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Effect of Temperature and Humidity on Formaldehyde Emissions in Temporary Housing Units

Srinandini Parthasarathy, Randy L. Maddalena, Marion L. Russell, and Michael G. Apte
*Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory,
Berkeley, CA*

ABSTRACT

The effect of temperature and humidity on formaldehyde emissions from samples collected from temporary housing units (THUs) was studied. The THUs were supplied by the U.S. Federal Emergency Management Administration (FEMA) to families that lost their homes in Louisiana and Mississippi during the Hurricane Katrina and Rita disasters. On the basis of a previous study, four of the composite wood surface materials that dominated contributions to indoor formaldehyde were selected to analyze the effects of temperature and humidity on the emission factors. Humidity equilibration experiments were carried out on two of the samples to determine how long the samples take to equilibrate with the surrounding environmental conditions. Small chamber experiments were then conducted to measure emission factors for the four surface materials at various temperature and humidity conditions. The samples were analyzed for formaldehyde via high-performance liquid chromatography. The experiments showed that increases in temperature or humidity contributed to an increase in emission factors. A linear regression model was built using the natural log of the percent relative humidity (RH) and inverse of temperature (in K) as independent variables and the natural log of emission factors as the dependent variable. The coefficients for the inverse of temperature and log RH with log emission factor were found to be statistically significant for all of the samples at the 95% confidence level. This study should assist in retrospectively estimating indoor formaldehyde exposure of occupants of THUs.

IMPLICATIONS

Maddalena et al. reported differences between formaldehyde concentrations in samples collected from the THUs during the morning and afternoon of the same day, highlighting the need to carry out further analysis on the effect of temperature and humidity on formaldehyde emissions. This study addresses the influence of temperature and humidity on formaldehyde emission factors from individual materials. The information provided can be incorporated into an exposure assessment study for the occupants of the FEMA trailers. However, because the experiments are carried out only on four samples from the THUs, they might not be representative of the entire fleet of THUs.

INTRODUCTION

This study is part of a larger effort to retrospectively estimate indoor formaldehyde exposures of the occupants of temporary housing units (THUs). The U.S. Federal Emergency Management Administration (FEMA) supplied over 100,000 emergency THUs to families that lost their homes in Louisiana and Mississippi during the Hurricane Katrina and Rita disasters. Concerns about the indoor environmental quality in the THUs emerged based on occupant health complaints and concerns. The U.S. Environmental Protection Agency (EPA)¹ reports that various studies^{2–4} have shown that acute exposure to formaldehyde can cause irritation in the eyes, nose, and throat. Human studies^{5,6} have reported increased levels of respiratory illnesses because of chronic formaldehyde exposure. Numerous studies^{7,8} showed that chronic exposure to formaldehyde caused increased cancer incidence in rodents. Some human studies^{9,10} have also reported higher rates of respiratory-site cancer occurrence among subjects regularly exposed to high formaldehyde levels. Formaldehyde is currently classified as a probable human carcinogen by EPA.¹¹ Measurements reported^{12,13} showed that formaldehyde concentrations observed in occupied and unoccupied THUs exceeded the National Institute for Occupational Safety and Health (NIOSH) recommended exposure limit (REL) of 0.016 parts per million (ppm),¹⁴ often by a factor of 10 or greater. The NIOSH REL was based on the analytical limit of detection and not on health effects data.

In the THUs, sources contributing to elevated indoor formaldehyde concentrations were related to building materials and furnishings. Maddalena et al.^{15,16} measured the indoor concentration and whole trailer emission factors in four unoccupied THUs for a range of volatile organic compounds (VOCs) and aldehydes. The study also determined the material-specific emission factors of the compounds from individual surface materials collected directly from the THUs. It was observed that all THUs had a significant portion of the internal surface area constructed with 1/8-in. plywood with a vinyl or polyvinyl chloride skin or simulated wood finish. All units had sheet vinyl flooring, and two of the four trailers also had carpeted areas. All countertops were particle-board surfaced with high-pressure laminate. Various wood products were used for the subfloor and for the bench and bed platforms. Formaldehyde was observed to be the only aldehyde emitted from these materials at rates sufficient to be of health

concern. A range of VOCs typically present when formaldehyde is observed¹⁷ are also emitted from materials. Like formaldehyde, which is a toxic air contaminant,^{1,18} many of the VOCs are known to have low odor thresholds, high potency as respiratory irritants, and in some cases carcinogenicity. On the basis of the previous study,^{15,16} the surface materials that dominated contributions to indoor formaldehyde were selected to analyze the effects of temperature and humidity on the emission factors. As detailed by Hawthorne et al.¹⁹ the mechanism of formaldehyde emissions depends on the production of formaldehyde in the bulk material, the transport through the bulk material, and the transfer of formaldehyde out of the bulk material and into the atmosphere. A few key studies have been carried out to measure the effect of temperature and relative humidity (RH) on formaldehyde emissions. Zhang et al.²⁰ conducted chamber experiments to understand the influence of temperature on the partition coefficient and diffusion coefficient and found that the partition coefficient decreases with an increase in temperature, and the diffusion coefficient increased with an increase in temperature. However, the equilibrium concentration of formaldehyde increased with an increase in temperature. Andersen et al.²¹ conducted field and chamber experiments on formaldehyde emissions from particle board. These chamber experiments showed that the emissions had a strong positive correlation with the prevailing temperature and humidity conditions. Van Netten et al.²² conducted chamber experiments on various materials (ceiling tile, gypsum board, shiplap, plywood, terracotta brick) that release formaldehyde and reported that higher emissions were observed with increases in temperature, humidity, or both.

In a literature review, Myers²³ reported that considerable variations existed between different kinds of boards in their response to varying environmental conditions. Myers²³ reports that the temperature coefficients (where log concentration was the dependent variable) for various types of composite wood materials fall within an approximate 2-fold range, as shown in Table 1. Further, humidity coefficients for various types of composite wood materials fall within the range of 0.005–0.038 (log RH⁻¹) for log RH.

The study also emphasized that significant variations existed among various types of wood in their response to changing temperature and humidity conditions. Myers²³ assumed an exponential relationship between the concentration and the inverse of temperature (in K) on the basis of the Arrhenius equation. On the basis of previous

studies, he assumes a linear relationship between concentration and RH.²⁴

EXPERIMENTAL METHODS

Materials

Surface materials were cut from THUs for analysis to determine material-specific formaldehyde emission factors. Samples selected for analysis were previously tested for emissions under 23 ± 1 °C in a controlled environmental chamber with a $0.06\text{-m}^3\text{ hr}^{-1}$ inlet flow of carbon-filtered preconditioned air at $50\% \pm 5\%$ RH. These materials included a subfloor ($416\ \mu\text{g m}^{-2}\text{ hr}^{-1}$) and cabinet wall ($488\ \mu\text{g m}^{-2}\text{ hr}^{-1}$) from trailer 1 manufactured in March 2006, a benchseat ($233\ \mu\text{g m}^{-2}\text{ hr}^{-1}$) from trailer 2 manufactured in October 2006, and a cabinet wall ($419\ \mu\text{g m}^{-2}\text{ hr}^{-1}$) from trailer 3 manufactured in October 2005. The samples were wrapped in two layers of aluminum foil and then stored in envelopes until the time of testing. The subfloor sample is made from particle board, and the benchseat, cabinet, and cabinet wall samples are all made from plywood.

Humidity Equilibration

Wood is a hygroscopic material; that is, it tends to adsorb or desorb moisture on the basis of the environmental conditions. Humidity equilibration experiments were carried out to determine the time taken by samples to attain equilibrium under conditions of altered humidity. The subfloor ($6 \times 6 \times 3/8$ in.) from trailer 1 and the cabinet wall ($6 \times 6 \times 1/8$ in.) from trailer 3 were selected for these experiments. All experiments were carried out in four chambers of 10.75-L capacity each. The air exchange rate was maintained at a constant value during all of the experiments. Each material was cut in half along the vertical axis, and the two samples were placed in chambers held at identical temperature and RHs of 50% and 85%. There were four stainless steel chambers installed in a Forma Scientific incubator for temperature control. House air was supplied to each chamber at a rate of 1 L/min. The air was passed through a carbon filter and a high-efficiency particulate air particle filter before being split to each chamber. Further, the purified air to each chamber was split again into a dry line and a wet line (bubbled through deionized water). RH adjustments were made by changing the flow between the wet or dry line that was supplied to each chamber. A Vaisala RH probe and temperature transmitter (P/N HMD30YB) was installed in each chamber to measure the RH changes. LabTech Notebook software was used to continuously log the data over the course of the experiment. The temperature and RH at which the experiments were carried out are listed in Table 2. A schematic diagram of the experimental setup is provided in Figure 1. The samples were weighed using a semimicroanalytical balance Ohaus model DV314C at the start of the experiment and weighed once daily until the weight of the sample reached a constant value. The temperatures in both chambers were altered when the samples attained equilibrium under the temperature and humidity conditions maintained previously.

Table 1. Temperature and humidity coefficient ranges reported in literature.

Material	Temperature Coefficient (K)		Humidity Coefficient (log RH ⁻¹)	
	Minimum	Maximum	Minimum	Maximum
Particle board	-11,120	-5,620	0.005	0.038
Plywood	-9,600	-7,430	0.006	0.033

Notes: Data from Myers.²³

Table 2. Humidity equilibration experiments.

Sample Number	Sample	Material	Temperature (°C)			RH (%)	
			15 ± 1	25 ± 1	35 ± 1	50 ± 2	85 ± 9
1	Cabinet wall		X			X	
2	Cabinet wall			X		X	
3	Cabinet wall	Plywood			X	X	
4	Cabinet wall		X				X
5	Cabinet wall			X			X
6	Cabinet wall				X		X
7	Subfloor		X			X	
8	Subfloor			X		X	
9	Subfloor	Particle board			X	X	
10	Subfloor		X				X
11	Subfloor			X			X
12	Subfloor				X		X

Formaldehyde Emissions under Various Temperature and Humidity Conditions

Two samples were cut from each material according to the dimensions specified in Table 3 and prepared for emission studies. Stainless steel backing plates were cleaned twice with methanol, air-dried, and baked overnight at 50 °C in an oven. The backing plates were taped to the back of the samples using Scotch 3M metal repair tape. Four pieces of tape were used to seal the edges of the material and to hold the backing plate in place.

The two samples from each material were placed in chambers held at identical temperature and RHs of 50% and 85%. The experiments were carried out at the various temperature and RH conditions specified in Table 3. The samples were placed in the chamber for an average of 1 hr before the air sampling was started. The air sampling for analysis was conducted daily for each chamber until the formaldehyde concentration was found to reach a steady value. Each sample was retained in the chamber under specified conditions of temperature and humidity until it equilibrated with the surrounding atmosphere. Blank samples were taken in

empty chambers and with tape to measure background formaldehyde concentrations.

Formaldehyde Sampling and Analysis

The air samples were drawn directly from each small emission chamber. Samples were collected using a vacuum pump (model DOAP104-AA; Gast) with sample flow rates regulated by electronic mass flow controllers. Aldehyde samples were collected on commercially available silica gel cartridges coated with 2,4-dinitrophenylhydrazine (XPOsure aldehyde sampler; Waters Corporation). Sample cartridges were capped, sealed in an aluminum envelope, and stored in the freezer until extraction. Cartridges were eluted with 2 mL of low-carbonyl-grade, high-purity acetonitrile into 2-mL volumetric flasks, and the eluent was brought to a final volume of 2 mL before analysis. Extracts were analyzed by high-performance liquid chromatography (HPLC) (1200 series; Agilent Technologies) using a C18 reverse-phase column with a 65%:35% water:acetonitrile mobile phase at 0.35 mL/min and ultraviolet detection at 360 nm. Multipoint calibrations were prepared using commercially available hydrazone derivatives of formaldehyde.

Quality Assurance

All samples were quantified with multipoint calibration curves prepared from pure chemicals. Analytical blanks were included in all analyses. Blanks for the emission experiments included backing plate and tape. Chamber blanks representing only the background in the chamber were also collected. Standards were purchased from Supelco. Dinitrophenylhydrazine derivatives of formaldehyde (P/N 47177), acetaldehyde (P/N 47340-U), and acetone (P/N 47341) were dissolved in acetonitrile and used to create a calibration curve for the HPLC analysis method. A midrange standard was run with each batch of samples to confirm the calibration and retention times of each analyte.

DATA ANALYSIS AND RESULTS

Emission Rate

The emission factors were normalized to the surface area of the samples. The steady-state form of the mass balance

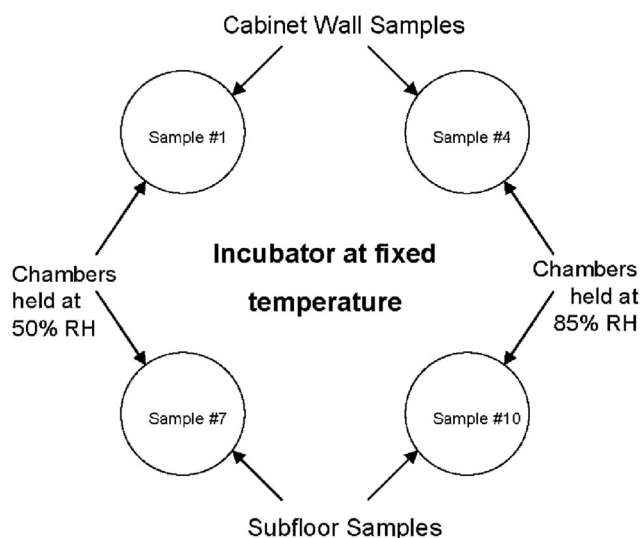


Figure 1. Schematic diagram of experimental setup humidity equilibration experiments.

Table 3. Overview of experiments and measured steady-state concentrations of formaldehyde in chamber experiments.

Sample Number	Sample	Temperature (°C)			RH (%)		Sample Dimensions			Ventilation Rate (L min ⁻¹)	Concentration (µg m ⁻³)	Emission Factor (µg m ⁻² hr ⁻¹)
		15 ± 1	25 ± 1	35 ± 1	50 ± 1	85 ± 3	Length (cm)	Breadth (cm)	Area Exposed (m ²)			
1	Benchseat	X			X					1.0	9.9	62
2	Benchseat		X		X		14.2	6.5	0.009	0.9	21	130
3	Benchseat			X	X					1.0	43	270
4	Benchseat	X				X				1.0	20	130
5	Benchseat		X			X	13.8	6.6	0.009	1.0	47	300
6	Benchseat			X		X				1.0	110	670
7	Cabinet	X			X					0.9	9.9	70
8	Cabinet		X		X		13.1	6.0	0.008	0.9	20	140
9	Cabinet			X	X					0.9	55	390
10	Cabinet	X				X				1.0	15	100
11	Cabinet		X			X	13.2	6.2	0.008	1.0	47	320
12	Cabinet			X		X				1.0	140	1000
13	Cabinet wall	X			X					1.0	9.4	60
14	Cabinet wall		X		X		14.1	6.6	0.009	1.0	17	110
15	Cabinet wall			X	X					1.0	45	280
16	Cabinet wall	X				X				1.0	18	110
17	Cabinet wall		X			X	14.0	6.5	0.009	1.0	40	260
18	Cabinet wall			X		X				1.0	100	640
19	Subfloor	X			X					0.9	15	100
20	Subfloor		X		X		14.0	6.5	0.009	0.9	44	270
21	Subfloor			X	X					0.9	120	770
22	Subfloor	X				X				1.0	24	140
23	Subfloor		X			X	14.2	7.0	0.010	1.0	85	480
24	Subfloor			X		X				0.9	270	1600

Notes: The steady-state concentrations presented in this table are corrected for formaldehyde emissions resulting from the backing plate, tape, and background formaldehyde levels in the air.

equation for calculating area-specific emission factors (*EF*; µg m⁻² hr⁻¹) in a well-mixed system is

$$EF = \frac{f \times (C - C_o)}{A} \quad (1)$$

where *f* (m³ hr⁻¹) is the ventilation rate, *A* (m²) is the exposed surface area of the sample, *C* (µg m⁻³) is the

measured steady-state concentration in the chamber, and *C*_o (µg m⁻³) is the background concentration in the chamber.

RESULTS AND DISCUSSION

Figures 2a and 2b show the results from the humidity equilibration experiments for the cabinet wall and subfloor samples. The goal of these experiments was to

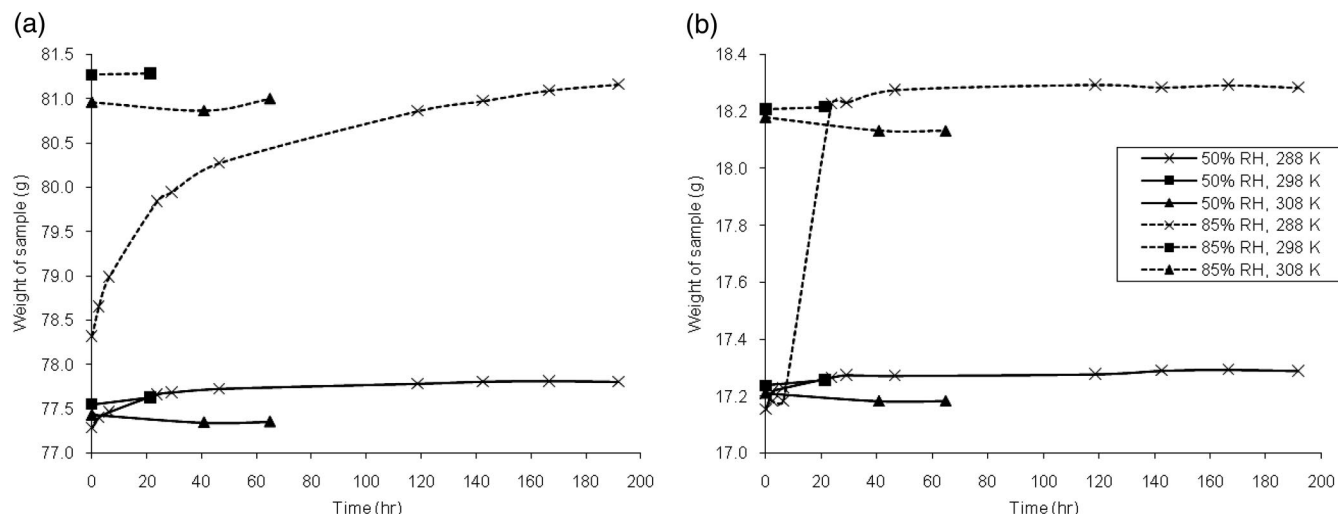


Figure 2. Results from humidity equilibration experiments: (a) cabinet wall sample and (b) subfloor sample.

estimate the time for the samples to equilibrate with the environmental conditions. It is seen in Figures 2a and 2b that the moisture gain by both samples was significantly high during the 50% RH, 15 °C and 85% RH, 15 °C conditions. The samples were first exposed to 50% RH, 15 °C and 85% RH, 15 °C conditions when the sampling began. The sudden exposure to high humidity after they were stored for a few months wrapped in aluminum foil resulted in higher levels of moisture uptake under these conditions. As shown by Figures 2a and 2b, the samples take approximately 40 hr to equilibrate. However, under the 85% RH, 15 °C conditions, the samples take approximately 240 hr to reach equilibrium.

To analyze the effect of temperature and humidity on aldehyde emissions, a total of six experiments were carried out for each sample. The experiments lasted until the steady-state concentration of formaldehyde remained constant. The results are tabulated in Table 3, where it can be seen that the concentration of formaldehyde increases between 1.9–3.5 times for a 10 °C rise in temperature depending on the sample type. Humidity does not influence the emissions as strongly as temperature. However, a 35% increase in humidity can increase the emissions by 1.8–2.6 times depending on the material. The effect of humidity on emission is more pronounced at higher temperatures.

Temperature and RH have a strong positive correlation with the emission factors for all of the samples. The correlation coefficient (R^2) between temperature and emission factors for all samples was found to be greater than 0.83, and the R^2 between RH and emission factor for all samples was found to be greater than 0.98. Temperature and RH were not correlated ($-0.05 < R^2 < 0.03$).

A linear regression model was built setting the natural log of emission factors as the dependent variable. Natural log of percent RH and inverse of temperature were used as the independent variables. The coefficients of inverse temperature and log RH with log emission factor were found to be statistically significant for all of the samples at the 95% confidence level, as shown in

Table 4. Figures 3a–3d show the Arrhenius plots of modeled and measured emission factors versus temperature and humidity. The inverse temperature coefficient for the benchseat, cabinet, cabinet wall, and subfloor were -6740 , -8500 , -7030 , and -9940 K, respectively. The log RH coefficients for the benchseat, cabinet, cabinet wall, and subfloor were 1.55, 1.47, 1.42, and 1.17 $\log \text{RH}^{-1}$, respectively. The regression model also yielded excellent fits with the experimental data as shown in Table 4.

Previous studies assumed a linear relationship between concentration and RH.²⁴ Hence, this study assumes a linear relationship between emission rate and RH. However, a direct comparison of the humidity coefficients generated in the study presented here with previous work is not possible. Wu²⁵ applied the wood water adsorption dynamics developed by Nelson²⁶ to wood composite products. The sorption isotherm exhibits a fairly linear trend between approximately 20 and 90% RH. On the basis of the analysis by Wu,²⁴ the results generated in this study could be applied over the 20–90% RH range. Myers²³ assumed an exponential relationship between the concentration and the inverse of temperature, and this study assumes an exponential relationship between the emission rate and the inverse of temperature. The ventilation rates and exposed area for each sample are fairly constant across all experimental conditions. The emission factors are related to the concentration by an almost constant factor for each sample. Hence, the log of emission factors and log of concentration vary in a similar linear fashion with change in temperature. Hence, an order of magnitude and sign comparison can be made between the temperature coefficients generated in the regression analysis and the values reported in the literature. Myers²³ states that the temperature coefficients reported for various types of composite wood materials fall in the range of $-11,120$ to -5620 K. The temperature coefficients estimated in this study for particle board falls within this

Table 4. Linear regression modeling results.

	Coefficients	Confidence Limits		Regression Statistics	
		Lower	Upper	R^2	P
Benchseat					
Inverse temperature coefficient (K)	-6,740	-7,640	-5,840	0.996	0.0002
log RH coefficient ($\log \text{RH}^{-1}$)	1.546	1.225	1.866		0.0006
Intercept	21.4	18.1	24.7		–
Cabinet					
Inverse temperature coefficient (K)	-8,500	-11,100	-5,940	0.979	0.002
log RH coefficient ($\log \text{RH}^{-1}$)	1.468	0.569	2.366		0.01
Intercept	27.8	18.0	37.0		–
Cabinet wall					
Inverse temperature coefficient (K)	-7,030	-8,660	-5,390	0.988	0.0008
log RH coefficient ($\log \text{RH}^{-1}$)	1.421	0.813	2.028		0.005
Intercept	22.9	16.8	28.9		–
Subfloor					
Inverse temperature coefficient (K)	-9,940	-11,500	-8,400	0.994	0.0003
log RH coefficient ($\log \text{RH}^{-1}$)	1.166	0.625	1.708		0.006
Intercept	34.4	29.0	40.0		–

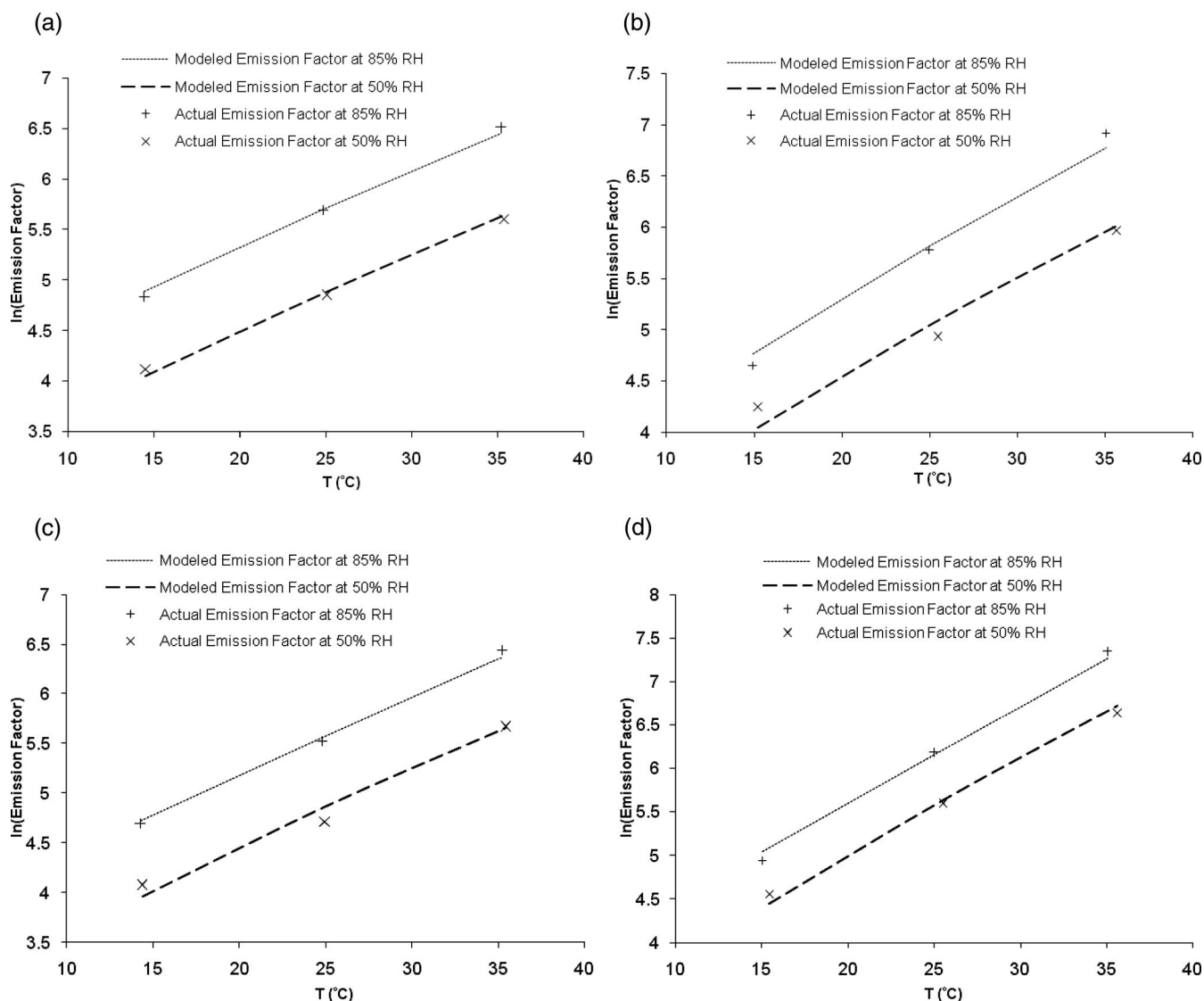


Figure 3. Arrhenius plot of modeled and predicted emission factors as a function of temperature and RH: (a) benchseat sample, (b) cabinet sample, (c) cabinet wall sample, and (d) subfloor sample.

range. Additionally, Myers²³ reports temperature coefficients for plywood in the range of -9600 to -7430 K. The temperature coefficient estimated for the particle board sample (-9940 K, subfloor) and plywood (-8500 K, cabinet) falls within this reported range whereas the temperature coefficients for the benchseat and cabinet wall plywood samples (-6740 and -7030 K, respectively) fall close to this reported range. Previous studies report that higher emission rates are observed with increase in ventilation rates; however, the current experiments were carried out at a constant ventilation rate of 5.7 hr⁻¹. The study is limited to observing the effects of temperature and RH on formaldehyde emission factors, when the ventilation rates are held constant and the formaldehyde concentrations in the chamber are constantly changing.

CONCLUSIONS

Chamber experiments were carried out to gauge the effect of temperature and humidity on formaldehyde emission

factors. The experiments established that 10 °C variation in temperature increased the formaldehyde emissions 1.9 – 3.5 times, and a 35% increase in RH can increase the emissions by a factor of 1.8 – 2.6 . Linear regression models were built in which the natural log of emission factors was the dependent variable and the natural log of RH and inverse of temperature served as the independent variables. The coefficient of inverse temperature was found to be in agreement with values previously reported in literature. Most of the available literature on temperature and RH effects on formaldehyde emissions was reported before 1990. A comparison of temperature coefficients calculated from this study with previously reported values also establishes that there has not been any significant change in the way composite wood surface materials respond to increases in temperature. The experiments were limited to a few samples from the THUs. However, the effects of temperature and humidity reported in this study could be incorporated into an exposure analysis for occupants of THUs.

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About the Authors

Michael Apte is a staff scientist, Randy Maddalena is a scientist, and Marion Russell and Srinandini Parthasarathy are senior research associates in the Environmental Energy Technologies Division of Lawrence Berkeley National Laboratory. Please address correspondence to: Michael Apte, Lawrence Berkeley National Laboratory, Indoor Environment Department MS 90-3058, 1 Cyclotron Road, Berkeley, CA 94720; phone: +1-510-486-4669; fax: +1-510-486-6658; e-mail: mgaite@lbl.gov.