

The effects of temperature and humidity on formaldehyde emission from UF-bonded boards: a literature critique

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Abstract

An analysis has been conducted on available data related to temperature and humidity effects on formaldehyde concentrations that are produced by emission from particleboard and hardwood plywood paneling. Temperature changes are described by an exponential relation while a linear relation suffices for humidity effects. Large variations exist in the results from different investigators on different boards for the exponential temperature coefficients B (-5620° to $-12,480^\circ\text{K}^{-1}$) and for the linear humidity coefficients β (0.005% to 0.038% $^{-1}$). The variations appear to be caused by experimental errors, procedural differences between laboratories, board-to-board differences, and board aging. Statistical treatment of all chamber test data as a composite set (normalized to unity at 25°C) yields a composite B of -8930°K^{-1} , with a 95 percent confidence interval from -8390° to -9470°K^{-1} ($\pm 6\%$ relative error). Using this composite B , calculated correction factors, $c(25^\circ\text{C})/c(T)$, varied from 4.89 at 10°C to 0.152 at 45°C. The statistical analysis also indicates that significant board-to-board and laboratory-to-laboratory variations occurred. Assuming this composite coefficient is indeed valid for a particular board or dwelling, it can be used to make temperature corrections to standard temperature with, for example, relative errors (95% confidence) in correction factors being $\pm 7\%$ percent for a $\pm 10^\circ\text{C}$ temperature difference. In the absence of knowledge about the specific applicability of a particular temperature coefficient, however, temperature corrections should probably be restricted to a 5°C range, except for general engineering or planning purposes. A similar composite analysis of all the humidity data (normalized to unity at 50% relative humidity (RH)) yields a composite β of 0.0195% $^{-1}$ with a 95 percent confidence interval from 0.014 to 0.025 ($\pm 28\%$ relative error). The corresponding correction factors, $c(50\%)/c(\text{RH})$, vary from 2.41 at 20 percent RH to 0.56 at 90 percent RH. Very large variations between investigators, and between boards, lead to poor confidence in such composite universal humidity correction factors, however, and no clear recommendation can be made

regarding correction factors to convert formaldehyde concentrations to a standard humidity. Small humidity changes ($\sim 10\%$ RH), however, probably can be neglected since they may be masked by rather minor temperature changes ($\pm 1^\circ$ to 2°C). Much more information is needed to clarify the reasons for the large variability and to increase confidence in temperature and humidity correction factors.

This is the fourth in a planned series of six critical reviews of the literature on different aspects of the problem of formaldehyde emission from adhesively bonded wood products. The series was initiated because of the need expressed by industry representatives for an independent evaluation and summation of data from a wide variety of studies. The six aspects being reviewed are concerned with effects of 1) formaldehyde-to-urea mole ratio (25), 2) ventilation rate and loading (26), 3) additions to wood furnish (28), 4) temperature and humidity, 5) post-manufacture treatments of boards, and 6) hydrolysis.

This paper analyzes the available data on effects of changing temperature or humidity (RH) on formaldehyde emission from particleboard and hardwood plywood paneling. As with the first three reviews (25, 26, 28), it is based upon a bibliography (27) derived from several sources (25), in this case covering the period from 1960 through May 1984.¹

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In undertaking this review of temperature and humidity effects, I considered it important to attempt to answer several questions:

- 1) What is the quality of the individual and the overall data bases?
- 2) What mathematical models reasonably describe the reported temperature and humidity data?
- 3) What is the variability in observed temperature and humidity coefficients or correction factors? Can that variability be related to differences such as product types (particleboard versus plywood), emission ranges (low versus high), or experimental procedures?
- 4) What differences exist between coefficients determined from laboratory tests on boards, and from measurements in actual living spaces?
- 5) What recommendations can be developed for usable coefficients or for additional work?

General comments

The available results on the influence of temperature and humidity are summarized in Tables 1 and 2. Quantitative analyses of the effects of these two variables will be presented separately since the uncertainties in the humidity data are so large and since, with the exception of results from one investigating group (10, 11), no interaction between the variables has been reported. First, however, some general comments about the nature of the studies are in order:

—The majority of the tests may be classified (Columns 2) as chamber types—ventilated or unventilated. A few (13, 20, 33, 34) are water sorption types (desiccator, WKI), while only two (7, 12) involve measurements of temperature effects in homes where, unfortunately, other conditions are poorly specified.

—Many of the studies reported data for only one or a few different boards (Columns 3); notable exceptions are those by Liiri, et al. (14) on particleboard (Table 2) and by Couch (7) on mobile homes (Table 1). Several investigators (9-11, 15, 38, 39) did not specify the numbers of boards that had been examined.

—Experimental details and range of variables are not always clearly stated (Columns 4-7). Where specified, temperature (Columns 4) and humidity (Columns 5) ranges are approximately 20° to 40°C and 20 to 90 percent, respectively. These are consistent with the usual ranges of interest for dwellings.

—Several reports did not supply data (Columns 8) but only made statements about an observed quantitative dependence upon temperature or humidity (7, 9-11, 13, 15, 38, 39). How extensive the data base underlying these statements may be is not always apparent. Nevertheless, I have deemed it improper to ignore such reported coefficients, particularly as the data base in some cases seemed likely to be relatively large (e.g., 9, 10, 39).

—Chamber formaldehyde concentrations can be very slow in achieving a new equilibrium or steady state after a humidity change (6, 14, 17). Consequently, reported chamber concentrations at different humidities may not represent steady state or equilibrium conditions. Although board emissions respond more quickly to temperature change, it is also not always apparent that such experiments have achieved steady state or equilibrium. Additionally, the possibility is very real that the formaldehyde supply within a board will be depleted during a test series (19, 21). Hence, I have labeled all chamber concentrations (Columns 8) apparent steady state or apparent equilibrium concentrations.

Temperature dependence

Data summary and analysis model

The listing in Table 1 is in order first of test type (Column 2)—e.g., chamber, home, water sorption. Within each test type, particleboard (PB) results preceded the less extensive results for hardwood plywood (PLY).

Some investigators (1, 16) have described the temperature dependence of formaldehyde emission in linear terms. More usually, however, an exponential dependence has been assumed and I have followed the latter practice by setting:

$$c \left(T, RH, \frac{N}{L} \right) = c^{\circ} (298^{\circ}\text{K}, RH, \frac{N}{L}) \exp \left[B \left(\frac{1}{T} - \frac{1}{298} \right) \right] \quad [1]$$

Here, T is temperature in °K, concentrations c are referred to 25°C as a standard, N is ventilation rate in hr.⁻¹, and L is board loading in m²/m³.

Although I have not carried out a statistical comparison to establish the optimum analytical expression, the selection of Equation [1] is warranted on several bases:

1) It has been the most widely used for this purpose and a number of laboratories have reported only values of the coefficient B , without data.

2) As Figure 1 and subsequent discussion show, a given data set covering a small temperature interval might reasonably be fit by a linear expression. Over a broader temperature range, however, any description must be strongly nonlinear.

3) Formaldehyde test values (chamber concentration, desiccator values, etc.) directly reflect a rate of formaldehyde emission from boards. The temperature dependence of both physical and chemical rate processes have been described traditionally and theoretically (Arrhenius equation) by exponential relations.

The weighting factors (Column 10) represent my somewhat arbitrary judgments about the relative merit (higher number = greater merit) attributable to each value of B . The factors were derived by multiplying separate weighting factors for test type (chamber > water sorption > home > elution from ground board), number of boards underlying the B value, number of temperatures used (emphasizing 15° to 40°C), and RH control. The Column 10 values were used in portions of the following statistical analyses of the variability in B .

¹Copies of the bibliographies (22-24, 27) may be requested from the Formaldehyde Institute, 1076 Central Park Ave., Scarsdale, NY 10583, after publication of the respective critiques.

TABLE 1. — Temperature effects.

Laboratory (Ref.) 1	Test 2	Material/No. ^a 3	Test conditions				Result ^b 8	Temperature coefficient, -B(°K ⁻¹) ^c 9	Weighting factor, ^d w 10
			Temp. (°C) 4	RH (%) 5	N/L(m/hr.) 6	Duration ^e 7			
Andersen, et al. (1)	Chamber	PB/1	17, 22, 27, 32	~20 to 40	0.3	5 d.	Apparent steady state values fit to: $c_s(RH, t, N/L) = (0.080t - 0.764) \cdot f(RH) \cdot g(N/L)$	5,620	4
Berge & Mellegaard (3)	Chamber	PB/1	21, 25, 30, 35	Controlled by board MC	0	3 hr.	Apparent equilibrium values given	10,150	3
Berge, et al. (4)	Chamber	PB/2	22, 28	30, 60	0.13, 0.31	7 d.	Apparent c_s values from partial factorial	9,800	4
Hanetho (9) and Sundin (38, 39)	Chamber	PB/?			0	1-2 d.	No data. Statement that 7°C ΔT doubles c_{eq}	8,780	5 each lab
Hoetjer (10) and Hoetjer & Koerts (11)	Chamber	PB/?			Range	hrs.	No data. Statement that c_{eq} increases 11% per °C.	9,290	5
Lundquist (15)	Chamber	PB/?	20-35		0		No data. Statement that 20° to 35°C increases c by 3-4 fold.	7,540	5
Myers & Nagaoka (29)	Chamber	PB/2	25, 40	75	0.06	days to wks	Apparent c_s values given	8,620; 8,740	3 each
Wittman (40)	Chamber	PB/1	40, 50, 60	50	0	2 hr.	Apparent c_{eq} values given	11,120	1
Matthews, et al. (18, 19)	Chamber	PB/3	23, 28, 38	50-80	Range	hrs.	Exponential coefficients given for fit of temperature, RH, N/L data to	8,280; 9,660; 8,080	3 each
	Chamber	PLY/2	23, 28, 38	50-80	Range	hrs.	Equation [A1]. ^f	7,430; 8,230	
	Chamber	PLY/1	23, 28, 33	50-80	Range	hrs.		8,190	
	Chamber	PLY/1	5 values, 22-30	4 values, 44-80	~0.1	5 d.	Apparent steady state values fit to: $c_s(t, RH) = c(30^\circ\text{C}, 80\%)[1 - 0.057(30 - t)] \cdot f(RH)$	7,530	4
Newton (30)	Chamber (equil. jar)	PB/3	6 values, 21-49	Controlled by board MC	0	?	Apparent equilibrium values given	8,660; 8,670; 10,020	3 each
Couch (7)	Chamber	PLY/2 ("low odor")	25, 35	60	0.5	6 hr.	No data. Exponential coefficient given	8,550; 9,600	3 each
Stöger (37)	Chamber	(PB + PLY)/1 ("low odor")					No data. Exponential coefficient given	8,800	3
	Elute ground PB	PB/1	20, 50, 70			hrs.	Initial evolution rate given	6,580	1
Cox & Moberly (8)	Chamber, total eluted CH ₂ O	UF coatings on wood	32, 43, 54, 66		0.35	2 hr.	Values for total eluted CH ₂ O given	8,170	1
Couch (7)	Air conc.	Mobile home/26					No data. Exponential coefficient given	7,520	4
Jewell (12)	Air conc.	Mobile home/1	15, 17, 24, 33			2 d. total	Air conc. values given	7,800	2
McVey (20)	Modified desiccator ^g	PB/1	12, 15, 17, 20, 22, 25	~100	0	2 hr.	Non-steady state values given	8,210	3
Newton (31)	Desiccator ^g	PB/2	23, 29, 37	~100	0	2 hr.	Non-steady state values given	8,630; 9,340	2 each
New Zealand (32)	Desiccator ^g	PB/1	8 values, 12-25	~100	0	2 hr.	Non-steady state values given	8,920	3
Roffael (33)	WKI ^h	PB/2	20, 40	~100	0	48 hr.	Non-steady state values given	6,970; 7,170	1 each
Roffael (34)	WKI ^h	PB/1	20, 30, 40	~100	0	48 hr.	Non-steady state values given	6,770	2
Rybicky, et al. (36)	Desiccator ^g	PB/3	15, 25, 35, 43	~100	0	2 hr.	Non-steady state values given	8,220; 7,910; 6,140	2 each
Kubota, et al. (13)	Modified desiccator ^g	PLY/4	22, 31	~100	0	24 hr.	No data. Statement that 22° to 31°C increases desiccator value ~3.5 ×	12,480	4

^aPB = particleboard. PLY = hardwood plywood paneling. Number indicates the apparent number of different boards which were tested. ? indicates number unspecified in original reference.

^bN/L = ventilation rate over board loading (26). c_s and c_{eq} = apparent steady state and equilibrium concentration of formaldehyde in chamber air. t = °C. f and g are functions specified in the reference.

^cB from linear regression of logarithmic form of Equation [1].

^dBased on test type (chamber > water sorption > home > elution), number of boards, number of temperatures (emphasis on 15° to 40°C), and RH control.

^eTime at each condition.

^fFrom authors' (18, 19) preliminary analysis.

^gTest involves vapor phase transfer of formaldehyde from small board specimens into water.

TABLE 2. — Humidity effects.^a

Laboratory (Ref.) 1	Test 2	Material/No. 3	Test conditions				Reported result 8	Humidity coefficient, $\beta(RH^{-1})^b$ 9	Weighting factor, ^c w 10
			Temp. (°C) 4	RH (%) 5	N/L (m/hr.) 6	Duration 7			
Liiri, et al. (14)	Chamber	PB, coated, uncoated, NH ₃ treated/~50	20	35, 85 cycles	~30	4 wk.	Seldom achieved steady state. Some initial transient spikes. Values at given RH may differ from cycle to cycle.	0.008 to 0.038 ^d	3 each of 9
Myers & Nagaoka (29)	Chamber	PB/2	25	25, 75	0.06	>1 wk.	Apparent steady values after initial transients.	0.0088, 0.0098	2 each
Berge, et al. (4)	Chamber	PB/2	22, 28	30, 60	0.1, 0.3	7 d.	Apparent steady state values from partial factorial fit to: $c_d(RH, T, N/L) = c_{eq}(60\%, 22^\circ\text{C})[1 + 0.0175(RH - 60)] \cdot f(T) \cdot g(N/L)^e$	0.024	4
Andersen, et al. (1)	Chamber	PB/1	17, 22, 27, 32	32, 39, 55, 68, 75	0.3	5 d.	Apparent steady state values fit to: $\alpha(RH, T, N/L) = (0.143H + 0.048) \cdot f(T) \cdot g(N/L)^{e,*}$	0.015	4
Bourne (5)	Chamber	PB/1	32	44, 68, 87	0.15	4-7 d.	Apparent steady state values fit to: $c = 1.50 + 0.0110 RH$	0.0047	3
Sundin (38)	Chamber	PB/?	20	30, 70	0	1-2 d.	No data. Statement that c_{eq} doubled between 30 and 70% RH	0.017	5
Sundin (39)	Chamber	PB/?	20	30, 90	0	1-2 d.	No data. Statement that c_{eq} doubled between 30 and 90% RH	0.012	5
Hoetjer (10) and Hoetjer & Koerts (11)	Chamber	PB/?				hrs.?	No data. $c_d(RH, T, N/L) = c_{eq}(RH, T)[1 + \frac{N/L}{K(RH)}]$ Correction table for $c_{eq}(RH, T)$ & $K(RH)$.	^f 0.024 ^g 0.021	5
Matthews, et al. (17, 18)	Chamber	PB/1	23	35, 50, 75	140	2 hr.	Non-steady state values	0.014	2
Matthews, et al. (19)	Chamber	PB/3	23, 28, 38	50, 60, 67, 80	Range	hrs.	Coefficients to Equation [A1] given from fit of temperature, RH, N/L data ^h	0.0186, 0.0060, 0.0370	2 each
Wittman (40)	Chamber	PB/1	70	35, 45, 50, 60, 70, 80	0	2 hr.	Presumably non-steady state values	0.0089	3
Matsumoto (16)	Chamber	PLY/1	5 values, 22-30	4 values, 44-80	~0.1	5 d.	$\alpha(RH, T) = c^*(30^\circ\text{C}, 80\%)[1 - 0.008(80 - RH)] \cdot f(T)^a$	0.0104	4
Matthews, et al. (19)	Chamber	PLY/3	23, 28, 38	50, 60, 67, 80	Range	hrs.	Coefficients to Equation [A1] given from fit of temperature, RH, N/L data	0.0064, 0.0330, 0.0095	2 each
Couch (6, 7)	Chamber	PLY/1	30	48, 80	~0.7	6 hr.	Non-steady state values	0.0059	1
Rudenko & Birjukova (35)	Chamber	UF mineral wood panels/2	40	20, 30, 40, 50, 60	$N = 1 \text{ hr.}^{-1}$	5-15 d.	Apparent steady state values	0.028, 0.020	3 each
Roffael (34)	WKI	PB/1	40	30, 75	0	2 d. 12 d.	Non-steady state values Non-steady state values	0.012 0.022	2

^aSee Table 1 for definition of terms not defined here.^b β from linear regression of Equation [2].^cBased on test type (chamber > WKI); number of boards; duration (≥ 1 day higher than <1 day), number of RH.^dCalculated for only 9 of board data sets.^e $H = gH_2O/\text{kg air}$.^fAssuming $K(46\%) = 0.1 \text{ m/hr.}$ from N/L 0.5 to 1.5 m/hr. (9, 10).^gAssuming $K(46\%) = 1.0 \text{ m/hr.}$ from N/L 0.5 to 1.5 m/hr. (9, 10).^hFrom authors' (19) preliminary analysis.

Variability in temperature Coefficients

The total range in B is over twofold, -5620 to $-12,480^\circ\text{K}^{-1}$, and is only slightly less when restricted to chamber tests, ~ -5600 to $\sim -11,100^\circ\text{K}^{-1}$. Interestingly, however, this twofold range is far less than that observed (~ 300 -fold) from various laboratories and boards for coefficients describing the dependence of

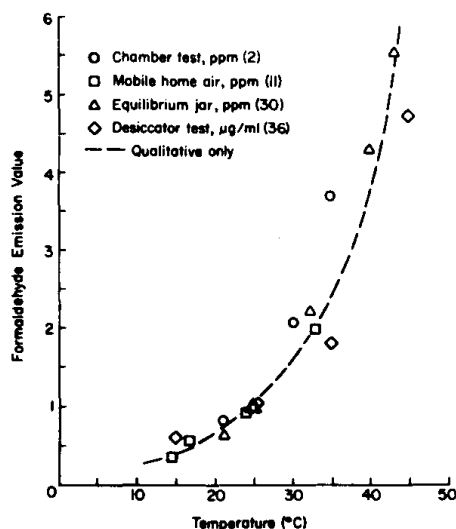


Figure 1. — illustration of non-linear temperature dependence.

formaldehyde concentration on ventilation rate and loading (26).

To obtain at least semiquantitative estimates of the variability in B and its causes, I have carried out several statistical treatments, yielding various mean B values and associated uncertainties listed in Table 3 (Columns 6 and 7). The table also includes the result of an approximate error analysis for chamber testing (Appendix). In considering the following discussion of Table 3 it must be kept in mind that the statistical analyses are only semiquantitative due to the grouping of data from different laboratories, boards, tests, etc.

Group I. — Mean B values were derived by averaging different subsets of the observed B 's from Table 1. Because standard deviations have dubious meaning here, the uncertainties are given as the observed ranges. (Nominal standard deviations for sets 2 and 3, for example, are 1370 and 1180, respectively, corresponding coefficients of variation being 16% and 14%). The various means for sets 1 to 6 are surprisingly close, and it does not seem possible to attribute any significance to their differences.

Group II. — Each investigator's formaldehyde test data were first normalized to 25°C , i.e., $c(T)/c(298)$ ratios were calculated. Then, different subsets of those normalized values were each treated as composite data sets and regressed against Equation [1]; for example, sample sets 7 and 8 of Table 3 each provided 100 normalized data points for a regression, set 9 provided 61, and set 10 provided 32.

TABLE 3. — Variability in temperature coefficient.

Analysis procedure	Sample set	No. of laboratories	No. of boards	No. of B values	$-B$	Variability
1	2	3	4	5	6	7
I. Averaging B's						Range of set
(Table 1)						
	1. All, weighted ^a	20	>50	^b 107	8510	5620-12,480
	2. All, unweighted	20	>50	37	8410	5620-12,480
	3. Chamber tests, unweighted	11	>20	22	8540	5620-11,120
	4. Desiccator & WKI tests, unweighted	6	14	11	8250	6140-12,480
	5. Plywood, unweighted	4	10	7	8860	7430-12,480
	6. "Low emission" boards, unweighted ^c	4	13	13	8700	7430-10,020
II. Regression of composite normalized data^d						95% confidence interval^e
(12°-40°C)						
	7. All, weighted ^b	20	>50		8520	8080-8960
	8. All, unweighted	20	>50		8480	8020-8940
	9. Chamber tests, unweighted	11	>20		8930	8390-9470
	10. Desiccator & WKI tests, unweighted	6	14		7670	6590-8750
III. Error analysis for chamber tests (Appendix)						± 2 Standard deviation range for $B = -8930$
	11. Low error (very good controls, broad T range)					8500-9360
	12. Intermediate error					8070-9790
	13. High error (moderate controls, narrow T range)					4290-13,570

^aWeighting factors from Table 1.

^b $\sum w_i/n_i$, where w_i and n_i are weighting factors and number of B values for laboratory i .

^cFor boards with chamber concentration <0.4 ppm at ventilation rate/loading ≤ 1 m/hr., or with desiccator values 1.3 $\mu\text{g}/\text{mL}$.

^dEach investigator data normalized to 25°C . Combined data for each set regressed against Equation [1].

^eThe approximate meaning of the latter is that there is a 95% probability that the true B for similar data sets will lie within the confidence interval.

Table 3 lists the resultant B values for several subsets, along with their 95 percent confidence intervals.² Figure 2 illustrates the results for chamber testing (set 9) and includes the normalized data points, the regression line, and 67 percent and 95 percent confidence intervals.³

This composite regression analysis leads to the following conclusions:

- The weighting factors have little influence on B .
- There exists significant ($p = 0.012$) board-to-board variation at a given laboratory.
- There exists significant ($p = 0.019$) additional laboratory-to-laboratory variation.
- There does not exist significant ($p = 0.80$) board type (plywood versus particleboard) differences.
- There may be a significant ($p = 0.040$) difference between chamber tests (set 9) and water sorption type tests (set 10).

-Data are unfortunately insufficient to establish whether the apparently low reported B values from mobile home measurements (-7520 , Ref. (6), and -7800 , Ref. (11), in Table 1) are really significantly below the chamber value of -8930 (set 9, Table 3). Note, however, that a B of -7520 yields a calculated $c(25^\circ\text{C})/c(40^\circ\text{C})$ only 25 percent greater than given by the composite B .

Group III. — An approximate error analysis was carried out (Appendix) for the case of a chamber test involving a single board, in order to assess the extent to which experimental errors might contribute to the observed uncertainties. Three rather arbitrary situations were evaluated: 1) A “low” error case that assumed a relatively broad temperature range and very good ex-

perimental controls over temperature, humidity, and ventilation rate, along with board parameters that moderated the influence of humidity and ventilation rate variations; 2) a “high” error case that assumed a relatively narrow temperature range and moderately good experimental controls, along with board parameters causing greater sensitivity to humidity and ventilation rate changes; 3) an “intermediate” error case.

The following conclusions are warranted from the error analysis:

—Without careful attention to experimental design and controls, the errors in B from a particular chamber experiment can be expected to approach the observed uncertainties (Group III coefficients of variation from 2% to 26% compared with Group I nominal coefficients of variation from $\sim 10\%$ to 20%).

—Experimental error can be influenced by board properties, i.e., sensitivity to humidity and ventilation rate variations.

From the analyses summarized in Table 3, therefore, it is likely that the variability in observed temperature coefficients has a variety of causes—different emission mechanisms from board to board, inter-laboratory procedural differences, basic differences in board response to very different tests, limitations in experiment design and control. A compounding factor is

²The approximate meaning of the latter is that there is a 95% probability that the true B for similar data sets will lie within the confidence interval.

³Here, the approximate meaning of the confidence intervals is that a future measurement of c will have a 67% or 95% probability of falling within the interval.

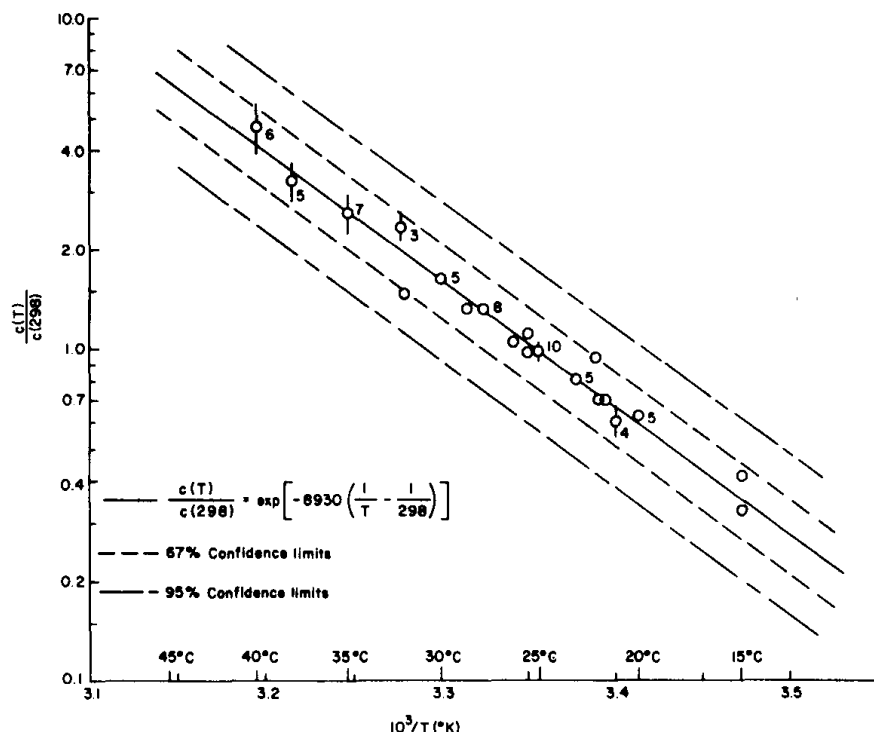


Figure 2. — Normalized temperature dependence of concentration (Circles = normalized data points; adjacent numbers = number of data points at a particular temperature while symbol and vertical line are the corresponding mean $\pm$ one standard deviation).

the likelihood (21, 26, 29) of different rates of board aging and formaldehyde depletion at higher test temperatures. Indeed, perhaps the most surprising outcome of this analysis is the extent of agreement among the observed temperature coefficients.

Recommended temperature corrections

The majority of board-emission temperature coefficients from data available for this review are consistent with an increase of two- to threefold per 10°C temperature rise. In fact the coefficient ($B = -8930^{\circ}\text{K}^{-1}$) for composite chamber data predicts an increase by a factor of 2.7 between 20° and 30°C (c.f., Fig. 2). Therefore, meaningful comparisons of laboratory test data or of home air contamination levels necessitate either that measurements be made at a standard temperature, or that they be corrected to a standard temperature.

In view of the combined high temperature sensitivity and observed high variability in temperature coefficients, however, what reasonable recommendations can be made at this time? I suggest the following:

1) Within any laboratory that has consistently observed a particular B value from board tests, that value obviously should be employed internally to correct similar test data.

2) For correcting home air levels to standard temperature (25°C), one universally acceptable coefficient is very desirable, and that same value would be useful for correcting chamber data in laboratories not having extensive background data of their own. Generally, the results of Berge, et al. (3, 4) appear to have been employed for these purposes. At this time, however, a more reasonable choice would seem to be the coefficient derived from the composite regression of normalized chamber data, i.e., $-8930^{\circ}\text{K}^{-1}$ in set 9, Table 3. Perhaps the most compelling reason for selecting this value now is the existence of at least some measure of its uncertainty and the resultant uncertainty in calculated temperature correction factors. Obviously, as more and better data become available, these correction factors and their uncertainties should be supplanted.

In accordance with this recommendation, Table 4 presents correction factors $c(298)/c(T)$ and their 95 percent confidence limits calculated from the statistical analysis of composite chamber data.⁴

3) It is imperative that measuring temperatures be kept as close as possible to the standard temperature, i.e., small ΔT . Table 4 demonstrates that the uncertainty in correction factors based upon the composite B value increases with greater divergence from 25°C. Similarly, Table 5 illustrates the increasing uncertainty with greater ΔT if a particular board were to possess a temperature coefficient different from the composite value. In this table, hypothetical measured concentrations are corrected to 25°C using three B values, the composite and the two reported (Table 1)

⁴In this case the 95% confidence intervals have the approximate meaning that the true factors for *similar* data sets have a 95% probability of being within those intervals.

TABLE 4. — Temperature correction factors from composite chamber data.

Temperature °C/°F/°K	Correction factor $c(298^{\circ}\text{K})/c(T^{\circ}\text{K})^d$	95% confidence interval ^b	
		Interval	Relative error, (%) ^c
10/50/283	4.89	4.41-5.42	± 9.7
15/59/288	2.83	2.65-3.03	± 6.9
20/68/293	1.67	1.61-1.72	± 3.4
25/77/298	1.00	1.00-1.00	0
30/86/303	0.610	0.591-0.630	± 3.2
35/95/308	0.378	0.355-0.403	± 6.5
40/104/313	0.238	0.217-0.261	± 9.7
45/113/318	0.152	0.135-0.172	± 11.5

^aexp $[-8930 (1/T - 1/298)]$

^bIn this case the 95% confidence intervals have the approximate meaning that the true factors for similar data sets have a 95% probability of being within those intervals.

^c100 $(1 - \text{interval}/2 \times \text{factor})$

TABLE 5. — Effect of extreme temperature coefficients on corrected concentration.

Hypothetical measurement		Concentration corrected to 25°C with B of ^a		
Temperature (°C)	Concentration (ppm)	-8930°K ⁻¹	-5620°K ⁻¹	-11,120°K ⁻¹
15	0.14	0.40	0.27 (33)	0.51 (27)
20	0.24	0.40	0.33 (18)	0.45 (13)
25	0.40	0.40	0.40 (0)	0.40 (0)
30	0.66	0.40	0.48 (20)	0.36 (10)
35	1.06	0.40	0.57 (43)	0.32 (20)
40	1.68	0.40	0.68 (70)	0.28 (30)

^aIn ppm. Numbers in parentheses are relative deviation from 0.40 ppm in percent.

extremes for chamber tests; the "measured" concentrations are such that the composite B produces a 25°C concentration of 0.40 ppm when corrected from all temperatures. Thus the extreme B values lead to under- or overcorrections up to 20 percent for a 5°C ΔT and up to 43 percent for a 10°C ΔT . It must be understood, moreover, that the uncertainties discussed here and in point 2 above are in addition to those caused by experimental errors in making the concentration measurement at some temperature.

4) Additional board testing by several laboratories would be highly desirable under carefully controlled conditions to verify correction factors for a variety of boards representing a range of emission rates and a range of emission-reducing methods.

5) Similarly, additional home testing would be highly desirable under controlled conditions to verify the applicability of the chamber correction factors to the far more complex situation within dwellings.

Humidity dependence

Data summary and analysis model

The investigations reporting humidity/emission results that I considered quantifiable are listed in Table 2. Note that no information is available for humidity effects in dwellings. The table includes abbreviated descriptions of the experiments and results. It also lists (Col. 9) the humidity coefficients, β , obtained from attempts to place each data set on the same basis by regression analysis against the equation

$$c(RH) = c(50\%) [1 + \beta (RH - 50)] \quad [2]$$

Column 10 has weighting factors similar to those for temperature but derived, in this case, by multiplying individual factors for test type, number of boards, likelihood of transient or steady state behavior, and number of humidities.

As with Equation [1], the choice of Equation [2] to describe humidity dependence is not based on a statistical comparison of different models. Instead, it is based upon the facts that humidity dependence of emission is much weaker than temperature dependence, and that several investigators (1, 4, 5, 16, 19) have previously found linear models to fit their data satisfactorily. As will be seen, moreover, a more complex model is hardly warranted at this time in view of the uncertainties surrounding measurements of humidity dependence.

Variability in humidity coefficients

The range of β values is about tenfold, i.e., two to three times greater than the observed range in temperature coefficients. Table 6 presents the results of analyses of this variability and its causes—analyses analogous to those for temperature dependence (Table 3) and subject to similar limitations:

Group I. — Mean β values (Col. 6, Table 6) were derived by averaging different subsets of the observed β 's from Table 2. Nominal coefficients of variation from the means for sets 1 to 5 are 51, 55, 86, 30, and 62 percent, respectively. Despite the broad ranges of values, the means are relatively close, and it is not likely that much significance can be attributed to their differences.

Group II. — Each investigator's data for a given board were first normalized to 50 percent RH, i.e., $c(RH)/c(50\%)$ ratios were calculated. This included actual experimental data points (~ 50 total) as well as points (~ 10 total) calculated from reported humidity coefficients (10, 11, 38, 39). Different subsets of those normalized values were then treated as composite data sets and regressed against Equation [2].

Table 6 gives the resultant β values only for weighted and unweighted regressions (sets 6 & 7), along with their 95 percent confidence intervals.⁴ Figure 3 illustrates the results for the unweighted case and includes the normalized data points, the regression line, and the 67 percent and 95 percent confidence intervals for future measurements.³ The analysis justifies the following conclusions:

—The weighting process did not affect the β coefficient.

—There exists significant ($p < 0.001$) board-to-board variation at a given laboratory.

—There does not exist significant additional board type ($p = 0.27$) or laboratory-to-laboratory ($p = 0.69$) differences.

Thus, for humidity dependence, board-to-board differences may be the major source of variation. Not explicitly accounted for by the statistical analysis, how-

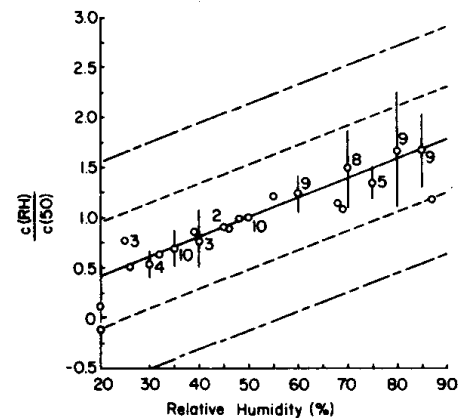


Figure 3. — Normalized humidity dependence for chamber tests (symbols as in Figure 2 except solid line from $c(RH)/C(50\%) = [1 + 0.0195 (RH - 50)]$).

TABLE 6. — Variability in humidity coefficients.

Analysis procedure	Sample set	No. of laboratories	No. of boards	No. of β values	β	Variability
1	2	3	4	5	6	I
I. Averaging β's						Range of set
(Table 2)						
	1. All, weighted ^a	13	>40	^b 89	0.017	0.005-0.038
	2. All, unweighted	13	>40	31	0.017	0.005-0.038
	3. Plywood, unweighted	3	5	5	0.013	0.005-0.033
	4. Coated particleboard, unweighted	1	6	6	0.021	0.017-0.030
	5. "Low emission" boards, unweighted ^c	1	5	5	0.021	0.006-0.037
II. Regression of composite normalized data^d						95% confidence interval^e
	6. All, weighted ^a	13	>40		0.020	0.014-0.025
	7. All, unweighted	13	>40		0.020	0.014-0.025

^aWeighting factors from Table 2.

^b $\sum w_i n_i$, where w_i and n_i are weighting factors and number of β values for laboratory i .

^cFor boards with chamber concentration ≤ 0.4 ppm at ventilation rate/loading ≤ 1.0 m/hr.

^dEach investigator data normalized to 50°C. Combined data regressed against Equation [2].

^eThe approximate meaning of the latter is that there is a 95% probability that the true B for similar data sets will lie within the confidence interval.

ever, is the very slow and somewhat erratic response of board emission to humidity changes (0, 7, 14, 18). This is well-illustrated in Figure 4 where some of the humidity cycling results of Liiri, et al. (14) are shown. Even 4 weeks after a humidity change, the chamber concentration did not always become constant or even reach the same value after each cycle. Moreover, upon changing humidity there may be an initial sharp change in emission in the opposite direction from the eventual change, as seen for Board A upon changing from 85 to 35 percent RH. These transients are postulated to be caused by water vapor carrying the formaldehyde with it as it leaves or enters the board under the driving force of a humidity change. To what extent these time effects may have influenced the calculated humidity coefficients β (Table 2, Col. 9) is not clear. Although test duration times (Table 2, Col. 7) varied from 4 weeks to only a few hours and some data sets almost certainly represented nonsteady state experiments, there is no obvious correlation between test times and β values.

Recommended humidity correction

For the following reasons I suggest that for the present it is most reasonable not to make humidity corrections unless approximate humidity coefficients β are available for the particular boards under test:

1) Correction factors for anticipated humidity ranges may be small relative to those for anticipated temperature ranges. The composite humidity coefficient of 0.0195 RH^{-1} , for example, predicts a concentration change of 20 to 25 percent per 10 percent RH change in the mid-RH region (Table 7); it also leads to a correction factor of 0.63 for converting measurements at 80 percent RH to 50 percent RH. In contrast the composite temperature coefficient predicts a concentration change of nearly 400 percent per 10°C temperature change, and it leads to a correction factor of 0.61 for converting measurements at 30°C to 25°C (Table 4). Without careful temperature control, therefore, humidity changes of up to 10 percent RH may well be hidden within the effects of temperature fluctuation.

2) Confidence in the applicability of the composite humidity coefficient to a given board is much lower than it is in the applicability of the composite temperature coefficient. Continuing the above example, the 95 percent confidence relative error in the 80 percent to 50 percent RH correction factor of 0.63 is ± 11 percent, whereas the analogous relative error in the 30°C to 25°C temperature correction factor of 0.61 is ± 3 percent (Table 4 versus Table 7).⁴ Expressed in other terms, the range of reported β values is much greater than the range of B and the existence of board-to-board variations appears to be much more likely for humidity response.

3) Contributing to the uncertainty in humidity correction factors is the slow and time-varying response of board emissions to humidity changes. Such temporal effects create the strong possibility of error in formaldehyde test values and in β values.

It follows from the above points that, as with temperature, measurements should be made as close as possible to the standard humidity (50% RH). Moreover,

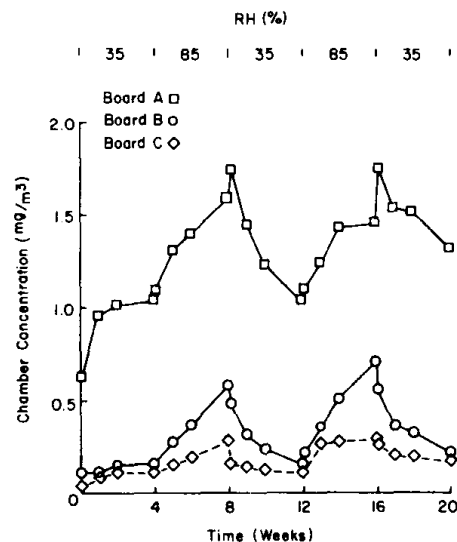


Figure 4. — Effect of humidity cycling on particleboard formaldehyde emission (ref. (14)).

TABLE 7. — Humidity correction factors from composite data treatment.

Relative humidity (%)	Correction factor $c(50\%)/c(RH)^a$	95% confidence interval ^b	
		Interval	Relative error ^c (%)
20	2.41	1.71-4.13	± 42
30	1.64	1.38-2.02	± 19
40	1.24	1.16-1.34	± 7
50	1.00	1.00-1.00	± 0
60	0.84	0.80-0.88	± 5
70	0.72	0.66-0.78	± 8
80	0.63	0.57-0.71	± 11
90	0.56	0.50-0.64	± 13

^a $[1 + 0.0195 (RH - 50)]^{-1}$.

^bIn this case the 95% confidence intervals have the approximate meaning that the true factors for similar data sets have a 95% probability of being within those intervals.

^c $100 (1 - \text{interval}/2 \times \text{factor})$

additional study is needed into the humidity dependence of formaldehyde concentrations in chamber tests and in dwellings, using a wide variety of boards under carefully controlled conditions.

Summary

Temperature dependence

1. Available results on the temperature dependence of formaldehyde concentration in chambers or buildings containing particleboard and hardwood plywood paneling were fit with an expression that is exponential in temperature (Equation [1]). For individual measurement sets the temperature coefficient B varied from -5620° to $-12,480^\circ\text{K}^{-1}$, and calculated values of $c(30^\circ\text{C})/c(20^\circ\text{C})$ varied from 1.9 to 4.1.

2. Statistical treatment of all chamber test data as a composite set (normalized to unity at 25°C) yields a composite B of -8930°K^{-1} , with a 95 percent confidence interval from -8390° to -9470°K^{-1} ($\pm 6\%$ relative error).⁷ Using this composite B , calculated correction factors, $c(25^\circ\text{C})/c(T)$, varied from 4.89 at 10°C to 0.152 at 45°C .

3. The statistical analysis indicates that significant board-to-board and laboratory-to-laboratory variations occurred. However, it is not clear that there are significant temperature response differences between board types (particleboard versus hardwood plywood paneling), between "low" and "high" emission boards, or between chamber tests and dwellings.

Humidity dependence

1. The response of board formaldehyde emission to humidity changes is more complex than that to temperature change and may not have achieved an equilibrium or steady state by the time the concentration measurements are performed. Nevertheless, the available data (primarily chamber tests) on humidity dependence were fit with an expression (Equation [2]) that is linear in percent RH. For individual measurement sets the humidity coefficient β varied from 0.005% to 0.038%⁻¹.

2. A composite analysis of all the data (normalized to unity at 50% RH) yields a composite β of 0.0195%⁻¹ with a 95 percent confidence interval from 0.014 to 0.025 (± 28 percent relative error).² The corresponding correction factors, $c(50\%)/c(\text{RH})$, vary from 2.41 at 20 percent RH to 0.56 at 90 percent RH.

Conclusions and recommendations

Temperature dependence

1. An exponential Arrhenius type expression (Equation [1]) provides adequate representation of the strong temperature dependence of formaldehyde emission by particleboard and hardwood plywood paneling.

2. Large variations in temperature coefficients are observed. These variations probably arise from inadequate experimental controls, laboratory procedural differences, board aging effects, and differences in board emission mechanisms. More work is needed to clarify this situation and provide more reliable (possibly more board-discriminating) temperature coefficients for both laboratory and dwelling tests.

3. Because of the strong temperature dependence of emission and the observed variability in temperature coefficients, formaldehyde emission measurements should be made as closely as possible to 25°C ($\pm 5^\circ\text{C}$) and the measured values corrected to 25°C.

4. Any laboratory having extensive and consistent experience in this area should, of course, make use of that experience in correcting internal data for temperature effects.

5. For laboratories without such experience and for formaldehyde concentrations in dwellings, I suggest for the present that the composite temperature coefficient (-8930°K^{-1}) and associated correction factors (Table 4) be used in correcting for temperature effects, with full appreciation for the uncertainties involved.

Humidity dependence

1. Very large variations between investigators, and between boards, lead to poor confidence in universal humidity correction factors.

2. It is probable that humidity response can vary from board to board. However, the entire situation is

clouded due to the complex, time-dependent response of board emission to humidity changes. Additional study would be very useful on a wide variety of boards and in buildings, with careful attention to time effects.

3. No clear recommendation can be made regarding correction factors to convert formaldehyde concentrations to a standard humidity. Small humidity changes ($\sim 10\%$ RH), however, probably can be neglected since they may be masked by rather minor temperature changes ($\pm 1^\circ$ to 2°C).

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Appendix

Error analysis for temperature coefficient

We assume first that a steady state chamber concentration c (ppm) depends upon temperature T (°K), relative humidity RH (%), ventilation rate N (hr.⁻¹), and

board loading L (m²/m³) according to (4, 27)

$$c = c^0 e^{B \left(\frac{1}{T} - \frac{1}{298} \right)} [1 + \beta(RH - 50)] \cdot \frac{1}{1 + \frac{1}{K} \frac{N}{L}} \quad [A1]$$

TABLE A1. — Illustrative error calculation for temperature coefficient.

Error source	Parameter ^a	Estimated value for		
		Low error	High error	Intermediate error
Temperature variation	T_2 (°K)	40	35	37.5
	T_1 (°K)	15	20	17.5
	ΔT (°K)	25	15	20.0
	s_T (°K)	0.1	0.3	0.2
	s^2/B^2 from s_T direct ^b	0.66×10^{-4}	0.16×10^{-2}	2.6×10^{-4}
Humidity variation	s^2/B^2 from s_T on c^c	4.5×10^{-4}	0.16×10^{-2}	10.4×10^{-4}
	RH (%)	80	30	50.0
	β (% ⁻¹)	0.01	0.03	0.02
	s_{RH} (%)	1	2	1.5
	s^2/B^2 from s_{RH} on c^c	0.19×10^{-4}	2.48×10^{-2}	4.73×10^{-4}
Ventilation rate variation	N/L (m/hr.)	1.5	0.5	1.0
	K (m/hr.)	1.0	0.1	0.5
	$(s_{N/L})/(N/L)$	0.01	0.02	0.015
	s^2/B^2 from $s_{N/L}$ on c^c	0.04×10^{-4}	3.88×10^{-2}	0.79×10^{-4}
Formaldehyde analysis	s_{an}/c	0.01	0.05	0.025
	s^2/B^2 from s_{an} on c^c	0.33×10^{-4}	0.28×10^{-2}	4.93×10^{-4}
Total Relative error at 67% confidence	s^2/B^2	5.7×10^{-4}	7.0×10^{-2}	23.5×10^{-4}
Relative error at 95% confidence	$\pm 100 s/B$	2.4%	26%	4.8%
	$\pm 200 s/B$	4.8%	52%	9.6%

^a T = temperature; s = standard deviation; B = temperature coefficient; RH = relative humidity; N/L = ventilation rate/loading; β = humidity coefficient; K = transport parameter.

^bEquation [A2], first two terms.

^cEquation [A3].

Here, B , β , and K are experimentally determined parameters and c° is the concentration at 25°C, 50 percent RH, and zero ventilation rate.

Assuming, further, that T , RH , and N/L act independently, we can write the relative error in determining B from a measurement of c at two temperatures and constant RH and N/L (2):

$$\left(\frac{\text{Variance } B}{B^2}\right)_{RH, \frac{N}{L}} = \frac{s_B^2}{B^2} + \left(\frac{T_2}{T_1 - T_2}\right)^2 \left(\frac{s_{T_1}}{T_1}\right)^2 + \left(\frac{T_1}{T_1 - T_2}\right)^2 \left(\frac{s_{T_2}}{T_2}\right)^2 + \left(\frac{1}{\ln(c_2/c_1)}\right)^2 \left[\left(\frac{s_{c_1}}{c_1}\right)^2 + \left(\frac{s_{c_2}}{c_2}\right)^2 \right] \quad [A2]$$

In [A2] the various s 's are the standard deviations for measuring the respective quantities. The errors in concentration (s_c/c) result from uncertainties in RH (s_{RH}), N/L , ($s_{N/L}$), and T (s_T), plus those in the analytical method for formaldehyde (s_{an}). Thus by standard propagation of error treatment on Equation [A1], the third term of Equation [A2] becomes

$$\left(\frac{1}{\ln \frac{c_2}{c_1}}\right)^2 \left[\left(\frac{s_{c_1}}{c_1}\right)^2 + \left(\frac{s_{c_2}}{c_2}\right)^2 \right] \approx \left(\frac{1}{\ln \frac{c_2}{c_1}}\right)^2 \times \left[\frac{\beta^2 s_{RH}^2}{[1 + \beta(RH - 50)]^2} + \frac{\frac{1}{K^2} s_{N/L}^2}{\left(1 + \frac{1}{K} \frac{N}{L}\right)^2} + \frac{B^2 s_T^2}{T^4} + \frac{s_{an}^2}{c^2} \right] \quad [A3]$$

Using reasonable estimates of the parameters and errors in Equations [A2] and [A3], I have evaluated the equations for low, high, and intermediate error cases (Table A1) in order to illustrate the effects that might be experimentally observed. The intermediate case is, of course, the most plausible since it more nearly randomizes the various influences towards low or high errors. The following conclusions appear warranted:

1. Total errors are at least potentially quite large and each source can be a major contributor.
2. The properties of the board under study and the choice of experimental conditions can have a major influence on the error and, therefore, on the degree of experimental control required. Error increases, for example, at high β and low K , as well as at low T , low RH , and low N/L .