDDs –2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, and 1,2,3,7,8,9-HxCDD– and four of the nine PCDFs –1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-PeCDF, 2,3,4,6,7,8-HxCDF, and 1,2,3,4,6,7,8-HpCDF. The relative differences in the medians for 2,3,7,8-TCDD (6%) and 1,2,3,7,8-PeCDD (2%) were very small and probably not meaningful ($p=0.65$ and 0.44 , respectively). There was a 20% difference in the median for 1,2,3,7,8,9-HxCDD ($p=0.36$), but workers with low exposure potential had a higher median than workers with high exposure potential. The relative differences in the median for 1,2,3,4,7,8-HxCDF

(6%) and 1,2,3,6,7,8-PeCDF (2%) were also very small and probably not meaningful ($p=0.55$ and 0.50 , respectively). There was an 18% difference in the median for 2,3,4,6,7,8-HxCDF ($p=0.77$) and a 12% difference in the median for 1,2,3,4,6,7,8-HpCDF ($p=0.88$), but workers with low exposure potential had a similar or higher median than workers with high exposure potential.

For all PCDDs and PCDFs except 2,3,7,8-TCDD and 1,2,3,7,8,9-HxCDD, high exposure-potential-workers no longer in these jobs had slightly higher median levels than those whose presumed exposure was current. PCDD and PCDF levels in workers with mixed exposure did not consistently follow a pattern suggestive of occupational exposure (i.e., intermediate between those with high and those with low exposure potential).

The highest values for specific PCDDs and PCDFs consistently occurred in seven workers. These individuals ranged in age from 37 to 64 and had worked in the mill for 10 to over 40 years in a variety of jobs. One had a job involving exposure to bleached paper dust in the finishing department for four years and to effluents for five years. Five had jobs involving exposure to bleached paper dust at the paper machines for six to 44 years. One never worked in an area with high exposure potential. Six of the seven had potential exposure outside work that could have contributed to these findings. Six ate locally caught fish, four had lived in a home heated with wood, two had applied weed killer at home, and one had used pentachlorophenol to treat wood.

Both the median and maximum values of the total I-TEQ were higher in workers in the low exposure potential group than in workers in the high potential exposure group or community residents (Table 3). The lowest value for the median I-TEQ occurred in workers in the high exposure potential group. The relative differences between the highest and the other median values, however, were small (11% to 26%, $0.32 \leq p \leq 0.96$). Two workers had considerably higher values than other workers or community residents. Both worked in the wood yard for at least 15 years. When these workers are excluded from the I-TEQ comparisons, the median and maximum values are decreased slightly for low-exposure-potential workers, but the pattern of the results was unchanged.

4. Conclusion

Current body burdens of PCDDs or PCDFs were within the background ranges previously reported for persons with no known occupational exposure. Differences between mill workers and community residents in PCDD and PCDF serum concentrations were small. Although higher maximum levels were found for several congeners in mill workers, these results were not clearly related to occupational characteristics. No meaningful relationship was seen between I-TEQ levels in serum and work in jobs thought to have a potential for exposure. Our conclusions were unaffected by the method we used to impute nondetectable values. Rosenberg et al. also failed to find any statistically significant differences in PCDD or PCDF concentrations in blood plasma between unexposed and potentially exposed workers in the bleach plant or the paper mill of a Finnish pulp and

paper mill.⁶⁾

Table 1. Lipid-adjusted Serum Concentrations of Polychlorinated Dibenzo-*p*-dioxins (PCDDs) in Pulp and Paper Mill Workers and Community Residents

Congener	Exposure Group	N ²	Concentration (ppt) ¹		
			Median	Low	High
2378-TCDD	Community Resident	16	1.8	1.5	3.5
	Worker - Low ³	14	1.9	0.9	5.2
	Worker - High ⁴	32	1.9	0.7	4.9
12378-PeCDD	Community Resident	15	5.6	2.0	7.8
	Worker - Low	14	5.3	3.7	12.3
	Worker - High	32	5.7	2.6	11.3
123478-HxCDD	Community Resident	16	6.2	1.8	11.3
	Worker - Low	12	7.4	1.8	19.1
	Worker - High	31	5.6	2.5	14.7
123678-HxCDD	Community Resident	16	67.0	48.3	101
	Worker - Low	14	79.7	33.1	145

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	Worker - High	25	65.9	29.4	117
123789-HxCDD	Community Resident	16	6.9	3.3	12.8
	Worker - Low	14	9.4	1.9	19.7
1234678-HpCDD	Worker - High	29	8.3	2.9	21.4
	Community Resident	15	95.2	64.1	115
	Worker - Low	13	91.2	47.5	230
1234679-HpCDD	Worker - High	31	73.9	34.0	161
	Community Resident	15	5.2	3.3	14.7
	Worker - Low	9	7.7	3.5	26.8
	Worker - High	19	7.0	3.3	36.5
OCDD	Community Resident	11	547	230	1042
	Worker - Low	10	673	288	1600
	Worker - High	23	541	285	1489

¹ppt = parts per trillion. Imputed values were calculated for results below the limit of detection.

²The number of samples may be fewer than 16 for community residents, 32 for high exposure workers, and 14 for low exposure workers because quality control criteria were not met in some samples.

³Low = low exposure potential (groundwood mill/long log area, wood yard, Kraft mill)

⁴High = high exposure potential (bleach plant, dry end of the paper machines, rewind areas, finishing areas, effluent treatment plant)

Table 2. Lipid-adjusted Serum Concentrations of Polychlorinated Dibenzofurans (PCDFs) in Pulp and Paper Mill Workers and Community Residents

Congener	Exposure Group	N ²	Concentration (ppt) ¹		
			Median	Low	High
2378-TCDF ³	Community Resident	14	1.3	0.8	4.9
	Worker - Low ⁴	14	1.4	0.7	6.7
	Worker - High ⁵	32	1.3	0.4	11.1
12378-PeCDF ³	Community Resident	15	1.3	0.8	2.8
	Worker - Low	14	1.3	0.4	2.1
	Worker - High	30	1.2	0.5	2.4
23478-PeCDF	Community Resident	16	6.4	3.5	13.0

	Worker - Low	14	7.8	3.9	14.1
	Worker - High	31	5.9	2.1	11.3
123478-HxCDF	Community Resident	16	6.4	2.4	9.6
	Worker - Low	14	6.9	2.2	16.2
	Worker - High	32	6.8	2.9	13.2
123678-HxCDF	Community Resident	16	4.8	2.5	7.7
	Worker - Low	14	5.3	2.2	11.7
	Worker - High	31	4.9	2.5	9.4
123789-HxCDF ³	Community Resident	16	1.6	0.8	2.3
	Worker - Low	14	1.4	0.4	3.4
	Worker - High	31	1.3	0.5	3.3
	Community Resident	15	3.9	2.5	14.6
234678-HxCDF ³	Worker - Low	7	4.7	2.3	7.9
	Worker - High	15	4.6	1.9	7.8
	Community Resident	13	15.5	9.5	31.3
1234678-HpCDF	Worker - Low	9	19.5	13.3	28.1
	Worker - High	22	17.3	7.5	33.7
1234789-HpCDF ³	Community Resident	12	2.7	0.8	11.0
	Worker - Low	8	3.4	1.6	4.7
	Worker - High	20	2.6	0.6	7.0

¹ppt = parts per trillion. Imputed values were calculated for results below the limit of detection.

²The number of samples may be fewer than 16 for community residents, 32 for high exposure workers, and 14 for low exposure workers because quality control criteria were not met in some samples.

³The proportion of detectables for this congener was less than 50% of all samples.

⁴Low = low exposure potential (groundwood mill/long log area, wood yard, Kraft mill)

⁵High = high exposure potential (bleach plant, dry end of the paper machines, rewind areas, finishing areas, effluent treatment plant)

Table 3. Toxic Equivalents (I-TEQ) in Pulp and Paper Mill Workers and Community Residents

Exposure Group

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	Community (n=16)	Worker-Low Exposure (n=14)	Worker-High Exposure (n=32)
PCDD Subtotal	13.5 (9.5-19.1) ¹	15.9 (6.5-31.8)	13.3 (7.5-24.9)
PCDF Subtotal	5.0 (3.4-8.8)	5.9 (3.2-11.0)	4.7 (1.9-8.1)
Total I-TEQ	19.1 (12.9-25.9)	21.2 (9.8-41.6)	18.1 (10.7-32.9)

¹ Numbers represent median, minimum, and maximum values.

² Low = low exposure potential areas (groundwood mill/long log area, wood yard, Kraft mill)

³ High = high exposure potential areas (bleach plant, dry end of the paper machines, rewind areas, finishing areas, effluent treatment plant)

5. References

- 1) Patterson DG Jr, Holler JS, Lapeza DR Jr, Alexander LR, Groce DF, O'Connor RC, Smith SJ, Liddle JA, Needham LL (1986): High-resolution gas chromatography/high-resolution mass spectrometric analysis of human adipose tissue for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin. *Anal Chem* 58:705-713.2
- 2) Patterson DG Jr, Hampton I, Lapeza CR Jr, Belser WT, Green V, Alexander IR, Needham LL (1987): High resolution gas chromatographic high resolution mass spectrometric analysis of human serum on a whole weight and lipid bases for 2,3,7,8,-tetrachlorodibenzo-*p*-dioxin. *Anal Chem* 59:2000-2005.
- 3) Akins JR, Waldrep K, Bernert JT (1989): The estimation of total serum lipids by a completely enzymatic summation method. *Clin Chem Acta* 184:219-226.
- 4) Hornung RW, LD Reed (1990): Estimation of average concentration in the presence of nondetectable values. *Applied Occupational and Environmental Hygiene* 5:46-51.
- 5) EPA: Interim Procedures for Estimating Risks Associated with Exposures to Mixtures of Chlorinated Dibenzo-*p*-Dioxins and -Dibenzofurans (CDDs and CDFs) and 1989 Update. Washington, D.C.: U.S. Environmental Protection Agency. EPA/625/3-89/016, March 1991.
- 6) Rosenberg C, Kintzas H, Tornaues J, Mutanen P, Jappinen P, Patterson DG Jr, Needham LL, Vainio H (1994): PCDD/PCDF levels in the blood of workers in a pulp and paper mill. *Organohalogen Compounds* 21:101-104, Kyoto University, Kyoto, 606-01, Japan.