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Occupational exposure to chemical agents in the paper industry

Received: 12 January 2004 / Accepted: 7 April 2004 / Published online: 10 September 2004
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Abstract As part of an International Agency for Research on Cancer (IARC) international epidemiological study of workers in the pulp and paper industry, previously unpublished exposure measurements were assembled in a database. This article summarizes the results of 3,873 measurements carried out in the production departments of paper and paperboard mills and recycling plants in 12 countries. In the paper and paperboard mills, most of the agents were measured in the pulping and refining departments and in on-machine coating and winding of paper/paperboard. Exposures to asbestos, carbon monoxide, formaldehyde, fungal spores, bacteria, nitrogen dioxide, mineral dusts, paper dust, sulphuric acid and different solvents sometimes exceeded exposure limit values. In the re-pulping and de-inking departments of recycling plants high exposures to formaldehyde, fungal spores, bacteria and paper dust were observed. High exposures to asbestos,

bioaerosols, carbon monoxide and paper dust were found in many departments; ammonia, formaldehyde, mineral and paper dust and solvents were found in coating machines; and diphenyl and polychlorobiphenyls (PCBs) were found in some special circumstances. Measurements in the newsprint and uncoated paper machine departments revealed only a few elevated exposures. In nearly all departments, measurements of epichlorohydrin, PCBs, sulphur dioxide, hydrogen sulphide and mercaptans tended to be low, often even below their detection limits. In spite of some uncertainties in the measurement data, the study provides new insights into the level and variation of occupational exposures of production workers in the paper and paperboard industry.

Keywords Occupational exposure · Measurements · Paper industry · Epidemiology

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Introduction

To investigate patterns of cancer incidence and mortality among employees of the pulp and paper industries, the International Agency for Research on Cancer (IARC) is co-ordinating a multicentric epidemiological study involving researchers from 15 countries and representing most paper production regions in the world. The cancer morbidity and mortality rates of more than 100,000 employees working in 75 mills in 15 countries will be studied.

The exposure database created during this large study contains a large number of previously unpublished results of exposure measurements in the pulp and paper industry. The database includes 31,502 exposure measurements of 246 agents sampled from the 1950s to the middle of the 1990s. The structure and content of the database has been described elsewhere (Kauppinen et al. 1997).

The purpose of this article is to summarize the results of exposure measurements carried out in paper and paperboard production and in recycling departments. A summary of occupational hygiene measurements conducted in the non-production departments of pulp, paper, and paper product mills has been published (Teschke et al. 1999). Hygiene measurements in pulp and paper product departments will be described in future publications.

Material and methods

Research institutes and government agencies from the 15 collaborating countries reported national industrial hygiene measurements using a structured format (Kauppinen et al. 1997). The database was compiled at the Finnish Institute of Occupational Health.

For each department and agent, the number of samples, the number of mills, the mean, median, minimum and maximum concentrations, the percentages of measurements below the limit of detection, the lowest measured concentrations, and the threshold limit values (TLVs) of the American Conference of Governmental Industrial Hygienists (ACGIH 1997) and the percentages of measurements over TLV were tabulated. When available, time-weighted average (TWA) data were given priority for reporting. Concentrations below detection limits were assigned the value 0 in the calculation of mean concentrations, because in many cases the limits of detection were not indicated.

Of the total of 31,502 exposure measurements in pulp, paper and paper product industries, 12% were measured in paper, paperboard and paper recycling production between 1950 and 1994. Most of the measurements were from 1980 to 1994, and only a few measurements were from the 1950s.

A total of 3,873 exposure measurements in paper and paperboard production and recycling were received from 12 countries: Canada ($n=88$), Denmark ($n=189$),

Finland ($n=1,288$), Germany ($n=537$), Japan ($n=295$), the Netherlands ($n=99$), Norway ($n=294$), Poland ($n=111$), Spain ($n=44$), Sweden ($n=619$), the UK ($n=23$) and the USA ($n=288$). Measurement data from recycled-paper mills were available from only seven countries: Canada, Denmark, Finland, Germany, Poland, Sweden and the USA. Data were compiled from mills producing a wide variety of products. The data were divided into ten departments in paper and paperboard mills and eight departments in recycled-paper mills.

A wide variety of agents was measured in the paper, paperboard and recycling industries. From the total number (3,873) of exposure measurements 1,535 (40%) were organic compounds other than solvents [e.g. formaldehyde 611, diphenyl 319, polychlorobiphenyls (PCBs) 153 and paper dust 128], 1,046 (27%) mineral and unspecified dusts (e.g. total dust 643, asbestos 135 and respirable dust 99), 516 (13%) inorganic gases (e.g. ammonia 158, chlorine 152 and carbon monoxide 80), 319 (8%) organic solvents (e.g. vinyl acetate 40, isopropanol 32 and solvent hygiene effect 29), 176 (5%) metals and metal compounds (e.g. sodium hydroxide 17, copper 16 and chromium 15), 159 (4%) bioaerosols (e.g. fungal spores 68, total germ count 46 and endotoxins 34) and 122 (3%) other exposures.

In paper and paperboard mills most measurements were made in winding and cutting departments ($n=902$), followed by pulping and refining ($n=593$), calendering or on-machine coating ($n=495$), fine-paper machines ($n=401$), paper machines ($n=357$), soft/tissue-paper machines ($n=284$), paperboard machines ($n=273$), newsprint-paper machines ($n=98$), other specified departments ($n=144$) and unspecified departments ($n=104$).

In the above-mentioned departments, the number of agents over the TLV compared with the number of measured agents was highest in pulping and refining (13/64), followed by winding and cutting (8/71), calendering or on-machine coating (7/35), fine-paper machines (6/23), paperboard machines (5/35), paper machines (4/28), soft/tissue-paper machines (4/12), newsprint-paper machines (3/15), other specified departments (2/19) and unspecified departments (2/31).

In recycled-paper mills most measurements were made in re-pulping departments ($n=103$), followed by de-inking ($n=36$), washing or screening ($n=35$), bleaching ($n=28$), drying ($n=3$), winding or cutting ($n=2$), other specified departments ($n=7$) and unspecified departments ($n=2$).

In the above-mentioned departments only in re-pulping departments did some formaldehyde measurements exceed its TLV value.

Results

Table 1 presents a summary of concentrations measured in paper and paperboard departments, and Table 2

Table 1 Concentrations measured in the paper and paperboard industries, by department and agent (agents with $n \geq 5$ only). – no comparable TLV, TWA time-weighted average of air samples, duration > 1 h, ST short-time air samples, duration < 1 h

Department agent	Unit	No. of samples	No. of mills	Arithmetic mean	Median	Minimum	Maximum	Not detected (%)	Lowest value	TLV 1997	Over TLV 1997(%)	TLV 2003	TWA/ST ^a
Pulping, refining, etc. of stock													
Acetic acid	ppm	10	3	8.1	7.7	0	20	20	3	10	33	10	TWA
Ammonia	ppm	31	6	20.7	8	0	70	3	2	25	31	25	TWA
Asbestos	fibres/cm ³	28	6	1.5	1.2	0	7.1	11	0.001	0.2	64	0.1	TWA
Carbon monoxide	ppm	18	5	1.20	33	0	700	11	4.5	25	61	25	TWA
Coal	mg/m ³	14	1	0.81	0.78	0.21	1.7	0	0.21	2	0	2	ST
Chlorine	ppm	41	2	0.24	0	0	1.9	73	0.2	1	15	1	ST
Dimethylsulphide	ppm	5	2	0.09	0	0	0.47	80	0.47	–	–	–	TWA
Diphenyl	ppm	62	1	2.3	0.4	0.1	13	0	0.1	0.2	84	0.2	TWA
Dust, respirable	mg/m ³	11	7	1.1	0.25	0	5.5	9	0.06	3	9	3	TWA
Dust, total dust	mg/m ³	30	11	0.37	0.21	0	1.6	13	0.05	10	0	10	TWA
Dyes	mg/m ³	76	2	0	0	0	0	100	0	–	–	–	TWA
Formaldehyde	ppm	25	8	0.5	0.4	0	3.1	4	0.01	0.3	56	0.3	ST
Hydrogen sulphide	ppm	10	4	0.15	0.05	0	0.54	40	0.05	10	0	10	TWA
Isopropanol	ppm	7	1	3.5	3.8	0.3	5.1	0	0.3	400	0	200	TWA
Kaolin	mg/m ³	25	4	0.94	0.001	0	21.5	24	0.001	10	4	2	TWA
Methyl mercaptan	ppm	8	3	0.04	0	0	0.33	88	0.33	0.5	0	0.5	TWA
Nitrogen dioxide	ppm	11	3	7.5	1.1	0.2	50	0	0.2	3	36	3	TWA
Polychlorinated biphenyls	µg/m ³	9	2	0.6	0	0	2.3	56	0.2	500	0	500	TWA
Sodium hydroxide	mg/m ³	5	2	0.001	0	0	0.002	60	0.001	2	0	2	TWA
Solvents, hygiene effect	Unitless	8	2	0.06	0.07	0.01	0.08	0	0.01	1	0	1	TWA
Solvents, total	ppm	8	2	3.9	4.5	1.1	5.5	0	1.1	–	–	–	TWA
Sulphur dioxide	ppm	7	3	0.07	0	0	0.28	57	0.1	2	0	2	TWA
Sulphuric acid	mg/m ³	10	1	4.1	2.2	0.3	11.5	0	0.26	1	60	1	TWA
Talc	mg/m ³	58	3	0.75	0.58	0.18	3.8	0	0.18	5	0	2	TWA
Titanium dioxide	mg/m ³	5	1	12.2	2.4	0	29	20	0	10	40	10	TWA
Vinyl acetate	ppm	7	1	0.34	0.5	0	0.7	29	0.2	10	0	10	TWA
Newsprint and uncoated paper machine													
Asbestos	fibres/cm ³	14	2	0.04	0	0	0.11	50	0.01	0.2	0	0.1	TWA
Bacteria	cfu/m ³	6	1	1,240	940	180	3,800	0	180	–	–	–	ST
Carbon monoxide	ppm	5	1	7	6	1	13	0	1	25	0	25	TWA
Dust, containing silica	mg/m ³	10	1	7.6	2.6	0.8	39	0	0.8	–	–	–	TWA
Dust, total dust	mg/m ³	18	3	0.42	0.35	0	1.3	17	0.14	10	0	10	TWA
Formaldehyde	ppm	7	2	0.15	0.04	0	0.46	0	0.04	0.3	29	0.3	ST
Fungal spores	cfu/m ³	6	1	775	485	230	2,100	0	230	–	–	–	ST
Methyl-isothiocyanate	ppm	5	1	246	140	70	650	70	70	–	–	–	TWA
Hydrogen sulphide	ppm	17	2	0.8	0.8	0	2.1	6	0.1	10	0	10	TWA
Fine and coated paper machine													
Ammonia	ppm	58	1	104	31	2	1,005	0	2	25	57	25	TWA
Asbestos	fibres/cm ³	6	1	0.02	0.007	0	0.06	17	0.002	0.2	0	0.1	TWA
Carbon monoxide	ppm	22	1	10.0	5.0	0.2	45	0	0.2	25	9	25	TWA
Chlorine	ppm	10	1	0	0	0	0	100	0	1	0	1	ST
Diphenyl	ppm	98	1	3.2	0.35	0.05	128	0	0.05	0.2	80	0.2	TWA
Dust, total dust	mg/m ³	111	2	1.7	1	0.2	23	0	0.2	10	1	10	TWA

Table 1 (Contd.)

Department agent	Unit	No. of samples	No. of mills	Arithmetic mean	Median	Minimum	Maximum	Not detected (%)	Lowest value	TLV 1997	Over TLV 1997(%)	TLV 2003	TWA/ST ^a
Formaldehyde	ppm	51	6	1.1	0.4	0.01	9.9	0	0.01	0.3	53	0.3	ST
Hydrazine	ppm	6	1	0.02	0.015	0.01	0.02	0	0.01	0.01	100	0.01	TWA
Isopropanol	ppm	5	2	5.6	1.3	0.03	16	0	0.03	400	0	200	TWA
Solvents, hygiene effect	Unitless	5	2	0.24	0.07	0.02	0.96	0	0.02	1	0	1	TWA
Solvents, total	ppm	5	2	21.9	10.1	1.5	79	0	1.45	-	-	-	TWA
Vinyl acetate	ppm	5	2	0.12	0	0	0.3	60	0.3	10	0	10	TWA
Soft/tissue-paper machine													
Asbestos	fibres/cm ³	7	2	1.5	0.74	0	7.3	14	0.25	0.2	100	0.1	TWA
Carbon monoxide	ppm	7	1	20	15	10	45	0	10	25	14	25	TWA
Dust, respirable	mg/m ³	26	3	4.1	0.8	0.3	33.6	0	0.3	3	19	3	TWA
Dust, total dust	mg/m ³	175	5	8.6	5.5	0	71.3	1	0.4	10	29	10	TWA
Fungal spores	cfu/m ³	57	1	1,494	714	40	13,480	0	40	-	-	-	ST
Paperboard machine													
Ammonia	ppm	12	2	6.5	6	0	20	17	0.85	25	0	25	TWA
Benzo(a)pyrene	µg/m ³	86	1	1.3	0.23	0	19	3	0.03	-	-	-	TWA
Dust, total dust	mg/m ³	29	2	2.3	1.4	0	8.8	3	0.09	10	0	10	TWA
Endotoxins	ng/m ³	17	2	1,239	101	3	9,700	0	3	-	-	-	TWA
Epichlorohydrin	ppm	8	1	0.03	0	0	0.09	63	0.07	0.5	0	0.5	TWA
Formaldehyde	ppm	8	1	0.52	0.26	0.18	2.2	0	0.18	0.3	50	0.3	ST
Organic dust	mg/m ³	15	2	2.7	0.64	0.14	13.7	0	0.14	5	25	5	TWA
Polychlorinated biphenyls	µg/m ³	6	1	0.33	0.15	0	1	50	0.3	500	0	500	TWA
Sulphur dioxide	ppm	9	1	0.41	0.3	0.1	0.81	0	0.1	2	0	2	TWA
Sulphuric acid	mg/m ³	5	1	0.89	0.77	0.22	1.6	0	0.22	1	40	1	TWA
Total microbes (mostly bacteria)	cfu/m ³	31	2	34,435	24,000	1,800	130,000	0	1,800	-	-	-	ST
Paper/paperboard machine from more than one of above categories													
Asbestos	fibres/cm ³	27	4	0.07	0.04	0	0.5	11	0.005	0.2	4	0.1	TWA
Dust, respirable	mg/m ³	6	3	4.3	0.19	0	25	17	0.06	3	17	3	TWA
Dust, total dust	mg/m ³	17	8	1.9	0.5	0	18.9	29	0.19	10	6	10	TWA
Epichlorohydrin	ppm	12	3	0	0	0	0.01	92	0.01	0.5	0	0.5	TWA
Formaldehyde	ppm	228	24	0.37	0.2	0	6.6	4	0.011	0.3	37	0.3	ST
Hydrogen sulphide	ppm	8	2	0.16	0.13	0	0.33	13	0.1	10	0	10	TWA
Paper dust	mg/m ³	5	1	1.2	1.2	1	1.4	0	1	5	0	5	TWA
Silica (quartz)	mg/m ³	7	1	0	0	0	0	100	0.0	0.1	0	0.05	TWA
Sodium hydroxide	mg/m ³	5	2	0.05	0	0	0.23	80	0.01	2	0	2	TWA
Sulphur dioxide	ppm	9	2	0.36	0.27	0	1.2	11	0.01	2	0	2	TWA
Sulphuric acid	mg/m ³	6	2	0.11	0.07	0	0.27	50	0.14	1	0	1	TWA
Calendering or on-machine coating													
Ammonia	ppm	51	5	9.2	5	0	70	16	0.27	25	12	25	TWA
Asbestos	fibres/cm ³	11	4	0.04	0	0	0.2	55	0.01	0.2	9	0.1	TWA
Butoxyethanol, 2-	ppm	11	1	0.1	0	0	0.6	82	0.5	25	0	20	TWA
Diphenyl	ppm	67	1	9.5	2.6	0.1	123	0	0.1	0.2	96	0.2	TWA
Dust, total dust	mg/m ³	28	7	1.3	0.2	0	19	18	0.09	10	4	10	TWA
Ethyl acetate	ppm	12	1	74	49	16	256	0	16	400	0	400	TWA
Formaldehyde	ppm	166	10	4.2	0.2	0	50	33	0.02	0.3	54	0.3	ST
Isopropanol	ppm	17	3	11.5	2.1	0	75	18	0.04	400	0	200	TWA

Ozone	ppm	14	2	0.09	0.05	0	0.4	7	0.01	0.05	50	0.05	TWA
Perchloroethylene	ppm	16	2	152	32	0	1,000	6	2	25	75	25	TWA
Solvents, hygienic effect	Unitless	14	2	0.07	0.03	0	0.46	7	0.01	1	0	1	TWA
Solvents, total	ppm	14	2	6.8	2.3	0.1	47	0	0.09	-	-	-	TWA
Toluene	ppm	5	2	12.4	8.9	1.4	32	0	1.4	50	0	50	TWA
Vinyl acetate	ppm	22	3	0.02	0	0	0.4	91	0.1	10	0	10	TWA
Winding, cutting, grading, etc.													
Asbestos	fibres/cm ³	31	8	0.39	0	0	10.5	84	0.2	0.2	16	0.1	TWA
Carbon monoxide	ppm	14	2	117	75	35	350	0	35	25	100	25	TWA
Diphenyl	ppm	90	2	1.6	1.1	0.05	17.9	0	0.05	0.2	88	0.2	TWA
Dust, respirable	mg/m ³	49	28	0.31	0	0	6.9	65	0.05	3	8	3	TWA
Dust, total dust	mg/m ³	214	32	4.3	0.7	0	120	5	0.06	10	7	10	TWA
Epichlorohydrin	ppm	5	2	0	0	0	0	100	0	0.5	0	0.5	TWA
Ethanol	ppm	10	3	94	83.5	0	226	10	3.18	1000	0	1000	TWA
Formaldehyde	ppm	111	17	0.2	0.1	0	1.1	6	0.01	0.3	15	0.3	ST
Methyl ethyl ketone	ppm	8	1	0.21	0	0	0.99	63	0.33	200	0	200	TWA
Organic dust	mg/m ³	28	4	0.56	0.42	0	1.9	4	0.13	5	0	5	TWA
Paper dust	mg/m ³	92	10	3.1	1.6	0	17	14	0.03	5	28	5	TWA
Paperboard dust	mg/m ³	16	3	0.81	0.35	0	2.6	6	0.1	5	0	5	TWA
Silica (quartz)	mg/m ³	31	2	0	0	0	0	100	0.0	0.1	0	0.05	TWA
Total microbes (mostly bacteria)	cfu/m ³	5	1	8,680	2,000	500	30,000	0	500	-	-	-	TWA
Other specified department													
Paperboard dust	mg/m ³	9	4	9.8	2.3	0.9	41	0	0.9	5	44	5	TWA
Polychlorinated biphenyls	µg/m ³	108	2	254	2.7	0	16,000	11	0.01	500	3	500	TWA
Unspecified department													
Asbestos	fibres/cm ³	9	3	0.06	0.009	0	0.22	33	0.007	0.2	11	0.1	TWA
Carbon monoxide	ppm	7	3	8.1	0	0	32	57	10	25	14	25	TWA
Methyl ethyl ketone	ppm	7	1	2.5	2.2	0.1	9.5	0	0.1	200	0	200	TWA
Paper dust	mg/m ³	22	1	1.2	0.7	0.1	4.4	0	0.1	5	0	5	TWA
Polychlorinated biphenyls	µg/m ³	23	2	5.91	3.2	0	17	13	0.3	500	0	500	TWA

"TLV selected as TWA or STEL, given type of measurements (TWA/ST)

Table 2 Concentrations measured in the recycled-paper industry, by department and agent (agents with $n \geq 5$ only). – no comparable TLV, TWA time-weighted average of air samples, duration > 1 h, ST short-time air samples, duration < 1 h

Department agent	Unit	No. of samples	No. of mills	Arithmetic mean	Median	Minimum	Maximum	Not detected(%)	Lowest value	TLV 1997	Over TLV 1997 (%)	TLV 2003	TWA/ST ^a
Re-pulping of waste paper													
Chlorine	ppm	24	1	0.06	0.06	0.06	0.06	0	0.06	1	0	1	ST
Dust, total dust	mg/m ³	10	3	2.3	2.8	0.3	3.5	0	0.3	10	0	10	TWA
Sulphuric acid	mg/m ³	10	1	0.0	0.0	0.0	0.0	100	0.0	1	0	1	TWA
Formaldehyde	ppm	8	2	0.21	0.22	0.05	0.40	0	0.05	0.3	50	0.3	ST
Endotoxins	ng/m ³	7	3	2.9	2.0	0.0	11.0	0	0.01	–	–	–	TWA
Organic dust	mg/m ³	7	2	0.42	0.26	0.16	1.00	0	0.16	5	0	5	TWA
Aluminum oxide	mg/m ³	6	1	0.0	0.0	0.0	0.0	100	0.0	10	0	10	TWA
Paper dust	mg/m ³	6	2	1.2	1.1	0.5	1.9	0	0.49	5	0	5	TWA
Silica	mg/m ³	5	1	0.0	0.0	0.0	0.0	100	0.0	0.1	0	0.05	TWA
De-inking of waste paper													
Chlorine	ppm	24	1	0.06	0.06	0.06	0.06	0	0.06	1	0	1	ST
Total microbes (mostly bacteria)	cfu/m ³	5	1	3,676	1,300	850	9,100	0	850	–	–	–	ST
Sodium hydroxide	mg/m ³	5	1	0.70	0.60	0.30	1.20	0	0.30	2	0	2	TWA
Washing, screening or riffling of recycled paper pulp													
Chlorine	ppm	24	1	0.06	0.06	0.06	0.06	0	0.06	1	0	1	ST
Bleaching of recycled paper pulp													
Chlorine	ppm	24	1	0.06	0.06	0.06	0.06	0	0.06	1	0	1	ST

^aTLV selected as TWA or STEL, given type of measurements (TWA/ST)

presents a similar summary in recycled-paper departments. Data are presented for all agents with at least five measurements/department.

From the total number (3,873) of exposure measurements in paper industries, 3,651 (94%) were conducted in paper and paperboard departments.

In pulping and refining departments, the most frequently measured agents were dyes (13%), diphenyl (11%) and talc. In newsprint and uncoated-paper machine departments, the most often measured agents were total dust (18%), hydrogen sulphide (17%) and asbestos (14%). In fine and coated-paper machine departments, the most often measured agents were total dust (28%), diphenyl (24%) and ammonia (15%). In soft/tissue-paper machine departments, the most frequently measured agents were total dust (62%), fungal spores (20%) and respirable dust (9%).

In paperboard machine departments, the most often measured agents were benzo(a)pyrene (32%), bacteria (11%) and total dust (11%). In paper or paperboard machine departments (from more than one category) the measured agents were mainly formaldehyde (64%), asbestos (8%) and total dust (5%). In calendering or on-machine coating departments, the most frequently measured agents were formaldehyde (34%), diphenyl (14%) and ammonia (11%). In winding, cutting or grading departments, the most often measured agents were total dust (24%), formaldehyde (12%) and paper dust (10%).

In other specified departments, the most frequently measured agents were PCBs (75%), paperboard dust (6%) and white spirits (3%). In unspecified departments of the paper industry, most of the measurements were PCBs (22%), paper dust (21%) and asbestos (9%).

Table 3 shows measurements exceeding the TLV value or high exposure concentrations for microbes (no TLVs available for these agents) in the paper and paperboard industries.

The highest concentrations of asbestos were measured in the pulping and refining and soft-paper departments. The mean concentration of carbon monoxide exceeded its TLV value by four to fivefold in some pulping/refining, winding and cutting departments. The TLV for formaldehyde was exceeded three to fourfold in some fine and coated-paper departments and 13–14-fold in some on-machine coating departments. It is worth noticing that during the recent 10 years the TLVs of, especially, asbestos and formaldehyde have been changed to a fraction of their older values. The highest concentrations of total dust were measured in winding and cutting, paperboard and other unspecified departments. The highest concentrations of paper dust were measured in pulping, soft-paper and other specified departments.

Of the total number of exposure measurements in the paper and recycled paper industry (3,873), only 216 (6%) measurements were conducted in recycled-paper departments.

Formaldehyde was the main component of several chemicals used in the paper and recycled-paper industry,

Table 3 Exposure measurements exceeding the TLV (×) in the paper and paperboard industry, by department and agent. (Endotoxins, bacteria, fungal spores and other microbes have no TLVs) (0 = all measurements below TLV; – = no measurements)

Agent	Unit	Total n	TLV exceeded in paper mill departments													
			P 1 ^a	P 2 ^b	P 3 ^c	P 4 ^d	P 5 ^e	P 6 ^f	P 7 ^g	P 8 ^h	P 9 ⁱ	P 10 ^j				
Agents with measurements $n \geq 5$ in a department (P1–P10):																
Acetic acid	ppm	18	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Ammonia	ppm	140	×	–	×	–	–	–	–	–	–	–	–	–	–	–
Asbestos	fibres/cm ³	135	×	0	0	×	–	–	–	–	–	–	–	–	–	–
Bacteria	cfu/m ³	9	–	×	–	–	–	–	–	–	–	–	–	–	–	–
Carbon monoxide	ppm	79	×	0	×	–	–	–	–	–	–	–	–	–	–	–
Chlorine	ppm	51	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Diphenyl	ppm	307	×	–	×	–	–	–	–	–	–	–	–	–	–	–
Dust, respirable	mg/m ³	92	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Dust, total dust ^k	mg/m ³	624	0	0	×	×	×	×	×	×	×	×	×	×	×	×
Endotoxins	ng/m ³	24	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Formaldehyde	ppm	603	×	×	×	–	–	–	–	–	–	–	–	–	–	–
Fungal spores	cfu/m ³	66	–	×	–	–	–	–	–	–	–	–	–	–	–	–
Hydrazine	ppm	8	–	–	×	–	–	–	–	–	–	–	–	–	–	–
Kaolin	mg/m ³	25	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Nitrogen dioxide	ppm	11	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Organic dust ^l	mg/m ³	45	–	–	–	–	–	–	–	×	–	–	–	–	–	–
Ozone	ppm	45	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Paper/paperboard dust	mg/m ³	150	–	–	–	–	–	–	–	–	–	–	0	–	–	×
Perchloroethylene	ppm	24	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Polychlorinated biphenyls	µg/m ³	153	0	–	–	–	–	–	–	–	–	–	–	–	–	–
Sulphur dioxide	ppm	34	0	–	–	–	–	–	–	–	–	–	–	–	–	–
Sulphuric acid	mg/m ³	21	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Talc	mg/m ³	58	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Titanium dioxide	mg/m ³	8	×	–	–	–	–	–	–	–	–	–	–	–	–	–
Total microbes (mostly bacteria)	cfu/m ³	36	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Number of agents over TLY			13	3	6	4	4	5	4	7	8	2	2	2	2	2

^aPulping and refining of stock

^bNewsprint and uncoated-paper machine

^cFine and coated-paper machine

^dSoft/tissue-paper machine

^ePaperboard machine

^fPaper/paperboard machine, with more categories

^gCalendering or on-machine coating

^hWinding, cutting or grading machine

ⁱOther specified department

^jUnspecified department

^kMainly inorganic non-toxic dusts (calcium carbonate, etc.)

^lDusts containing unspecified organic compounds, mainly paper dust

and the concentrations that exceeded the present TLV value were measured during the re-pulping of waste pulp. Microbes may cause problems because of the warm and wet conditions in many departments, and high concentrations of bacteria were measured, especially in the de-inking processes, in wastewater.

Discussion and conclusions

These exposure data from the paper and paperboard industry provide a good starting point for describing industrial exposures for epidemiological studies. In the assessment of relevant exposures for the international epidemiological study, a team of industrial hygienists has integrated the measurements in the database with mill-specific information from the questionnaires answered by the participating companies, to create a pulp and paper department-exposure matrix (PAPDEM) for 25 major exposure agents. For some agents only qualitative assessment was feasible. The assessment was based on over 31,000 industrial hygiene measurements, company questionnaires, and the experience of the assessment team. A description of this matrix and a summary of exposure estimates have been published elsewhere (Kauppinen et al. 2002).

In the pulping and refining departments of paper and paperboard mills, and in on-machine coating and winding departments, exposures to ammonia, asbestos, carbon monoxide, formaldehyde, fungal spores and bacteria, nitrogen dioxide, mineral and organic dusts, sulphuric acid and different solvents often exceeded the TLV values. In recycling plants in the re-pulping and de-inking departments, high exposures to formaldehyde, fungal spores, bacteria and paper dust were observed. In other departments of the paper and paperboard mills, the measurements in several departments indicated high exposures to asbestos, bioaerosols, carbon monoxide and paper dust and, in some departments, exposures to diphenyl and PCBs. Measurements in the newsprint and uncoated-paper machine departments revealed only a few elevated exposures. In nearly all departments, measurements of epichlorohydrin, PCBs, sulphur dioxide, hydrogen sulphide and mercaptans tended to be low, often below the detection limits.

The TLVs for the measured agents in the 2003 catalogue (ACGIH 2003) were the same, with only a few exceptions, as in the 1997 catalogue used for statistical calculations of measurement data.

It is interesting to consider potential sources of some of these exposures. As would be expected in this industry, paper or paperboard dust or organic dust was a very common exposure in many departments. The occupational exposure limit of organic dust (paper dust) was often exceeded, especially in soft-paper mills. Asbestos exposure was due to asbestos-containing insulating material or brake linings of winding machines or forklifts mainly used during the earlier years.

Nowadays, in many factories they have been changed to asbestos-free ones. Respirable dust, total dust, talc and titanium dioxide (from mineral-coating chemicals) were common high exposure agents in many departments with on-machine coating, calendering, winding and cutting machines. The exposure limits for inorganic dusts were also often exceeded.

Ammonia (a component in mineral-coating paste) exposure exceeded the exposure limit in the stock-preparation and on-machine coating areas of special-paper mills. Elevated carbon monoxide and nitrogen dioxide exposures probably arose from the exhausts of forklifts. Formaldehyde is the main component of many chemicals used in paper or paperboard industry, and high concentrations were measured in many departments. High concentrations of formaldehyde and different solvent vapours from many kinds of coating and laminating chemicals occurred in the calendering area and in the on-machine coating of paper. Sulphuric acid has been, and still is, widely used in the paper and paperboard industry as a pH-adjusting chemical. Hydrazine was mainly used for eliminating oxygen in water for steam generation.

PCBs were used in the earlier eras of the paper industry in capacitors or transformers. The highest concentrations have been measured during accidental exposures. Diphenyl was used as a preservative in fruit wrapping paper only in one mill, where it prompted complaints followed by intense measuring activity revealing locally high concentrations.

High levels of microbes were measured in many departments because of the warm and wet conditions there, when no or little slimicides were used for prevention for bacterial growth. In those departments, high concentrations of endotoxins and bacteria were measured, especially in soft-paper mills.

The database has some limitations. Although it is comprised of measurements from more than 100 mills in 13 countries, the number of mills reporting data for any one agent in a particular department was usually less than five and, in many cases, only one or two. When measurement data for an agent is large and consistent across several departments and agrees with measurements reported in the literature, the results are more likely to be generalizable. The database did not contain information about ventilation, enclosures or other potential exposure determinants. Many measurements were below detection limits, and, in most cases, the detection limits were not reported.

From all exposure measurements in the pulp and paper industry, the most often measured agent groups were organic compounds (40%) other than solvents, followed by mineral and unspecified dusts (27%), inorganic gases (13%), organic solvents (8%), metals and metal compounds (5%), bioaerosols (4%) and other exposures (3%). The most frequently measured exposure agents were total dust (17%), formaldehyde (16%), diphenyl (8%), ammonia (4%), chlorine (4%), asbestos (3%) and paper dust (3%).

The PAPDEM database consists of many measurement data from the pulp and paper industry in Finland (33%), Sweden (16%), Germany (14%), Japan (8 %) and Norway (8%). Few such data (<3% per country) have come from Poland, the Netherlands, Canada, Spain and the UK.

Relatively limited exposure data have been published from the industries under study. Organochlorine compounds have also been identified, usually in trace amounts, in the treatment of the spent liquors in kraft pulp mills (Crawford et al. 1992; Rosenberg et al. 1991, 1994). Exposure to polychlorinated dioxins or furans during the manufacture of pulp, paper or paper products has been estimated and measured (Crandall et al. 1992; Kelada 1990; Krishnan et al. 1990). Paper dust has been measured in different kinds of paper mills (Bullock and Laird 1994; Heederik et al. 1987; Janhunen et al. 1984; Liukkonen et al. 1990; Andreassen-Rix and Lynge 1996; Sahle et al. 1990). Micro-organisms and endotoxins have been measured in recycling and other paper mills (Liukkonen et al. 1990; Andreassen-Rix and Lynge 1996; Liesivuori et al. 1994; Niemelä et al. 1985). National reports are available from Finland on exposures in the pulp industry (Mäkinen et al. 1986), paper industry (Janhunen et al. 1984), paper-products industry (Liukkonen et al. 1988) and pulp/paper recycling mills (Liukkonen et al. 1990), and occupational exposure measurement data of those studies are included in the database. Most of the data reported in the literature are consistent with those in the database.

The database did not collect data on exposures to physical agents such as noise, vibration, ionizing and non-ionizing radiation, etc., which are known to occur in paper and paperboard mills. There are few reports in the literature on physical agents, for example, electric and magnetic field investigations in pulp and paper mills in Canada (Barroetavena et al. 1994).

Chemical exposures in the paper industry may cause various health effects and diseases

A review article that included all relevant reports prior to 1995 (Thorén et al. 1996a) on health effects of workers in the pulp and paper industry revealed that high exposure to chlorine compounds and paper dust was associated with an increased prevalence of impaired lung function, allergic respiratory diseases and death, and reduced sulphur compounds were associated with increased mortality rates due to ischaemic heart diseases.

Increased lung function and respiratory symptoms among workers exposed to soft-paper dust have been reported in some studies (Heederik et al. 1987; Ericsson et al. 1988; Thorén et al. 1989a; Sahle et al. 1990; Thorén et al. 1994). In addition, death from asthma and chronic obstructive pulmonary diseases has been found among workers exposed to soft-paper dust (Thorén et al. 1989b).

The evidence for malignant diseases in pulp and paper mills was reported in a second review article (Thorén et al. 1996b). The authors reported an in-

creased risk for lung cancer among workers exposed to asbestos, chlorine compounds, wood dust, terpenes and slimicides. Weak, although inconsistent, associations for increased cancer mortality rates have also been found in a retrospective follow-up study of 3,241 workers in the pulp and paper industry in Spain (Sala-Serra et al. 1996). In a large Canadian cohort mortality study of 30,157 male workers in pulp and paper mills, an increased mortality rate for malignant cancers was found (Band et al. 1997). Those malignancies were associated with the following known or suspected carcinogens to which the workers were exposed: asbestos (pleura), biocides (kidney), formaldehyde (kidney, brain, Hodgkin's disease) and hypochlorite (oesophagus).

Acknowledgements The authors would like to thank the following industrial hygienists, epidemiologists and other experts in the mills and research institutions for their contributions to this database: Aage Andersen, Shirley Fincham, Murray Finkelstein, Paul Henneberger, E. Hiltun, Danuta Kielkowski, Reiko Kishi, Jean Claude Limasset, Robert Rajan, Alain Robert, Maria Sala-Serra and Allison Tepper. Health hazard evaluation data of the National Institute for Occupational Safety and Health (NIOSH) were included in the database. The valuable help of Agneta Hollander and Steve Prahacs (Canadian Pulp and Paper Association), and of Paavo Jäppinen (Stora Enso, Finland) in the construction of the classifications for this project is gratefully acknowledged. The study was partly supported by the European Commission BIOMED-1 and BIOMED-2 programmes.

References

- ACGIH (1997) TLVs and BEIs. In: Threshold limit values (TLVs) for chemical substances and physical agents and biological exposure indices (BEIs). Cincinnati
- ACGIH (2003) TLVs and BEIs. In: Threshold limit values (TLVs) for chemical substances and physical agents and biological exposure indices (BEIs). Cincinnati
- Andreassen-Rix B, Lynge E (1996) Industrial hygiene measurements in a new industry—the repulping and deinking of paper waste. *Am J Ind Med* 30:142–147
- Band PR, Nhu DL, Fang R, Threlfall WJ, Astrakianakis G, Anderson JTL, Keefe A, Krewski D (1997) Cohort mortality study of pulp and paper mill workers in British Columbia, Canada. *Am J Epidemiol* 146:186–194
- Barroetavena MC, Ross R, Teschke K (1994) Electric and magnetic fields at three pulp and paper mills. *Am Ind Hyg Assoc J* 55:358–363
- Bullock WH, Laird LT (1994) A pilot study of the particle size distribution of dust in the paper and wood products industry. *Am Ind Hyg Assoc J* 55:836–840
- Crandall MS, Kinnes GM, Hartle RW (1992) Levels of chlorinated dioxins and furans in three occupational environments. *Chemosphere* 25:255–258
- Crawford RJ, Cloutier SL, Rovell-Rix DC (1992) Evaluation of OSHA method 5 for measuring chloroform in pulp and paper industry workplace and ambient atmosphere. *Am Ind Hyg Assoc J* 53:210–215
- Ericsson K, Jarvholm B, Norin F (1988) Respiratory symptoms and lung function following exposure in workers exposed to soft paper tissue dust. *Int Arch Occup Environ Health* 60:341–345
- Heederik D, Burdorf L, Boleij J, Willems H, van Bilsen J (1987) Pulmonary function and intradermal tests in workers exposed to soft-paper dust. *Am J Ind Med* 11:637–645

- Janhunen H, Korhonen K, Mäkinen R (1984) Chemicals in the paper and paperboard industry—use, exposures, and health risks in Finland (In Finnish with English summary). *Työterveyslaitoksen tutkimuksia* 2:215–229
- Kauppinen T, Teschke K, Savela A, Kogevinas M, Boffetta P (1997) International data base of exposure measurements in the pulp, paper and paper product industries. *Int Arch Occup Environ Health* 70:119–127
- Kauppinen T, Teschke K, Keefe A, Nicol A-M, Astrakianakis G, Boffetta P, Colin D, Korhonen K, Liukkonen T, Pannett B, Westberg, H (2002) Assessment of exposure in an international study on cancer risks among pulp, paper and paper product workers. *Am Ind Hyg Assoc J* 63:254–261
- Kelada FS (1990) Occupational intake by dermal exposure to polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans in the pulp mill industry. *Am Ind Hyg Assoc J* 51:519–521
- Krishnan R, Meyer C, Goodman B, Foerst MB (1990) Estimated worker exposure to 2378-TCDD and 2378-TCDF in the manufacture, processing, and commercial use of pulp, paper and paper products. *Government Reports Announcements and Index (GR&I)*, Issue 24, Washington
- Liesivuori J, Kotimaa M, Laitinen S, Louhelainen K, Pönni J, Sarantila R, Husman K (1994) Airborne endotoxin concentrations in different work conditions. *Am J Ind Med* 25:123–124
- Liukkonen T, Korhonen K, Mäkinen R, Lindroos L (1988) Use of chemicals in the paper refinement industry (In Finnish with English summary). *People Work* 2:27–40
- Liukkonen T, Lindroos L, Kotimaa M (1990) Chemical and biological health hazards in recycled pulp production (In Finnish with English summary). *People Work* 4:257–266
- Mäkinen R, Kangas J, Korhonen K (1986) Chemicals in the pulp industry—use, exposures, and health risks in Finland (In Finnish with English summary). *Työterveyslaitoksen tutkimuksia* 4:17–30
- Niemelä SI, Vääränen P, Mentu J, Jokinen A, Jäppinen P, Sillanpää P (1985) Microbial incidence in upper respiratory tracts of workers in the paper industry. *Appl Environ Microbiol* 50:163–168
- Rosenberg C, Nylund L, Aalto T, Kontsas H, Norppa H, Jäppinen P, Vainio H (1991) Volatile organohalogen compounds from the bleaching of pulp—occurrence and genotoxic potential in the work environment. *Chemosphere* 23:1617–1628
- Rosenberg C, Kontsas H, Jäppinen P, Tornaues J, Hesso A, Vainio H (1994) Airborne chlorinated dioxins and furans in a pulp and paper mill. *Chemosphere* 29:1971–1979
- Sahle W, Sällsten G, Thoren K (1990) Characterization of airborne dust in a soft paper mill. *Ann Occup Hyg* 34:55–75
- Sala-Serra M, Sunyer J, Kogevinas M, McFarlane D, Antó JM (1996) Cohort study on cancer mortality among workers in the pulp and paper industry in Catalonia, Spain. *Am J Ind Med* 30:87–92
- Teschke K, Ahrens W, Andersen A, Boffetta P, Fincham S, Finkelstein M, Henneberger P, Kauppinen T, Kogevinas M, Korhonen K, Liss G, Liukkonen T, Osvoll P, Savela A, Szadkowska-Stanczyk I, Westberg H, Widerkiewicz K (1999) Occupational exposure to chemical and biological agents in the nonproduction departments of pulp, paper and paper product mills: an international study. *Am Ind Hyg Assoc J* 60:73–83
- Thorén K, Sallsten G, Bake B, Drake U, Jarvholm B, Sahle W (1989a) Lung function and respiratory symptoms among workers in a soft paper mill. *Int Arch Occup Environ Health* 61:467–471
- Thorén K, Jarvholm B, Morgan U (1989b) Mortality from asthma and chronic obstructive pulmonary disease among workers exposed in a soft paper mill: a case referent study. *Br J Ind Med* 46:192–195
- Thorén K, Jarvholm B, Sallsten G, Thiringer G (1994) Respiratory symptoms and asthma among workers exposed to paper dust: a cohort study. *Am J Ind Med* 26:489–496
- Thorén K, Hagberg S, Westberg H (1996a) Health effects of working in pulp and paper mills: exposure, obstructive airways diseases, hypersensitivity reactions and cardiovascular diseases. *Am J Ind Med* 29:111–122
- Thorén K, Persson B, Wingren G (1996b) Health effects of working in pulp and paper mills: malignant diseases. *Am J Ind Med* 29:123–130