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(First received 13 January 1975 and in final form 14 May 1975)

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A mathematical model for the room air concentration of formaldehyde has been established. The model is

$$E = \frac{(RT + S)(aH + b)}{1 + (nc/\alpha)} \text{mg HCHO m}^{-3} \text{ air}$$

where a , b , c , S and R are constants depending on type and surface coating of chipboard. T is air temperature $^{\circ}\text{C}$, n ventilation, air changes h^{-1} and α area board/room volume, $\text{m}^2 \text{m}^{-3}$. The constants a , b , c and S were determined on the basis of the chamber measurements, while R has to be calculated for each room from the results of measurements in the room. The model reproduces the measurements in dwellings and in the climate chamber with correlation coefficients of 0.88 and 0.94 respectively in the intervals 17–32 $^{\circ}\text{C}$, 5–13 g H_2O kg air $^{-1}$ and 0.4–3 air changes h^{-1} and may be used for prediction of formaldehyde concentrations in rooms containing chipboard.

The adverse health effects of low levels of formaldehyde are irritation of the upper airways and conjunctivitis. The need for air quality standards and control programmes for indoor air in the home is stressed.

INTRODUCTION

Off-gassing properties of materials are normally regarded as a problem only in confined spaces designed for long-term stay, e.g. nuclear submarines or manned spacecraft (Hodgson and Pustinger, 1966). The problem, however, is of general hygienic importance as a multitude of airborne contaminants are being generated within the home environment from building materials, fixtures, furniture, textiles and various household products. The purpose of this paper is to analyze the emanation of formaldehyde from chipboard used as a building material. The dependence of the emanation on indoor climatic parameters is described and a mathematical model is developed for prediction purposes.

Chipboard

The use of chipboard has increased rapidly during the last decade, and the world consumption in 1970 was $1.8 \times 10^6 \text{ m}^3$ (1.2×10^6 tons) (FAO, 1970). In the home environment the material is used for partitions, ceilings and floors, for wardrobes and furniture. Chipboard consists of woodshavings held together by a glue, usually a urea-formaldehyde glue with an approximate molecular ratio of 1:1.4. At high pressure

and temperature the glue polymerizes. During this condensation and the subsequent hardening, which is influenced by acid-base catalysis (De Jong and De Jonge, 1952; Mellegaard, 1967) water vapour and formaldehyde are released, but 0.04 per cent free formaldehyde remains in the board, which in the course of time is released, and partly replaced by formaldehyde regenerated from the polymerized urea-formaldehyde glue.

MATERIALS AND METHODS

Identical equipment for suction of 50 l. room air through two washing bottles was used for the field measurements and the measurements in the climate chamber. The laboratory analysis was carried out with the chromotropic acid method (Nordisk Metodisk Komite, 1964; Altschüller *et al.*, 1961) with a reproducibility of ± 5 per cent at 1.0 mg formaldehyde m^{-3} air, and a detection limit of 0.1 mg m^{-3} air. The simultaneous ventilation-measurement was performed by injection of a radioactive gas (^{85}Kr) and after a mixing period the decrease in radioactivity was monitored for 6–8 h. Indoor climatic parameters were measured with a barograph, a thermohygrograph and a psychrometer. In addition, outdoor wind speed and

cluded 5.5, 9.5 and 13 g H₂O kg⁻¹ dry air all at 22°C and 0.5 air change h⁻¹. The study of ventilation included 0, 0.5, 1 and 3 air changes h⁻¹ all at 22°C and 5.5 g H₂O kg⁻¹ dry air, and finally the ageing of the plates was studied with an interval of 13 months; the temperature was 22°C, the humidities 5.5 and 13 g H₂O kg⁻¹ dry air and the air changes 0 and 0.5 h⁻¹. At each of these combinations formaldehyde concentration in the air was followed to the equilibrium value from the zero level established with a ventilation of 20 air changes h⁻¹ at a humidity and temperature corresponding to the subsequent climatic conditions. This preliminary phase was established for 24 h and then the ventilation was decreased to the value in question. The formaldehyde concentration in the chamber then increased; it was followed for 5 days with four samples per day.

RESULTS

1. Field measurements

The results of the field measurements are given in Table 1. The means and variations of air temperature, humidity, ventilation and formaldehyde concentration were 22.8°C ± 14 per cent, 7.1 g water vapour kg⁻¹ dry air ± 15 per cent (41 per cent r.h.), 0.8 air change h⁻¹ ± 24 per cent and 0.62 mg formaldehyde m⁻³ ± 32 per cent respectively. The results do not follow a normal distribution, thus the range of variations is expressed as a percentage of the mean. The lowest formaldehyde concentration was 0.08 and the highest 2.25 mg m⁻³ air.

The frequency distribution of the measured formaldehyde concentrations is shown in Fig. 1. In two rooms (8 per cent) the average concentration of formaldehyde exceeded the German threshold limit (MAK) for occupational exposure, which at present is 1.2 mg m⁻³ (Henschler, 1972). In both rooms the temperature was higher than the average room temperature. For the home environment no limit exists,

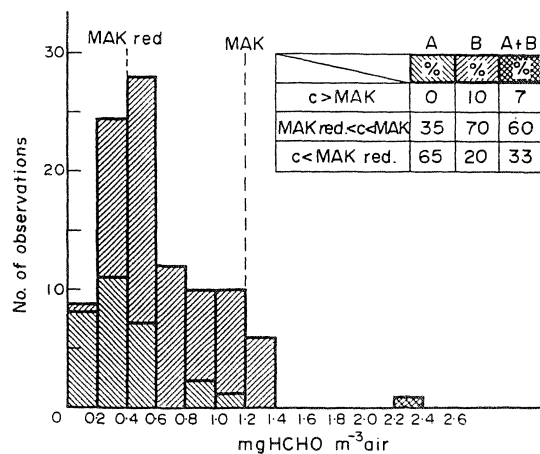


Fig. 1. The distribution of the formaldehyde concentrations in dwellings where boards were used for fixtures only (A), and where boards were used as a building material (B).

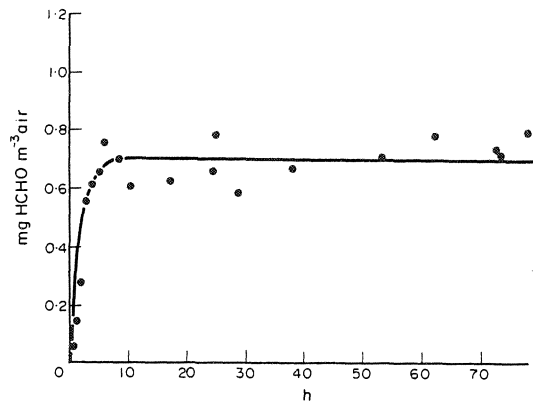


Fig. 2. Increase of formaldehyde concentration from zero to equilibrium during a climate chamber experiment. Ventilation 0.5 change h⁻¹, air temperature 22°C, humidity 5.5 g H₂O kg⁻¹ air (35 per cent r.h.).

but in comparison with the occupational value of 1.2 mg m⁻³ a reduction at least to 1/3 should take place due to the possible 24 h a day exposure in the home environment. This reduced value of 0.40 mg m⁻³ air is in this paper called MAK_{red}. The average formaldehyde concentration in the rooms was higher than this MAK_{red} in 17 rooms (68 per cent) and lower in eight rooms (32 per cent). All values were higher than the German limit for continuous exposure to outdoor air the so-called MIK value, which is 0.03 mg m⁻³ air (VDI, 1966). The measured formaldehyde concentration was correlated with the age of the house, but not with the climatic parameters. The high concentrations were only found in new houses, but all houses, even those several years old, had formaldehyde concentrations higher than the MIK value. It also appears from Fig. 1 that the formaldehyde concentration was higher in rooms where chipboard was used as a building material than in rooms where it was used only for fixtures.

2. Climate chamber measurements

Figure 2 shows the variation of formaldehyde concentration with time. After a preliminary 24-h period with ventilation of 20 air changes h⁻¹ the starting concentration is zero but in the experiment the formaldehyde concentration rapidly rose to the equilibrium concentration E_{cham} . An analysis showed that the air concentration approached exponentially to the equilibrium concentration with a time constant of 0.41 h⁻¹. The value was typical for all concentrations at ventilations larger than 0.5 air change h⁻¹. At zero ventilation the time constant was 18 h⁻¹. This time constant is an important factor for measurements under field conditions, as equilibrium concentration is achieved only after a period of five times the time constant (= 2 h) provided that constant climatic conditions have previously been established.

The results of the measurements at different climatic conditions are given in Fig. 3(a-c). From Fig. 3(a) it appears that equilibrium concentration of formaldehyde is directly proportional to temperature; a rise

Table 1. Results of the field measurements

Object No.	Age of* houses (months)	V_{rt} net volume of room (m^3)	$\alpha = \frac{P_1 + P_2}{Vn}$	Air temperature ($^{\circ}\text{C}$)		Humidity ($\text{g} (\text{kg air})^{-1}$)		Air changes (h^{-1})		HCHO-concentration ($\text{mg} (\text{m}^3 \text{air})^{-1}$)	
				No.	\bar{x}	No.	\bar{x}	No.	\bar{x}	No.	\bar{x}
1	0.3	20	1.6	7	17.1	7	6.0	4	0.5	7	0.65
2	0.3	20	1.6	4	21.4	4	4.7	2	0.5	6	0.77
3	0.3	20	1.6	2	8.6	2	4.8	2	0.4	2	0.43
4	0.3	97	1.1	3	20.1	3	4.9	2	0.9	3	0.89
5	0.3	97	1.1	1	37.8	1	9.5	1	0.8	1	2.24
6	5	14	1.7	4	26.1	4	5.6	3	1.2	4	0.33
7	5	12	1.1	3	25.4	3	5.0	2	2.3	3	0.33
8	5	14	1.7	3	22.6	3	5.9	2	0.9	3	0.60
9	5	14	1.7	3	23.3	3	6.7	2	0.3	3	0.68
10	5	14	1.7	4	23.0	4	8.0	2	1.2	5	0.71
11	5	14	1.7	3	23.8	3	6.5	2	0.6	3	0.58
12	29	14	0.4	2	21.4	2	4.8	1	1.3	2	0.08
13	29	14	0.4	3	26.8	3	4.8	2	0.9	3	0.20
14	29	14	0.4	3	31.1	3	5.4	2	1.1	3	0.26
15	29	14	0.4	1	24.4	1	4.2	1	4.6	1	0.09
16	2	16	1.3	7	26.0	7	7.3	4	0.4	7	0.67
17	0.2	16	1.3	3	25.9	3	10.2	1	0.1	3	1.29
18	0.2	16	1.3	2	19.6	2	7.7	—	—	3	0.67
19	37	22	1.8	8	21.6	8	6.6	2	0.3	10	0.49
20	42	18	1.1	5	23.3	5	9.7	1	0.2	5	1.06
21	42	18	1.8	3	25.8	3	10.7	—	—	3	1.18
22	56	21	0.3	5	22.3	5	9.4	3	0.3	6	0.42
23	54	15	0.3	5	21.6	5	7.1	2	0.6	5	0.36
24	0.2	21	1.0	5	22.1	5	8.7	2	0.4	5	0.74
25	0.2	21	1.0	4	19.4	4	8.5	2	1.4	4	0.20
Mean	15.3	23	1.2	3.7	22.8	3.7	7.1	1.9	0.8	4	0.62

* From the day central heating system functioned.

† Net volume is the volume restricted by walls, floor and ceiling exclusive closed wardrobes.

‡ α = surface area of particle boards per m^3 net volume of room. P_1 and P_2 are area of boards with no or sparse surface treatment and heavy surface treatment respectively.

direction were measured. The field measurements took place from February to September 1973, and included 25 rooms in 23 dwellings (19 houses and 4 flats), all situated in suburban areas in Jutland, Denmark. Twenty rooms were in new buildings, which were uninhabited, but ready for moving in, while five were inhabited. New houses were chosen from five different building firms, whereas some of the older houses were from the production of the same firms and some were traditional brick buildings with small amounts of chipboard. The new houses were selected at random. The only criterion was to include houses with different contents of chipboard. Chipboard was used as a construction material in 17 rooms and for fixtures only in eight rooms. Twenty-three of the measurements were in a bedroom, and in two houses one extra room was investigated. The data of the rooms are given in Table 1.

The equipment was placed in the room before the measurements. Doors and windows were closed, and only the natural ventilation through leaks and cracks remained. The radiators were set for a room temperature of 20–25°C, and the measurements were carried out in rooms without furniture. The amounts of chipboard was defined according to three categories, depending on surface treatment. Closed cupboards and wardrobes were not included in the room volume, and only their free outer surfaces were included in the area. The room was left closed overnight to reach equilibrium between emanation of formaldehyde and removal by the natural ventilation in the room.

The measurements under controlled conditions took place in the climate chamber at the Institute of Hygiene, Aarhus, Denmark. The chamber volume is 48 m^3 and all surfaces are aluminium, which does not react with formaldehyde (Andersen and Lundqvist, 1970). Air was supplied to the chamber through a double system of filters, absolute filters and charcoal filters. No particles could be detected in the air supplied to the chamber, and recirculation was never used. Fourteen boards from the normal production line of a Danish factory were placed in the room. They were made with urea-formaldehyde glue, and measured 1220 × 2440 mm and 19 mm thick. Their total weight was 523 kg and surface area 81 m^2 , equal to the total inner surface of the climate chamber. The boards were set up vertically with 25 cm between each board. Two fans ensured uniform mixing of the air in the chamber.

The emanation of formaldehyde from the boards was determined under nine different climatic conditions, selected among the 48 possible combinations of the air temperatures 17, 22, 27 and 32°C, water vapour contents 5.5, 9.5, 13.0 $\text{g H}_2\text{O kg}^{-1}$ dry air, and 0, 0.5, 1 and 3 air changes per hour. The sequence of the different combinations was arranged so that only one parameter was changed when changing from one combination to the next. Nine combinations were arranged in four groups, each with only one varying parameter. The study of temperature variations included temperatures of 17, 22, 27 and 32°C, all at a humidity of 5.5 $\text{g H}_2\text{O kg}^{-1}$ dry air and 0.5 air change h^{-1} . Study of the humidity variations in-

which expresses the formaldehyde concentration if m_f is a constant in T , H and n . This expression has been used by Stöger (1965) and Neusser and Zentner (1968), but m_f is not constant, for which reason the model must contain more detailed information about the conditions inside the board.

Analysis of the investigations mentioned above, suggested the following formula to predict the room air concentration of formaldehyde due to emanation from the surface of the board to the air at given circumstances to be:

$$E = \frac{(RT + S)(aH + b)}{1 + (nc/\alpha)} \text{ mg HCHO m}^{-3} \text{ air}, \quad (2)$$

where $\alpha = AV^{-1}$; ratio between volume of room and free area of boards in room

H = humidity g H₂O kg⁻¹ dry air

n = air change rate per hour

T = temperature °C,

and where

a , b , c , R and S are constants depending on the board under consideration determined from the experimentally formed slope of the function $E = E(T, H, n)$ in the controlled environment.

The model in the form of expression (2) indicates that E is a linear function increasing with T and H , while $1/E$ is a linear function increasing with n . The slope for $E(T)$ and $E(H)$ found in the chamber experiments, as indicated by the lines on Fig. 3(a and b), is in agreement with this. In the case of $1/E(n)$ (Fig. 3c), the linear relation is valid only for ventilation exceeding 0.4 air change h⁻¹.

The constants a , b , c , R and S are primarily deduced from the experimental data expressed through the regression lines. These zero-order values for the constants are inserted in (2), which then is used for correction of the original data. Based on these corrected data a new calculation (first order) has been carried out.

The model was tested in each order, the hypothesis was:

$$E_{\text{cham}} = E_{\text{theor}}, \quad (3)$$

where E_{cham} is the experimental value and E_{theor} is the corresponding value according to the model. The coefficients of regression, the slope and the intersection with the ordinate were 0.95, 0.82 and 0.09 respectively in the zero-order expression; 0.94, 1.03 and 0.05 respectively in the second-order expression. The second-order version of expression (2) is chosen and has the following form:

$$E(T, H, n) = \frac{(0.143H + 0.048)(0.080T - 0.764)}{1 + (n0.304/\alpha)}. \quad (4)$$

This expression is strictly only valid for the conditions mentioned in the chamber because (a) the expression does not include variation with time and uncontrolled systems are seldom in a state of equilibrium, (b) air and wall temperatures are normally different, (c) the history and surface treatment of boards will differ from those used in the climate chamber. These and other limitations will be described below.

THE MODEL USED ON THE FIELD MEASUREMENTS

One hundred measurements of formaldehyde concentrations were made in 24 rooms. Simultaneous measurements of formaldehyde concentration and ventilation was carried out in 47 cases, and 20 of these (indicated as E_{field}) had all climatic parameters in the operating range of the model. A comparison of the 20 E_{field} values and the corresponding calculated values E_{theor} according to expression (4) was carried out with a linear regression analysis. The slope of the regression line and the correlation coefficient were 0.30 and 0.33 respectively, which means that the model in the form of (4) is unable to predict

the measured formaldehyde concentration in field conditions. The reason for this was examined with a t -test on $\{E_{\text{field}}\}$ and on the errors $\{\delta = E_{\text{theor}} - E_{\text{field}}\}$. The results are shown in Table 3, which indicates that the concentrations are correlated to age, α and n , but not to T and H . These two variables influence the error δ . It is known from the climate chamber measurements that the concentration of formaldehyde in the air is a function of temperature and humidity, and the fact that this was not found in the field measurements indicates that the measurement of air temperature and humidity in this case was inadequate. Temperature and humidity in the room air varies through a day, which means that chipboard in a room will not be in thermal and hygroscopic equilibrium with room air. In order to improve the model the following procedure has been introduced. Two to seven measurements were carried out in each room, and on the basis of concentrations and parameters measured, each room has been assigned mean values of E , T , H and n : (\bar{E} , \bar{T} , \bar{H} , \bar{n}). These mean values are supposed to represent the normal situation of the room. Using these data a new value for R was calculated for each room according to the following expression, which is derived from expression (2).

$$R_{\text{corr}} = \frac{1}{\bar{T}} \left\{ \frac{1 + (\bar{n}c/\alpha)}{a\bar{H} + b} \bar{E}_0 - S \right\}, \quad (5)$$

where a , b , c and S are the constants from the chamber studies. The difference between R and the R_{corr} values are supposed to express differences between the chamber and field situations, and might, together with α , be considered as building parameters. Inserting these in (2) with a , b , c and S from the chamber measurements, an expression is formed, which includes in the mathematical model the variation of the formaldehyde concentration in a given room. This is valid for air temperature, humidity and ventilation of the same magnitude as the mean values on which R_{corr} were based.

The procedure has been tested by comparing E_{field} and the concentrations E_{corr} calculated according to expression (2) using α , R_{corr} and the measured values of T , H and n . A linear regression analysis gave $r = 0.88$. The intersection point on the Y -axis was 0.2 mg m⁻³, which, however, is not significantly different from an intersection through the origin as the S.D. of the E_{field} values was 0.28 mg m⁻³. The value $r = 0.88$ indicates that the procedure used leads to acceptable predictions of formaldehyde concentration in the rooms with climatic conditions in the range measured during the field experiments. This was confirmed by a statistical test on $\gamma = E_{\text{corr}} - E_{\text{field}}$. This test (Table 3) showed no correlation between γ and the parameters: age, α , T , H and n .

The ability of the model to reproduce the range of concentrations in Fig. 1 has been tested. The field measurements were for this purpose represented by the observed means and variations given in Table 1, together with the mean value ($\bar{R}_{\text{corr}} = 0.64 \pm 30$ per cent) of the calculated R_{corr} for each room. These values represent the normal range of climate and construction parameters. Figure 4 shows the variation of $E_{\text{theor}} = E_{\text{theor}}(\alpha)$ at different conditions. Curve I represents $E(\alpha)$ at the mean conditions (\bar{T} ,

Table 3. The influence of age, α , T , H and n on measured formaldehyde concentrations in dwellings (E_{field}), δ and γ . Values greater than 1.73 indicate a significant difference at the 95 per cent level. For definitions of δ and γ , see text

	Age	α	T	H	n
E_{field}	1.95	-1.73	1.03	0.33	2.60
δ	-0.39	-0.06	-3.24	-1.23	-0.13
γ	0.04	-0.12	-0.96	0.29	-0.67

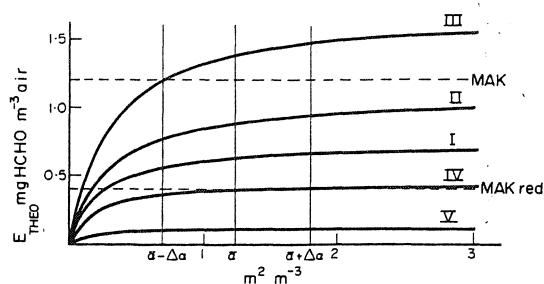


Fig. 4. The concentration of formaldehyde in air as a function of the amount of particle board α . The curves (I–V) represent different climate and construction parameters (see text).

\bar{H} , \bar{n} , \bar{R}_{corr}). It appears that $\alpha = \bar{\alpha}$ gives $E_{\text{theor}} = 0.62 \text{ mg m}^{-3}$, which is in excellent agreement with the mean of the 100 measured concentrations (0.62 mg m^{-3}), and that α in the observed interval ($\bar{\alpha} \pm \Delta\alpha$) is only affecting E_{theor} slightly. Curves III and V represent the highest and the lowest formaldehyde emission in the room at the conditions for III: ($\bar{T} + \Delta T$, $\bar{H} + \Delta H$, $\bar{n} - \Delta n$, $\bar{R}_{\text{corr}} + \Delta R$) and for V: ($\bar{T} - \Delta T$, $\bar{H} - \Delta H$, $\bar{n} + \Delta n$, $\bar{R}_{\text{corr}} - \Delta R$). It appears that α -values in the interval $[\bar{\alpha} - \Delta\alpha, \bar{\alpha} + \Delta\alpha]$ may result in formaldehyde concentrations from 0.10 to 1.46 mg m^{-3} . Ninety-five per cent of the observed concentrations were within this interval. Four per cent were below and 1 per cent higher. The interval mentioned is in excellent agreement with the frequency distribution of concentrations shown in Fig. 1.

CONCLUSIONS AND HEALTH ASPECTS

The range of formaldehyde concentrations to be expected in the "average" house has been estimated by calculating $E_{\text{theor}} = E_{\text{theor}}(\alpha)$ at the conditions: II: ($\bar{T} + \Delta T$, $\bar{H} + \Delta H$, $\bar{n} - \Delta n$, \bar{R}_{corr}) and IV: ($\bar{T} - \Delta T$, $\bar{H} - \Delta H$, $\bar{n} + \Delta n$, \bar{R}_{corr}). The resulting curves are shown in Fig. 4 as curves II and IV respectively. It appears that the normal variations in climate parameters in the same dwelling may result in variations in formaldehyde concentrations over a range of 2–2.5.

Formaldehyde may affect the human body by absorption through the skin, inhalation or by oral intake, of which the last is of no importance in the home environment. In dermatology, toxic or allergic skin infections caused by formaldehyde are often met. Formaldehyde, however, is widely used in shampoos, cosmetic deodorants, leather, paper, textiles, etc. (Walker, 1964), for which reason it is difficult to locate the releasing factor. Occupational allergy to formaldehyde is also well known, but so far it has not been possible to state a threshold for development of skin disease (Stokinger and Coffin, 1968). The literature gives no examples of skin disease caused by inhalation of formaldehyde or of formaldehyde allergy caused by a home environment with building materials, furniture, etc. made of chipboard.

The adverse health effects of formaldehyde are due to its great affinity to water (1980 vol formaldehyde vapour S.T.P./vol water) and its protein-denaturing property. The most important effect of low concentrations of formaldehyde is irritation of the mucus membranes in the nose and in the upper airways,

which occurs at concentrations higher than $2\text{--}3 \text{ mg m}^{-3}$; the long-term effects on the airways of lower concentrations are not known. The odour threshold is about 1 mg m^{-3} and conjunctivitis may occur at a threshold from 0.12 to 0.30 mg m^{-3} (Henschler, 1972). (The threshold limit values for occupational exposure and the limit values for continuous outdoor exposure to formaldehyde in U.S.A., West Germany and U.S.S.R. are given in Table 4.)

In the present study the TLV value of $1.2 \text{ mg formaldehyde m}^{-3} \text{ air}^{-1}$ was exceeded in two dwellings, and only two dwellings (0.07 and 0.08 mg m^{-3}) had lower concentrations than the American value for outdoor exposure. These dwellings were 3.5 y old and had a very low content of chipboard ($0.5 \text{ m}^2 \text{ m}^{-3}$ room). We conclude that the use of chipboard in its present form, made on the basis of a formaldehyde-urea glue, may result in higher indoor formaldehyde concentrations than permitted for continuous outdoor exposure. Either restrictions on the use of this material or a re-design of the production process to reduce the emanation of formaldehyde, therefore, should be introduced.

The present study indicates that the rather extensive system of air quality standards and control programs for workroom air and for community air should be supplemented by air quality standards and control procedures for indoor air in the home. This applies to pollutants emanating from building materials, fixtures, furniture, textiles, paints, etc. as well as to pollutants penetrating from the outside.

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INDOOR AIR POLLUTION DUE TO CHIPBOARD USED AS A CONSTRUCTION MATERIAL

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Abstract—Chipboard (particle board) is a common building construction material made of wood-shavings held together with a urea-formaldehyde glue. Due to this composition there is a continuous emanation of formaldehyde from chipboard.

Measurements in 25 rooms in 23 Danish dwellings where chipboard was used in walls, floors and ceilings showed that the average concentration was 0.62 and the range 0.08–2.24 mg formaldehyde m⁻³ air exceeding the German threshold limit for occupational exposure (1.2 mg m⁻³). In all rooms the concentration exceeded the German limit for continuous exposure in outdoor air (0.03 mg m⁻³). In climate chamber experiments the equilibrium concentration of formaldehyde from chipboard was found to be directly proportional with temperature and water vapour concentration in the air (H g H₂O kg⁻¹). A hyperbolic decrease in formaldehyde concentration occurred at increasing ventilation rates.

A mathematical model for the room air concentration of formaldehyde has been established. The model is

$$E = \frac{(RT + S)(aH + b)}{1 + (nc/x)} \text{ mg HCHO m}^{-3} \text{ air}$$

where a , b , c , S and R are constants depending on type and surface coating of chipboard. T is air temperature °C, n ventilation, air changes h⁻¹ and x area board/room volume, m² m⁻³. The constants a , b , c and S were determined on the basis of the chamber measurements, while R has to be calculated for each room from the results of measurements in the room. The model reproduces the measurements in dwellings and in the climate chamber with correlation coefficients of 0.88 and 0.94 respectively in the intervals 17–32°C, 5–13 g H₂O kg air⁻¹ and 0.4–3 air changes h⁻¹ and may be used for prediction of formaldehyde concentrations in rooms containing chipboard.

The adverse health effects of low levels of formaldehyde are irritation of the upper airways and conjunctivitis. The need for air quality standards and control programmes for indoor air in the home is stressed.

INTRODUCTION

Off-gassing properties of materials are normally regarded as a problem only in confined spaces designed for long-term stay, e.g. nuclear submarines or manned spacecraft (Hodgson and Pustinger, 1966). The problem, however, is of general hygienic importance as a multitude of airborne contaminants are being generated within the home environment from building materials, fixtures, furniture, textiles and various household products. The purpose of this paper is to analyze the emanation of formaldehyde from chipboard used as a building material. The dependence of the emanation on indoor climatic parameters is described and a mathematical model is developed for prediction purposes.

Chipboard

The use of chipboard has increased rapidly during the last decade, and the world consumption in 1970 was 1.8×10^6 m³ (1.2×10^6 tons) (FAO, 1970). In the home environment the material is used for partitions, ceilings and floors, for wardrobes and furniture. Chipboard consists of woodshavings held together by a glue, usually a urea-formaldehyde glue with an approximate molecular ratio of 1:1.4. At high pressure

and temperature the glue polymerizes. During this condensation and the subsequent hardening, which is influenced by acid-base catalysis (De Jong and De Jonge, 1952; Mellegaard, 1967) water vapour and formaldehyde are released, but 0.04 per cent free formaldehyde remains in the board, which in the course of time is released, and partly replaced by formaldehyde regenerated from the polymerized urea-formaldehyde glue.

MATERIALS AND METHODS

Identical equipment for suction of 50 l. room air through two washing bottles was used for the field measurements and the measurements in the climate chamber. The laboratory analysis was carried out with the chromotropic acid method (Nordisk Metodisk Komite, 1964; Altshüller *et al.*, 1961) with a reproducibility of ± 5 per cent at 1.0 mg formaldehyde m⁻³ air, and a detection limit of 0.1 mg m⁻³ air. The simultaneous ventilation-measurement was performed by injection of a radioactive gas (⁸⁵Kr) and after a mixing period the decrease in radioactivity was monitored for 6–8 h. Indoor climatic parameters were measured with a barograph, a thermohygrograph and a psychrometer. In addition, outdoor wind speed and

Table 1. Results of the field measurements

Object No.	Age of* houses (months)	† Net volume of room (m ³)	$\alpha = \frac{P_1 + P_2}{Vn}$	Air temperature (°C)		Humidity (g (kg air) ⁻¹)		Air changes (h ⁻¹)		HCHO-concentration (mg (m ³ air) ⁻¹)	
				No.	\bar{x}	No.	\bar{x}	No.	\bar{x}	No.	\bar{x}
1	0.3	20	1.6	7	17.1	7	6.0	4	0.5	7	0.65
2	0.3	20	1.6	4	21.4	4	4.7	2	0.5	6	0.77
3	0.3	20	1.6	2	8.6	2	4.8	2	0.4	2	0.43
4	0.3	97	1.1	3	20.1	3	4.9	2	0.9	3	0.89
5	0.3	97	1.1	1	37.8	1	9.5	1	0.8	1	2.24
6	5	14	1.7	4	26.1	4	5.6	3	1.2	4	0.33
7	5	12	1.1	3	25.4	3	5.0	2	2.3	3	0.33
8	5	14	1.7	3	22.6	3	5.9	2	0.9	3	0.60
9	5	14	1.7	3	23.3	3	6.7	2	0.3	3	0.68
10	5	14	1.7	4	23.0	4	8.0	2	1.2	5	0.71
11	5	14	1.7	3	23.8	3	6.5	2	0.6	3	0.58
12	29	14	0.4	2	21.4	2	4.8	1	1.3	2	0.08
13	29	14	0.4	3	26.8	3	4.8	2	0.9	3	0.20
14	29	14	0.4	3	31.1	3	5.4	2	1.1	3	0.26
15	29	14	0.4	1	24.4	1	4.2	1	4.6	1	0.09
16	2	16	1.3	7	26.0	7	7.3	4	0.4	7	0.67
17	0.2	16	1.3	3	25.9	3	10.2	1	0.1	3	1.29
18	0.2	16	1.3	2	19.6	2	7.7	—	—	3	0.67
19	37	22	1.8	8	21.6	8	6.6	2	0.3	10	0.49
20	42	18	1.1	5	23.3	5	9.7	1	0.2	5	1.06
21	42	18	1.8	3	25.8	3	10.7	—	—	3	1.18
22	56	21	0.3	5	22.3	5	9.4	3	0.3	6	0.42
23	54	15	0.3	5	21.6	5	7.1	2	0.6	5	0.36
24	0.2	21	1.0	5	22.1	5	8.7	2	0.4	5	0.74
25	0.2	21	1.0	4	19.4	4	8.5	2	1.4	4	0.20
Mean	15.3	23	1.2	3.7	22.8	3.7	7.1	1.9	0.8	4	0.62

* From the day central heating system functioned.

† Net volume is the volume restricted by walls, floor and ceiling exclusive closed wardrobes.

‡ = surface area of particle boards per m³ net volume of room. P_1 and P_2 are area of boards with no or sparse surface treatment and heavy surface treatment respectively.

direction were measured. The field measurements took place from February to September 1973, and included 25 rooms in 23 dwellings (19 houses and 4 flats), all situated in suburban areas in Jutland, Denmark. Twenty rooms were in new buildings, which were uninhabited, but ready for moving in, while five were inhabited. New houses were chosen from five different building firms, whereas some of the older houses were from the production of the same firms and some were traditional brick buildings with small amounts of chipboard. The new houses were selected at random. The only criterion was to include houses with different contents of chipboard. Chipboard was used as a construction material in 17 rooms and for fixtures only in eight rooms. Twenty-three of the measurements were in a bedroom, and in two houses one extra room was investigated. The data of the rooms are given in Table 1.

The equipment was placed in the room before the measurements. Doors and windows were closed, and only the natural ventilation through leaks and cracks remained. The radiators were set for a room temperature of 20–25°C, and the measurements were carried out in rooms without furniture. The amounts of chipboard was defined according to three categories, depending on surface treatment. Closed cupboards and wardrobes were not included in the room volume, and only their free outer surfaces were included in the area. The room was left closed overnight to reach equilibrium between emanation of formaldehyde and removal by the natural ventilation in the room.

The measurements under controlled conditions took place in the climate chamber at the Institute of Hygiene, Aarhus, Denmark. The chamber volume is 48 m³ and all surfaces are aluminium, which does not react with formaldehyde (Andersen and Lundqvist, 1970). Air was supplied to the chamber through a double system of filters, absolute filters and charcoal filters. No particles could be detected in the air supplied to the chamber, and recirculation was never used. Fourteen boards from the normal production line of a Danish factory were placed in the room. They were made with urea-formaldehyde glue, and measured 1220 × 2440 mm and 19 mm thick. Their total weight was 523 kg and surface area 81 m², equal to the total inner surface of the climate chamber. The boards were set up vertically with 25 cm between each board. Two fans ensured uniform mixing of the air in the chamber.

The emanation of formaldehyde from the boards was determined under nine different climatic conditions, selected among the 48 possible combinations of the air temperatures 17, 22, 27 and 32°C, water vapour contents 5.5, 9.5, 13.0 g H₂O kg⁻¹ dry air, and 0, 0.5, 1 and 3 air changes per hour. The sequence of the different combinations was arranged so that only one parameter was changed when changing from one combination to the next. Nine combinations were arranged in four groups, each with only one varying parameter. The study of temperature variations included temperatures of 17, 22, 27 and 32°C, all at a humidity of 5.5 g H₂O kg⁻¹ dry air and 0.5 air change h⁻¹. Study of the humidity variations in-

cluded 5.5, 9.5 and 13 g H₂O kg⁻¹ dry air all at 22°C and 0.5 air change h⁻¹. The study of ventilation included 0, 0.5, 1 and 3 air changes h⁻¹ all at 22°C and 5.5 g H₂O kg⁻¹ dry air, and finally the ageing of the plates was studied with an interval of 13 months; the temperature was 22°C, the humidities 5.5 and 13 g H₂O kg⁻¹ dry air and the air changes 0 and 0.5 h⁻¹. At each of these combinations formaldehyde concentration in the air was followed to the equilibrium value from the zero level established with a ventilation of 20 air changes h⁻¹ at a humidity and temperature corresponding to the subsequent climatic conditions. This preliminary phase was established for 24 h and then the ventilation was decreased to the value in question. The formaldehyde concentration in the chamber then increased; it was followed for 5 days with four samples per day.

RESULTS

1. Field measurements

The results of the field measurements are given in Table 1. The means and variations of air temperature, humidity, ventilation and formaldehyde concentration were 22.8°C ± 14 per cent, 7.1 g water vapour kg⁻¹ dry air ± 15 per cent (41 per cent r.h.), 0.8 air change h⁻¹ ± 24 per cent and 0.62 mg formaldehyde m⁻³ ± 32 per cent respectively. The results do not follow a normal distribution, thus the range of variations is expressed as a percentage of the mean. The lowest formaldehyde concentration was 0.08 and the highest 2.25 mg m⁻³ air.

The frequency distribution of the measured formaldehyde concentrations is shown in Fig. 1. In two rooms (8 per cent) the average concentration of formaldehyde exceeded the German threshold limit (MAK) for occupational exposure, which at present is 1.2 mg m⁻³ (Henschler, 1972). In both rooms the temperature was higher than the average room temperature. For the home environment no limit exists,

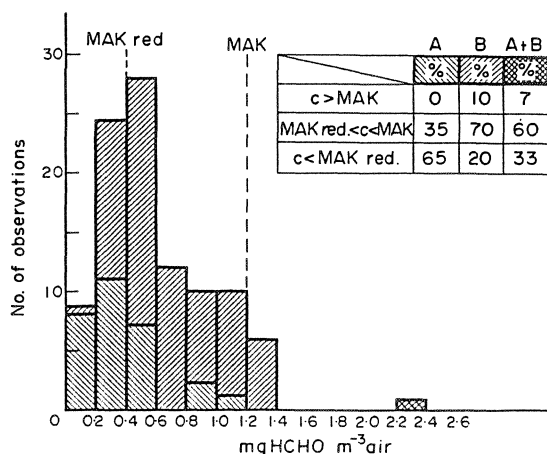


Fig. 1. The distribution of the formaldehyde concentrations in dwellings where boards were used for fixtures only (A), and where boards were used as a building material (B).

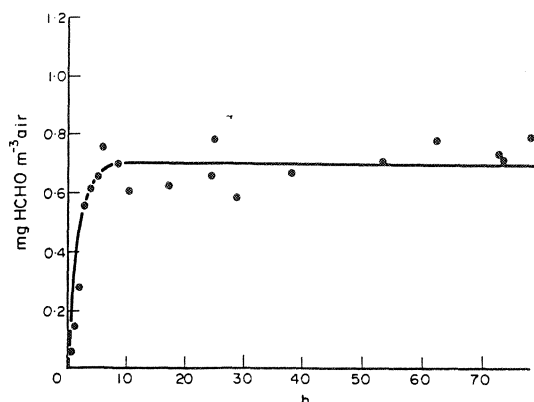


Fig. 2. Increase of formaldehyde concentration from zero to equilibrium during a climate chamber experiment. Ventilation 0.5 change h⁻¹, air temperature 22°C, humidity 5.5 g H₂O kg⁻¹ air (35 per cent r.h.).

but in comparison with the occupational value of 1.2 mg m⁻³ a reduction at least to 1/3 should take place due to the possible 24 h a day exposure in the home environment. This reduced value of 0.40 mg m⁻³ air is in this paper called MAK_{red}. The average formaldehyde concentration in the rooms was higher than this MAK_{red} in 17 rooms (68 per cent) and lower in eight rooms (32 per cent). All values were higher than the German limit for continuous exposure to outdoor air the so-called MIK value, which is 0.03 mg m⁻³ air (VDI, 1966). The measured formaldehyde concentration was correlated with the age of the house, but not with the climatic parameters. The high concentrations were only found in new houses, but all houses, even those several years old, had formaldehyde concentrations higher than the MIK value. It also appears from Fig. 1 that the formaldehyde concentration was higher in rooms where chipboard was used as a building material than in rooms where it was used only for fixtures.

2. Climate chamber measurements

Figure 2 shows the variation of formaldehyde concentration with time. After a preliminary 24-h period with ventilation of 20 air changes h⁻¹ the starting concentration is zero but in the experiment the formaldehyde concentration rapidly rose to the equilibrium concentration E_{cham} . An analysis showed that the air concentration approached exponentially to the equilibrium concentration with a time constant of 0.41 h⁻¹. The value was typical for all concentrations at ventilations larger than 0.5 air change h⁻¹. At zero ventilation the time constant was 18 h⁻¹. This time constant is an important factor for measurements under field conditions, as equilibrium concentration is achieved only after a period of five times the time constant (= 2 h) provided that constant climate conditions have previously been established.

The results of the measurements at different climatic conditions are given in Fig. 3(a-c). From Fig. 3(a) it appears that equilibrium concentration of formaldehyde is directly proportional to temperature; a rise

which expresses the formaldehyde concentration if m_f is a constant in T , H and n . This expression has been used by Stöger (1965) and Neusser and Zentner (1968), but m_f is not constant, for which reason the model must contain more detailed information about the conditions inside the board.

Analysis of the investigations mentioned above, suggested the following formula to predict the room air concentration of formaldehyde due to emanation from the surface of the board to the air at given circumstances to be:

$$E = \frac{(RT + S)(aH + b)}{1 + (nc/x)} \text{ mg HCHO m}^{-3} \text{ air}, \quad (2)$$

where $\alpha = AV^{-1}$; ratio between volume of room and free area of boards in room

H = humidity g H₂O kg⁻¹ dry air

n = air change rate per hour

T = temperature °C,

and where

a , b , c , R and S are constants depending on the board under consideration determined from the experimentally formed slope of the function $E = E(T, H, n)$ in the controlled environment.

The model in the form of expression (2) indicates that E is a linear function increasing with T and H , while $1/E$ is a linear function increasing with n . The slope for $E(T)$ and $E(H)$ found in the chamber experiments, as indicated by the lines on Fig. 3(a and b), is in agreement with this. In the case of $1/E(n)$ (Fig. 3c), the linear relation is valid only for ventilation exceeding 0.4 air change h⁻¹.

The constants a , b , c , R and S are primarily deduced from the experimental data expressed through the regression lines. These zero-order values for the constants are inserted in (2), which then is used for correction of the original data. Based on these corrected data a new calculation (first order) has been carried out.

The model was tested in each order, the hypothesis was:

$$E_{\text{cham}} = E_{\text{theor}}, \quad (3)$$

where E_{cham} is the experimental value and E_{theor} is the corresponding value according to the model. The coefficients of regression, the slope and the intersection with the ordinate were 0.95, 0.82 and 0.09 respectively in the zero-order expression; 0.94, 1.03 and 0.05 respectively in the second-order expression. The second-order version of expression (2) is chosen and has the following form:

$$E(T, H, n) = \frac{(0.143H + 0.048)(0.080T - 0.764)}{1 + (n0.304/x)}. \quad (4)$$

This expression is strictly only valid for the conditions mentioned in the chamber because (a) the expression does not include variation with time and uncontrolled systems are seldom in a state of equilibrium, (b) air and wall temperatures are normally different, (c) the history and surface treatment of boards will differ from those used in the climate chamber. These and other limitations will be described below.

THE MODEL USED ON THE FIELD MEASUREMENTS

One hundred measurements of formaldehyde concentrations were made in 24 rooms. Simultaneous measurements of formaldehyde concentration and ventilation was carried out in 47 cases, and 20 of these (indicated as E_{field}) had all climatic parameters in the operating range of the model. A comparison of the 20 E_{field} values and the corresponding calculated values E_{theor} according to expression (4) was carried out with a linear regression analysis. The slope of the regression line and the correlation coefficient were 0.30 and 0.33 respectively, which means that the model in the form of (4) is unable to predict

the measured formaldehyde concentration in field conditions. The reason for this was examined with a t -test on $\{E_{\text{field}}\}$ and on the errors $\{\delta = E_{\text{theor}} - E_{\text{field}}\}$. The results are shown in Table 3, which indicates that the concentrations are correlated to age, α and n , but not to T and H . These two variables influence the error δ . It is known from the climate chamber measurements that the concentration of formaldehyde in the air is a function of temperature and humidity, and the fact that this was not found in the field measurements indicates that the measurement of air temperature and humidity in this case was inadequate. Temperature and humidity in the room air varies through a day, which means that chipboard in a room will not be in thermal and hygroscopic equilibrium with room air. In order to improve the model the following procedure has been introduced. Two to seven measurements were carried out in each room, and on the basis of concentrations and parameters measured, each room has been assigned mean values of E , T , H and n : (\bar{E} , \bar{T} , \bar{H} , \bar{n}). These mean values are supposed to represent the normal situation of the room. Using these data a new value for R was calculated for each room according to the following expression, which is derived from expression (2).

$$R_{\text{corr}} = \frac{1}{\bar{T}} \left\{ 1 + \frac{(nc/x)\bar{E}_0}{a\bar{H} + b} - S \right\}, \quad (5)$$

where a , b , c and S are the constants from the chamber studies. The difference between R and the R_{corr} values are supposed to express differences between the chamber and field situations, and might, together with α , be considered as building parameters. Inserting these in (2) with a , b , c and S from the chamber measurements, an expression is formed, which includes in the mathematical model the variation of the formaldehyde concentration in a given room. This is valid for air temperature, humidity and ventilation of the same magnitude as the mean values on which R_{corr} were based.

The procedure has been tested by comparing E_{field} and the concentrations E_{corr} calculated according to expression (2) using α , R_{corr} and the measured values of T , H and n . A linear regression analysis gave $r = 0.88$. The intersection point on the Y -axis was 0.2 mg m⁻³, which, however, is not significantly different from an intersection through the origin as the S.D. of the E_{field} values was 0.28 mg m⁻³. The value $r = 0.88$ indicates that the procedure used leads to acceptable predictions of formaldehyde concentration in the rooms with climatic conditions in the range measured during the field experiments. This was confirmed by a statistical test on $\gamma = E_{\text{corr}} - E_{\text{field}}$. This test (Table 3) showed no correlation between γ and the parameters: age, α , T , H and n .

The ability of the model to reproduce the range of concentrations in Fig. 1 has been tested. The field measurements were for this purpose represented by the observed means and variations given in Table 1, together with the mean value ($\bar{R}_{\text{corr}} = 0.64 \pm 30$ per cent) of the calculated R_{corr} for each room. These values represent the normal range of climate and construction parameters. Figure 4 shows the variation of $E_{\text{theor}} = E_{\text{theor}}(x)$ at different conditions. Curve 1 represents $E(x)$ at the mean conditions (\bar{T} ,

Table 3. The influence of age, α , T , H and n on measured formaldehyde concentrations in dwellings (E_{field}), δ and γ . Values greater than 1.73 indicate a significant difference at the 95 per cent level. For definitions of δ and γ , see text

	Age	α	T	H	n
E_{field}	1.95	-1.73	1.03	0.33	2.60
δ	-0.39	-0.06	-3.24	-1.23	-0.13
γ	0.04	-0.12	-0.96	0.29	-0.67

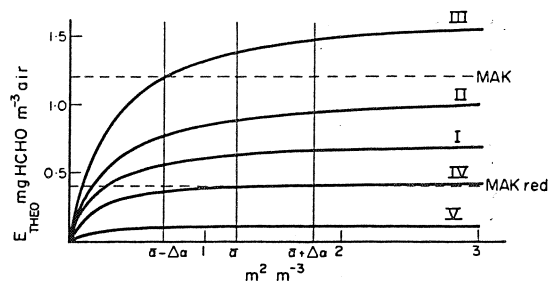


Fig. 4. The concentration of formaldehyde in air as a function of the amount of particle board α . The curves (I–V) represent different climate and construction parameters (see text).

\bar{H} , \bar{n} , \bar{R}_{corr}). It appears that $\alpha = \bar{\alpha}$ gives $E_{theor} = 0.62 \text{ mg m}^{-3}$, which is in excellent agreement with the mean of the 100 measured concentrations (0.62 mg m^{-3}), and that α in the observed interval ($\bar{\alpha} \pm \Delta\alpha$) is only affecting E_{theor} slightly. Curves III and V represent the highest and the lowest formaldehyde emission in the room at the conditions for III: ($\bar{T} + \Delta T$, $\bar{H} + \Delta H$, $\bar{n} - \Delta n$, $\bar{R}_{corr} + \Delta R$) and for V: ($\bar{T} - \Delta T$, $\bar{H} - \Delta H$, $\bar{n} + \Delta n$, $\bar{R}_{corr} - \Delta R$). It appears that α -values in the interval $[\bar{\alpha} - \Delta\alpha, \bar{\alpha} + \Delta\alpha]$ may result in formaldehyde concentrations from 0.10 to 1.46 mg m^{-3} . Ninety-five per cent of the observed concentrations were within this interval. Four per cent were below and 1 per cent higher. The interval mentioned is in excellent agreement with the frequency distribution of concentrations shown in Fig. 1.

CONCLUSIONS AND HEALTH ASPECTS

The range of formaldehyde concentrations to be expected in the "average" house has been estimated by calculating $E_{theor} = E_{theor}(\alpha)$ at the conditions: II: ($\bar{T} + \Delta T$, $\bar{H} + \Delta H$, $\bar{n} - \Delta n$, \bar{R}_{corr}) and IV: ($\bar{T} - \Delta T$, $\bar{H} - \Delta H$, $\bar{n} + \Delta n$, \bar{R}_{corr}). The resulting curves are shown in Fig. 4 as curves II and IV respectively. It appears that the normal variations in climate parameters in the same dwelling may result in variations in formaldehyde concentrations over a range of 2–2.5.

Formaldehyde may affect the human body by absorption through the skin, inhalation or by oral intake, of which the last is of no importance in the home environment. In dermatology, toxic or allergic skin infections caused by formaldehyde are often met. Formaldehyde, however, is widely used in shampoos, cosmetic deodorants, leather, paper, textiles, etc. (Walker, 1964), for which reason it is difficult to locate the releasing factor. Occupational allergy to formaldehyde is also well known, but so far it has not been possible to state a threshold for development of skin disease (Stokinger and Coffin, 1968). The literature gives no examples of skin disease caused by inhalation of formaldehyde or of formaldehyde allergy caused by a home environment with building materials, furniture, etc. made of chipboard.

The adverse health effects of formaldehyde are due to its great affinity to water (1980 vol formaldehyde vapour S.T.P./vol water) and its protein-denaturing property. The most important effect of low concentrations of formaldehyde is irritation of the mucus membranes in the nose and in the upper airways,

which occurs at concentrations higher than 2–3 mg m^{-3} ; the long-term effects on the airways of lower concentrations are not known. The odour threshold is about 1 mg m^{-3} and conjunctivitis may occur at a threshold from 0.12 to 0.30 mg m^{-3} (Henschler, 1972). (The threshold limit values for occupational exposure and the limit values for continuous outdoor exposure to formaldehyde in U.S.A., West Germany and U.S.S.R. are given in Table 4.)

In the present study the TLV value of $1.2 \text{ mg formaldehyde m}^{-3} \text{ air}^{-1}$ was exceeded in two dwellings, and only two dwellings (0.07 and 0.08 mg m^{-3}) had lower concentrations than the American value for outdoor exposure. These dwellings were 3.5 y old and had a very low content of chipboard ($0.5 \text{ m}^2 \text{ m}^{-3}$ room). We conclude that the use of chipboard in its present form, made on the basis of a formaldehyde-urea glue, may result in higher indoor formaldehyde concentrations than permitted for continuous outdoor exposure. Either restrictions on the use of this material or a re-design of the production process to reduce the emanation of formaldehyde, therefore, should be introduced.

The present study indicates that the rather extensive system of air quality standards and control programs for workroom air and for community air should be supplemented by air quality standards and control procedures for indoor air in the home. This applies to pollutants emanating from building materials, fixtures, furniture, textiles, paints, etc. as well as to pollutants penetrating from the outside.

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