

Occupational exposure to chemical agents in the particleboard industry

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KAUPPINEN TP, NIEMELÄ RI. Occupational exposure to chemical agents in the particleboard industry. *Scand J Work Environ Health* 11 (1985) 357–363. Air contaminants were measured at eight Finnish particleboard plants in 1965–1984. Most of the measurements concerned formaldehyde and wood dust. The other substances measured included terpenes, solvents, and heptachlor. Before 1975 the formaldehyde concentration regularly exceeded 2 ppm during many workphases. Considerable improvements in ventilation and the composition of glues have occurred since then, and today the exposure level is below 2 ppm during most workphases. High peak concentrations, 20–30 ppm at the highest, were characteristic of exposure in earlier years. The concentrations of wood dust have also decreased, eg, from over 5 mg/m³ to 1 mg/m³ or below during forming. These data have been used to evaluate past exposures in an epidemiologic study on cancer risks in the particleboard, plywood, and sawmill industries.

Key terms: formaldehyde, wood dust.

The main chemical agents used in particleboard plants are the glues applied to bind the chips together for the formation of the board. Urea-formaldehyde resins (carbamide resins) were introduced in Finland in the 1950s, and they are still the most popular glues. Urea-melamine and phenol-formaldehyde resins are used to a less extent. Some boards for export are glued with resins which contain pesticides (eg, heptachlor), but on the whole pesticides are used in glue mixtures only rarely.

The other substances present in Finnish particleboard plants include wood dust, the volatile components of spruce and pine, the exhaust gases from forklift trucks, and the thermal degradation products of resins emitted during hot pressing. If the plant has a coating department, the ambient air also contains solvent vapors. A potent carcinogen, bis(chloromethyl)ether (BCME), may also be formed if urea-formaldehyde resins, which contain ammonium chloride, are used (23).

Workers exposed to formaldehyde often experience irritation of the eyes, nose, and throat (10). Irritation of the nose and throat can also be associated with exposure to terpenes present in the drying vapors of coniferous woods. These effects have been studied in more detail in sawmills (12). Still another weak irritant is wood dust from birch, spruce, or pine (5). These woods are not known to cause other hazards, such as asthma or skin allergy. Allergic symptoms, however, may be caused by exposure to formaldehyde, a well-known sensitizing agent.

The suspected or known carcinogens found in particleboard plants include formaldehyde (1, 9), wood dust (7), and heptachlor (6). The exhaust gases given off by forklift trucks contain carcinogenic polycyclic aromatic hydrocarbons (8). Very little is known about the possible exposure to bis(chloromethyl)ether and the thermal degradation products of glues. To our knowledge, no epidemiologic studies have been published on cancer risks in the particleboard industry.

Only a few reports have been published on exposure to formaldehyde in the particleboard industry. Flick (4) used the pararosaniline method for 21 formaldehyde measurements taken in four particleboard plants in the Federal Republic of Germany. The average formaldehyde concentration was 2.0 (range 0.5–5.3) ppm. The formaldehyde concentration varied from 0.5 to 2.6 ppm at the control panels of the hot presses and from 0.6 to 3.8 ppm at the exits of the hot presses and at the cooling lines. The highest concentration (5.3 ppm) was measured at a resin-mixing stand. Wayne et al (22) reported the results of measurements taken in two particleboard plants in the United States. The measurements were performed with the chromotropic acid method. A total of 15 air samples was taken at the first plant. The average formaldehyde concentration was 1.28 (range 0.6–2.7) ppm. The concentration averaged 1.48 (range 0.9–2.5) ppm in the assembly line area, 1.30 (range 1.0–2.7) ppm in the finishing area, and 1.13 (range 0.6–2.1) ppm in the warehouse. The second plant had three principal areas, an assembly line for particleboard, a plywood plant, and a joint finishing area for particleboard and plywood. Six samples were taken along the particleboard assembly-line and eight samples in the joint finishing area. The concentrations averaged 0.92 (range 0.5–1.4) ppm at the assembly line and 0.92 (range 0.7–1.5) ppm in the joint finishing area. Rosén et al (17) measured formaldehyde in two Swedish plants. Their method was based

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on solid sorbent sampling and liquid chromatography. The mean concentration of the 21 samples was 0.33 ppm, with a standard deviation of 0.20 ppm.

Since 1965 the Finnish Institute of Occupational Health has made hygienic measurements at eight particleboard plants. The present article is a summary of the data on past and new measurements. These data have been used to evaluate exposures in an epidemiologic case-referent study of a cohort of Finnish particleboard, plywood, and sawmill workers.

Methods

Most of the measurements concerned either formaldehyde (334 samples) or wood dust (71 samples). Terpenes, solvents, and heptachlor were measured less often. Chiefly because of analytical difficulties neither bis(chloromethyl)ether nor thermal degradation products were measured at all. The authors themselves made measurements in four plants and evaluated the remaining data collected from the measurement reports of the Institute of Occupational Health. Such reports include a description of the analytical methods used, remarks on prevailing circumstances during the measurements, the sampling sites and times, the results, and pertinent comments. The analytical methods applied have been tested in the Institute of Occupational Health.

Formaldehyde

The standard method used to measure formaldehyde in the workroom air comprises sampling into a dilute sodium bisulfite solution and spectrophotometric determination after evaporation of the sample solution and the addition of a color-forming reagent, chromotropic acid (20). The sampling pumps used were battery operated. The typical sampling rate was 1 l/min, and the sampling time about 1 h. The lowest detectable concentration was approximately 0.02 ppm (16). The samples were taken as close to a worker's breathing zone as practically possible.

The formaldehyde concentration was continuously monitored in two plants in a study of temporal fluctuations. A 12-channel infrared-spectrophotometer (Miran multipoint ambient air analyzer with Miran 1A measuring head, Foxboro Analytical) was used for this purpose. The analyzing wavelength was 3.48 μm , and the path length 20.25 m. Since the accuracy of the method was insufficient at the level of 1 ppm, the readings were regularly calibrated according to the chromotropic acid procedure. The Miran readings were averaged so as to correspond to a sampling time of 1 h.

The results obtained before 1975 are based on a somewhat different procedure. The sample was collected in a dilute sodium bisulfite solution, which was then titrated with iodine, with starch as the indicator. The titration and the chromotropic acid methods were

compared in laboratory tests, with about the same sample volumes and formaldehyde concentrations as in a typical hygienic sample. The differences in the results were slight, and so the results of these two methods were considered comparable.

Wood dust

The Finnish standard measurement method for dust in the workroom air involves its collection on a membrane filter (Millipore filter AAWP 037) and weighing (18). Electric-circuit driven pumps with a sampling rate of about 20 l/min were used for the area sampling. Battery-operated pumps and a sampling rate of 1 l/min were used in the personal sampling. With the common sampling time of 4 to 6 h the detection limits of the method are below 0.1 mg/m³ for area sampling and 0.2–0.3 mg/m³ for personal sampling.

Terpenes

The volatile components from pine and spruce contain monoterpenes, eg, α -pinene, β -pinene, and Δ -carene (11). These substances were sampled into charcoal tubes and determined by gas chromatography according to the Finnish standard method (19). The battery-operated pumps had a rate of 0.2 l/min, and the sampling time was about 1 h. Terpenes were desorbed from charcoal with dimethylformamide and determined by a gas chromatograph equipped with a flame ionization detector. The column was 10 % Carbowax 20M on Chromosorb AW DMCS 80/100. The column temperature was 85°C. The detection limits were under 0.1 ppm for α - and β -pinene and about 1 ppm for Δ -carene.

Other substances

The Finnish standard method, based on charcoal tube sampling and gas chromatographic analysis, was used to measure solvent vapors given off by coating mixtures which contained, eg, xylene, toluene, or white spirit (19). Heptachlor was sampled into a toluene solution and determined by a gas chromatograph equipped with an electron capture detector. The column was 3 % QF-1 on Chromosorb AW DMCS 80/100 at 170°C. Hydrochloric acid was collected into a 0.005-M sodium hydroxide solution, neutralized with nitric acid, and titrated with 0.01-M silver nitrate, with potassium chromate as the indicator.

Sampling sites

The samples were taken at sites and during workphases where exposure was thought to be probable. Area sampling was generally used, as most workphases are carried out in rather stable sites. Area sampling was preferred also because the sampling method of formaldehyde is based on liquid absorption and because area sampling allowed the use of stronger pumps, which permit the collection of larger air volumes in the sampling of wood dust.

Results

The results of the formaldehyde measurements are presented in table 1. The ranges of the means in different workphases were 0.8–4.8 ppm in 1965–1974 and about 0.1–2.3 ppm in 1975–1984. The corresponding results were also calculated with plant medians and means only to balance any possible effect caused by

different numbers of measurements in different plants. The results of these calculations were very similar on the whole and thus indicated that this effect was small. The typical fluctuation in the formaldehyde concentration over time is shown in figure 1 for one particleboard plant. Table 2 gives two examples of the effects of the free formaldehyde content in the glue.

Table 1. Formaldehyde concentration in the workroom air of the particleboard plants by workphase or worksite and period of measurement.

Workphase or worksite	Formaldehyde concentration (ppm)			Number of plants	Number of measurements
	Range	Median	Mean		
Chipping 1975–1984	< 0.1 and 0.1	.	.	1	2
Preparation of glue mixture 1975–1984	0.3–4.9	1.9	2.2	1	10
Blending 1965–1974	0.1–2.0	1.0	1.0	2	10
1975–1984	< 0.1–1.4	0.7	0.7	2	8
Forming 1965–1974	< 0.5–4.6	1.3	1.7	4	26
1975–1984	0.1–4.8	0.9	1.4	5	32
Prepressing 1965–1974	1.0–6.1	5.1	3.4	1	5
Hot pressing 1965–1974	1.1–9.5	3.1	3.4	5	35
1975–1984	0.2–4.6	1.4	1.7	5	61
Celling crane 1965–1974	4.0–8.6	4.9	4.0	1	6
1975–1984	0.5–1.0	0.7	0.7	1	4
Planing 1965–1974	1.5–6.4	4.2	4.0	2	3
1975–1984	0.4–0.8	0.7	0.6	1	4
Sawing 1965–1974	0.7–9.2	4.1	4.8	3	17
1975–1984	< 0.1–3.3	0.6	1.0	4	36
Grinding 1965–1974	0.7 and 0.9	.	.	1	2
1975–1984	< 0.1 and 0.2	.	.	1	2
Grading 1965–1974	0.1–2.5	0.8	1.1	1	9
Packing 1965–1974	0.9 and 0.9	.	.	1	2
1975–1984	1.4–3.0	2.5	2.3	1	5
Coating 1965–1974	0.5–1.8	0.9	1.0	1	7
1975–1984	0.1–1.2	0.3	0.4	2	12
Jointing of particleboard 1965–1974	2.1–4.0	3.0	3.0	1	3

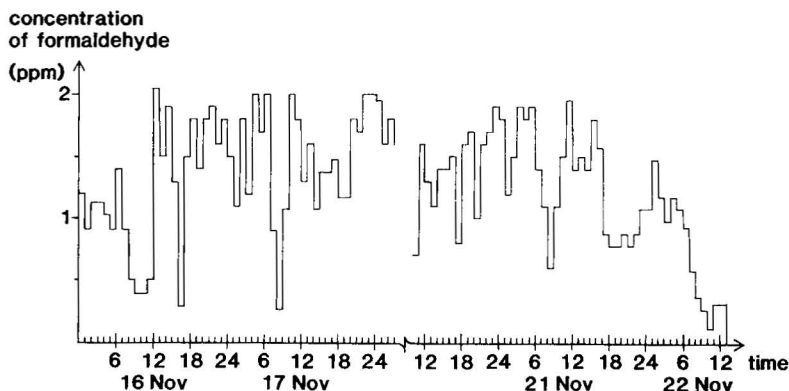


Figure 1. Typical temporal variation of formaldehyde concentration at the continuously operating press in one particleboard plant.

Table 2. Formaldehyde concentration in the workroom air by content of free formaldehyde of glue in two particleboard plants.

Plant and season	Work phase									Free formaldehyde content of glue (%)
	Forming			Hot pressing			Finishing			
	Number of measurements	Formaldehyde concentration (ppm)		Number of measurements	Formaldehyde concentration (ppm)		Number of measurements	Formaldehyde concentration (ppm)		
		Mean	SD		Mean	SD		Mean	SD	
Plant A, summer	6	3.4	1.3	13	2.7	1.1	6	1.4	0.6	0.4
Plant A, winter	3	0.5	0.1	3	1.2	0.3	6	0.7	0.1	≤0.2
Plant B, autumn	26	0.5	0.1	26	1.3	0.4	5	1.1	0.4	≤0.5
Plant B, spring	8	0.7	0.2	8	2.8	0.7	8	2.3	0.8	0.7

Table 3. Wood dust concentration in the workroom of the particleboard plants by workphase and period of measurement.

Workphase	Wood dust concentration (mg/m ³)			Number of plants	Number of measurements
	Range	Median	Mean		
Hogging ^a 1975—1984	0.1—29	3.9	11	1	3
Chipping ^a 1965—1974	1.1	.	.	1	1
1975—1984	0.7—1.7	1.0	1.1	2	3
Drying of chips ^a 1965—1974	24—29	.	.	1	2
Blending ^a 1965—1974	1.0—8.0	7.0	5.3	1	3
1975—1984	0.6—0.9	0.8	0.8	1	3
Forming 1965—1974	4.0—26	12	13	3	9
1975—1984	<0.1—0.5	0.4	0.4	2	4
Prepressing 1965—1974	6.0 and 7.0	.	.	1	2
Hot pressing 1965—1974	1.0—6.1	4.6	4.1	4	6
1975—1983	<0.1—2.1	0.5	0.8	2	5
Planing 1965—1974	0.6	.	.	1	1
Sawing 1965—1974	10—20	14	14	1	4
1975—1984	<0.1—2.3	1.1	1.1	2	9
Grinding 1975—1984	0.6 and 0.8	.	.	1	2
Grading 1975—1984	1.1	.	.	1	1
Packing 1975—1984	<0.6 and 0.6	.	.	1	2
Matching 1975—1984	1.2	.	.	1	1

^a Exposure short in duration.

The indicator tube measurements revealed high peak concentrations of formaldehyde in the 1960s during the opening of the hot press. The highest concentrations measured were 20—30 ppm. In recent measurements the concentrations have been lower (1—7 ppm).

High concentrations were occasionally measured in exceptional situations. In one old plant the formaldehyde concentration rose to over 20 ppm during the hot pressing and forming when natural ventilation was restricted by the shutting of the doors and windows. The formaldehyde concentration was over 2 ppm also during other workphases, eg, blending, sawing, grinding, grading, and packing, as well as in the ceiling

crane. These results, from 1971, are based on 33 samples (sampling time about 30 min).

The concentrations of wood dust are shown in table 3. At stable worksites the means ranged from 4.1 to 14 mg/m³ in 1965—1974 and from 0.4 to 1.1 mg/m³ in 1975—1984. The wood is usually a mixture of Finnish species, such as spruce, pine, aspen, and birch. Higher concentrations of wood dust were measured in a special situation, when particleboard was manufactured from sawdust. The concentration was 26—360 mg/m³ during forming (four samples), 23—58 mg/m³ during sawing (three samples), 15—17 mg/m³ during

prepressing (two samples), and 13 mg/m³ during hot pressing (one sample).

Terpenes evaporating from coniferous raw materials of particleboard were measured in one plant. The total concentration of monoterpenes (α - and β -pinene and Δ -carene) was 8.1 ppm in the chipping area and 7.5 ppm in the nearby blending area. The concentration of terpenes measured during operations after the drying of chips (eg, forming, hot pressing, grinding, and sawing) was below the detection limit of the method, which was 1–2 ppm, depending on the sample volume.

The concentrations of solvents during the coating of particleboard depended on the solvent mixture used and the ventilation. The following concentrations were measured during puttying (nine samples): 1–5 ppm of toluene, 3–44 ppm of xylene, and from slight traces to 120 ppm of white spirit. Three samples taken during polyvinyl chloride coating contained 4–6 ppm of toluene and 3–4 ppm of xylene.

Heptachlor was measured in one plant during a trial run of particleboard protected with pesticides. The concentrations varied from below 10 μ g/m³ during grading (near the end of the process) to 120 μ g/m³ during forming (near the beginning of the process). The concentrations during workphases between the forming and grading were 20–40 μ g/m³. Nine samples were taken.

Hydrochloric acid was measured in the hot pressing area of one plant for a determination of whether bis-(chloromethyl)ether could be formed when urea-formaldehyde glues, which contain ammonium chloride, are used. The concentration of hydrochloric acid was 0.5 ppm when the formaldehyde concentration at the same site and time (sampling time 64 min) was 1.1 ppm.

Discussion

The presently valid Finnish exposure limit for formaldehyde in the workroom air is 1 ppm (3). Earlier the value was 5 ppm (before 1972) and 2 ppm (1972–1980). The American Conference of Governmental Industrial Hygienists (ACGIH) has adopted a threshold limit value (TLV) of 2 ppm, with the intention of a change to 1 ppm in 1984–1985 (2).

In particleboard plants the level of 1 ppm was exceeded regularly during many workphases in the 1960s and early 1970s. Even levels above 2 ppm were common. The heaviest exposure, typically 2–5 ppm, usually occurred in the hot pressing area or in its vicinity (eg, prepressing, the ceiling crane, planing, and sawing). The concentrations measured in the mid-1970s are compatible with the results of Flick (4) and Wayne et al (22). The results are also consistent with those obtained in Sweden (17) in the 1980s. Differences in ventilation, the composition of glues, and process parameters are naturally reflected in the concentrations

measured. From the point of view of general exposure levels, however, the concentrations in these particleboard plants are similar.

The measured concentrations are on the same level as the lower and intermediate concentrations used for a carcinogenesis bioassay of formaldehyde in rats. None of the 236 rats with inhalation exposure to 2.0 ppm contracted nasal cancer. After exposure to 5.6 ppm, 2 of 235 rats developed nasal cancer. The highest concentration tested was 14.3 ppm; it caused 103 nasal cancers among 232 rats (9). Man's risk of nasal cancer cannot, however, be concluded simply by comparisons between exposures in the work environment and in animal experiments.

The formaldehyde concentration may vary considerably over time at the same measuring site, as shown in figure 1. In figure 1 the concentration monitored for 3.5 d at the control panel of a continuously operating hot press is presented. The sharp drops in the concentration profile occurred either when the large transport door between the manufacturing hall and the warehouse was open or when there were breaks in the process.

That the formaldehyde concentration has decreased over time is obvious when the data collected before and after 1975 are compared. This decrease is visible in the median and mean values shown in table 1; it occurred in every workphase with comparable results and for which at least three samples had been taken. The median concentrations detected in 1975 or after are 14–70 (average 38) % of the corresponding values measured before 1975. Should formaldehyde prove to be a human carcinogen, this improved exposure situation indicates that recently exposed particleboard workers are at less risk of contracting (respiratory) cancer caused by formaldehyde than particleboard workers exposed in the past.

The formaldehyde concentration in the air of particleboard plants depends on the composition of glue, the production parameters, and the efficiency of ventilation. The amount of free formaldehyde in urea-formaldehyde resins has a considerable effect on formaldehyde release (13). Additives in the hardener may reduce the content of free formaldehyde in particleboard. Process parameters, such as the press temperature, the press time, and the moisture of the wood also affect formaldehyde release.

In order to improve the work conditions in Finnish particleboard plants, the composition of glue and the process parameters have been modified in recent years. According to data of the Finnish Particleboard Association the perforate value, which describes the amount of free and loosely bound formaldehyde in particleboard, has decreased from 0.10 % (before 1970) to 0.03 % (in 1979). The content of free formaldehyde in glue seems to have a considerable effect on the formaldehyde concentration in the air of the plant (table 2). The formaldehyde concentrations in plant A were reduced by about 50 % when glue with a free formal-

dehyde content of 0.4 % was replaced with glue containing less than 0.2 % free formaldehyde. Changes in the recirculation of air also contributed to this improvement. The concentrations in plant B increased appreciably at the press and in the finishing area when a glue with 0.7 % of free formaldehyde was substituted for a glue with a free formaldehyde content of below 0.5 %.

More effective ventilation has also contributed to the decrease in formaldehyde concentration that has occurred over time. Twenty years ago ventilation rates were usually from two to three changes per hour according to the experience of the Institute of Occupational Health. The presses were open, with no enclosures or exhaust hoods. Today, the air exchange rates range from 7 to 20 per hour (15). In modern plants the hot presses and other sources of formaldehyde emission are equipped with enclosures or exhaust hoods to prevent the dispersion of formaldehyde vapor.

Before 1975 the concentrations of wood dust near forming and sawing machines were usually over 5 mg/m³, the present exposure limit in Finland (3) and the TLV for softwood dust adopted by the ACGIH (2). The concentrations show, however, a clear decrease during all workphases for which the data before and after 1975 are comparable. (See table 3.) The present exposure level is usually well below 5 mg/m³, often even below 1 mg/m³. But it should be noted that the wood dust in particleboard plants may contain uncured formaldehyde resin and other components of glue mixtures, and these ingredients may contribute to the toxicologic properties of wood dust. The main reason for the decrease in exposure to wood dust over time is likely to be the improved local ventilation at the forming and sawing machines. Changes in the composition of the raw materials may also have affected the measured results, as was clearly indicated by the results for the manufacture of particleboard from sawdust.

Exposure to terpene vapor in the processing of chips was rather low (below 10 ppm) compared to the concentrations measured in sawmills when pine is being sawed (usually 20–60 ppm) (11). This low result, however, is based on only a few samples, and the concentration of terpenes depends on the amount of coniferous wood in raw materials and on the ventilation. Evaluation of the results is further complicated by the lack of exposure limits for terpenes.

The formation of bis(chloromethyl)ether is theoretically conceivable on the basis of the simultaneous occurrence of hydrochloric acid and formaldehyde in hot pressing. Further studies on this compound in the air of particleboard plants are required, as exposure to even a very low (0.1 ppm) concentration has caused cancer in animal experiments (21). Because the formation of bis(chloromethyl)ether is a complex process which depends on the phase (liquid or gas), the temperature, the pH, and the water content of the

medium (23), the substance should be measured directly. A tested mass spectrometric method, which reportedly avoids the many pitfalls of bis(chloromethyl)ether analysis, is available (14).

Acknowledgments

We want to thank Mr H Havukkala for collecting the old hygienic data, Mr S Aatola, Mr E Kähkönen, Mr P Mäkelä, Mr H Riipinen, and Mr A Vähä-Söyrinki for assisting with the field measurements, and Mr E Priha and Mr I Ahonen for testing the measurement methods for formaldehyde.

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Received for publication: 3 May 1985