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Publication Date
2010-05-31

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April 2010

This work was supported by interagency agreement 08FED894632 by the National Center for Environmental Health (NCEH) of the Centers for Disease Control and Prevention (CDC) and the Assistant Secretary for Energy Efficiency and Renewable Energy, Building Technologies Programs of the U.S. Department of Energy (DOE) under Contract DE-AC02-05CH11231.

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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. Based on a previous study [1, 2], 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 natural log of percentage relative humidity (RH) and inverse of temperature (in K) as predictor variables, and natural log of emission factors as the target variable. The coefficients of both inverse temperature and log relative humidity with log emission factor were found to be statistically significant for all the samples at the 95% confidence level. This study should assist to retrospectively estimate indoor formaldehyde exposures of occupants of temporary housing units (THUs).

IMPLICATIONS

Maddalena et al., [1, 2] 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. The current report addresses the influence of temperature and humidity on the formaldehyde emission factors from individual materials. The information provided in this study can be incorporated into an exposure assessment study for the occupants of the FEMA trailers. However, since 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 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. Measurements reported [3, 4] showed that formaldehyde concentrations observed in both occupied and unoccupied THUs exceeded the National Institute for Occupational Safety and Health (NIOSH) recommended exposure limit (REL) of 0.016 ppm [5], 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., [1, 2] 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 ⅛-inch plywood with a vinyl or PVC skin or simulated wood finish. All units had sheet vinyl flooring, while two of the four trailers also had carpeted areas. All countertops were particle board surfaced with high-pressure laminate. A variety of wood products were used for the sub-floor 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 observed [6], are also emitted from materials. Like formaldehyde, which is a toxic air contaminant [7, 8], many of the VOCs are known to have low odor thresholds, high potency as respiratory irritants, and in some cases carcinogenicity. Based on the previous study [1, 2], 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., [10] 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 on formaldehyde emissions. Zhang et al., [11] conducted chamber experiments to understand the influence of temperature on the partition coefficient and diffusion coefficient and found that the partition coefficient decreases with increase in temperature, and the diffusion coefficient increased with increase in temperature. However, the equilibrium concentration of formaldehyde increased with increase in temperature. Andersen et al., [12] 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. [13] conducted chamber experiments on various materials (ceiling tile, gypsum board, shiplap, plywood, terracotta brick) that release formaldehyde, and reported higher emissions were observed with increases in temperature, humidity or both.

In a literature review, Myers [14] reported that considerable variations existed between different kinds of boards in their response to varying environmental conditions. Myers [14] reports that the temperature coefficients (where log-concentration was the dependent variable) for various types of composite wood materials fall within an approximate two-fold range, as shown in Table 1. Further, humidity coefficients for various types of composite wood materials falls within the range of 0.005 to 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. Myers [14] assumed an exponential relationship between the concentration and the inverse of temperature, based on the Arrhenius equation. Based on the Berge Equation, he assumes a linear relationship between concentration and $(1 + \beta * RH)$, where β is the humidity coefficient, and RH is the relative humidity.

EXPERIMENTAL METHODS

Materials

Surface materials were cut from THUs for analysis to determine material specific formaldehyde emission factors. Based on the previous study [1, 2], four surface materials with dominant formaldehyde emissions in the THUs were selected for analysis in small chambers to determine the effect of temperature and humidity on formaldehyde emission factors. Samples selected for the analysis were previously tested for emissions under 23 ± 1° C in a controlled environmental chamber with a $0.06\ m^3\ hr^{-1}$ inlet flow of carbon filtered preconditioned air at 50% ± 5% relative humidity. These materials included a sub floor ($416\ \mu g\ m^{-2}\ hr^{-1}$) and cabinet wall ($488\ \mu g\ m^{-2}\ hr^{-1}$) from Gulfstream Coach Cavalier manufactured March 2006, a benchseat ($233\ \mu g\ m^{-2}\ hr^{-1}$) from Coachmen's Spirit of America manufactured October 2006, and a cabinet wall ($419\ \mu g\ m^{-2}\ hr^{-1}$) from Pilgrim International manufactured 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, it tends to adsorb or desorb moisture based on 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 Gulfstream Coach Cavalier's subfloor (3” x 6” x 3/8”) and Pilgrim International's cabinet wall (3” x 6” x 1/8”) 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 the experiments. Each material was cut in half, and the two samples were placed in chambers held at identical temperature and relative humidities of 50% and 85%. The temperature and relative humidities at which the experiments were carried out are listed in Table 2. A schematic diagram of the experimental set-up is provided in Figure 1. The samples were weighed using a semi-micro analytical balance Ohaus model DV314C at the start of the experiment and weighed once daily till the weight of the sample reached a constant value.

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 in 50° C oven overnight. The backing plates were taped to the back of the samples using Scotch 3M Metal Repair tape. The two samples from each material were placed in chambers held at identical temperature and relative humidities of 50% and 85%. The experiments were carried out at the various temperature and relative humidity conditions specified in Table 3. The samples were placed in the chamber for an average of 1 hour 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-dinitrophenyl-hydrazine (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 65%:35% H_2O:Acetonitrile mobile phase at 0.35 mL/minute and UV 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.

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 equation for calculating area-specific emission factors, EF, (µg m^{-2} hr^{-1}) in a well-mixed system is

$$EF = \frac{f \times (C - C_0)}{A} \qquad (1)$$

where, f (m^3 hr^{-1}) is the ventilation rate, A (m^2) is the exposed surface area of the sample, C (µg m^{-3}) is the measured steady-state concentration in the chamber, and C (µg m^{-3}) is the background concentration in the chamber.

RESULTS AND DISCUSSION

Figures 2 a and b show the results from the humidity equilibration experiments for the cabinet wall and subfloor samples. The goal of these experiments was to estimate the time for the samples to equilibrate with the environmental conditions. As shown by Figures 2 a and b, the samples take about 40 hours to equilibrate. However, under the 85% RH, 288 K conditions the samples take about 240 hours 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 relative humidity have a strong positive correlation with the emission factors for all 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 correlation coefficient between relative humidity and emission factor for all samples was found to be greater than 0.98. Poor correlation was found between the temperature and relative humidity ($-.05<R^2<.03$).

A linear regression model was built setting natural log of emission factors as the target variable. Natural log of percentage relative humidity and inverse of temperature (in K) were used the predictor variables. The coefficients of both inverse temperature and log relative humidity with log emission factor were found to be statistically significant for all the samples at the 95% confidence level, as shown in Table 4. Figures 3 a to d 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 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 the concentration and (1 + β*RH), based on the Berge Equation, where β is the humidity coefficient, and RH is the relative humidity [14]. However, this study assumes a linear relationship between emission rate and relative humidity and therefore a direct comparison of the humidity coefficients generated in the current study with previous work is not possible. Myers [14] assumed an exponential relationship between the concentration and the inverse of temperature, and this study assumes an exponential relationship between the emission rate and 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 literature. Myers [14] states that the temperature coefficients reported for various types of composite wood materials fall in the range of -11120 to -5620 (K). The temperature coefficients estimated in this study for particle board falls within this range. Additionally, Myers [14] 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 while 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 0.1hr^{-1}. The study is limited to observing the effects of temperature and relative humidity 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 relative humidity can increase the emissions by a factor of 1.8 – 2.6. Linear regression models were built in which natural log of emission factors was the dependent variable and natural log of relative humidity and inverse of temperature served as the predictor 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 relative humidity effects on formaldehyde emissions was reported prior to 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 small number of 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.

ACKNOWLEDGEMENTS

This work was supported by interagency agreement 08FED894632 by the National Center for Environmental Health (NCEH) of the Centers for Disease Control and Prevention (CDC) and the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Building Technology, State, and Community Programs of the U.S. Department of Energy (DOE) under Contract DE-AC02-05CH11231. The authors would like to thank William Fisk and Mohamad Sleiman for their reviews of this paper.

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TABLES

Table 1 : Temperature and Humidity Coefficients ranges reported in literature

Material	Temperature coefficient (K)		Humidity coefficient (log RH^{-1})	
	Minimum	Maximum	Minimum	Maximum
Particle board	-11120	-5620	0.005	0.038
Plywood	-9600	-7430	0.006	0.033

Source : Myers (1985)

Table 2: Humidity Equilibration Experiments

Sample Number	Sample	Material	Temperature (K)			Relative Humidity (%)	
			288 ± 1	298 ± 1	308 ± 1	50 ± 2	85 ± 9
1	Cabinet Wall	Plywood	X			X	
2	Cabinet Wall			X		X	
3	Cabinet Wall				X	X	
4	Cabinet Wall		X				X
5	Cabinet Wall			X			X
6	Cabinet Wall				X		X
7	Subfloor	Particle board	X			X	
8	Subfloor			X		X	
9	Subfloor				X	X	
10	Subfloor		X				X
11	Subfloor			X			X
12	Subfloor				X		X

Table 3: Overview of Experiments and Measured Steady-State Concentrations of Formaldehyde in Chamber Experiments

Sample Number	Sample	Temperature (K)			Relative Humidity (%)		Sample Dimensions			Ventilation Rate	Concentration (µg m^{-3})	Emission Factor (µg m^{-2} hr^{-1})
		288 ± 1	298 ± 1	308 ± 1	50 ± 1	85 ± 3	Length (cm)	Breadth (cm)	Area Exposed (m^2)	(L min^{-1})		
1	Bench Seat	X			X		14.2	6.5	0.009	1.0	9.9	62
2	Bench Seat		X		X					0.9	21	130
3	Bench Seat			X	X					1.0	43	270
4	Bench Seat	X				X	13.8	6.6	0.009	1.0	20	130
5	Bench Seat		X			X				1.0	47	300
6	Bench Seat			X		X				1.0	110	670
7	Cabinet	X			X		13.1	6.0	0.008	0.9	9.9	70
8	Cabinet		X		X					0.9	20	140
9	Cabinet			X	X					0.9	55	390
10	Cabinet	X				X	13.2	6.2	0.008	1.0	15	100
11	Cabinet		X			X				1.0	47	320
12	Cabinet			X		X				1.0	140	1000
13	Cabinet Wall	X			X		14.1	6.6	0.009	1.0	9.4	60
14	Cabinet Wall		X		X					1.0	17	110
15	Cabinet Wall			X	X					1.0	45	280
16	Cabinet Wall	X				X	14.0	6.5	0.009	1.0	18	110
17	Cabinet Wall		X			X				1.0	40	260
18	Cabinet Wall			X		X				1.0	100	640
19	Subfloor	X			X		14.0	6.5	0.009	0.9	15	100
20	Subfloor		X		X					0.9	44	270
21	Subfloor			X	X					0.9	120	770
22	Subfloor	X				X	14.2	7.0	0.010	1.0	24	140
23	Subfloor		X			X				1.0	85	480
24	Subfloor			X		X				0.9	270	1600

Note: 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.

Table 4: Linear Regression Modeling Results

	Coefficients	Confidence Limits		Regression Statistics	
Benchseat		**Lower**	**Upper**	**R^2**	**p**
Inverse Temperature Coefficient (K)	-6740	-7640	-5840	0.996	0.0002
log RH Coefficient (log RH^{-1})	1.546	1.225	1.866		0.0006
Intercept	21.4	18.1	24.7		--
Cabinet					
Inverse Temperature Coefficient (K)	-8500	-11100	-5940	0.979	0.002
log RH Coefficient (log RH^{-1})	1.468	0.569	2.366		0.01
Intercept	27.8	18.0	37.0		--
Cabinet Wall					
Inverse Temperature Coefficient (K)	-7030	-8660	-5390	0.988	0.0008
log RH Coefficient (log RH^{-1})	1.421	0.813	2.028		0.005
Intercept	22.9	16.8	28.9		--
Subfloor					
Inverse Temperature Coefficient (K)	-9940	-11500	-8400	0.994	0.0003
log RH Coefficient (log RH^{-1})	1.166	0.625	1.708		0.006
Intercept	34.4	29.0	40.0		--

FIGURES

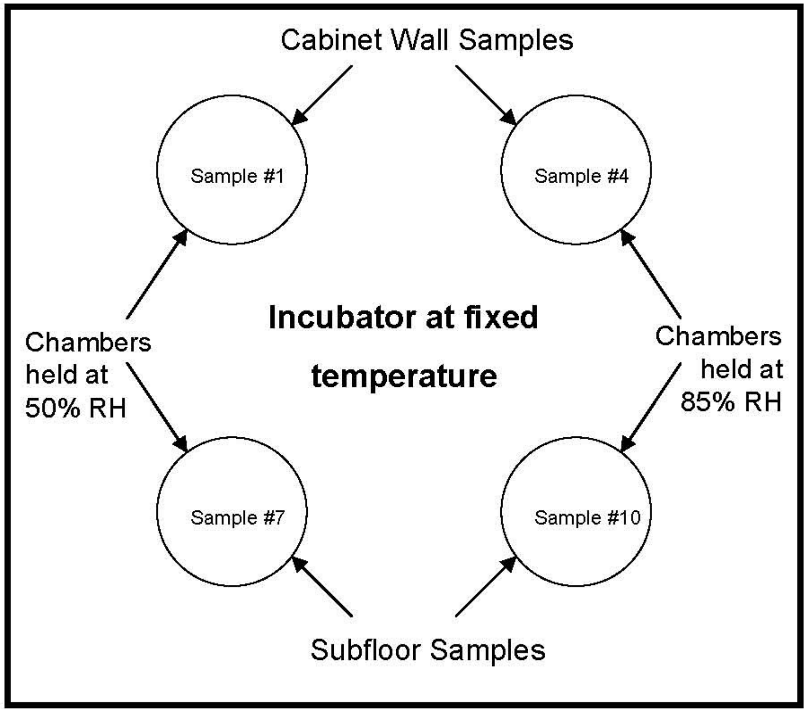


Figure 1. Schematic diagram of experimental set-up Humidity Equilibration Experiments

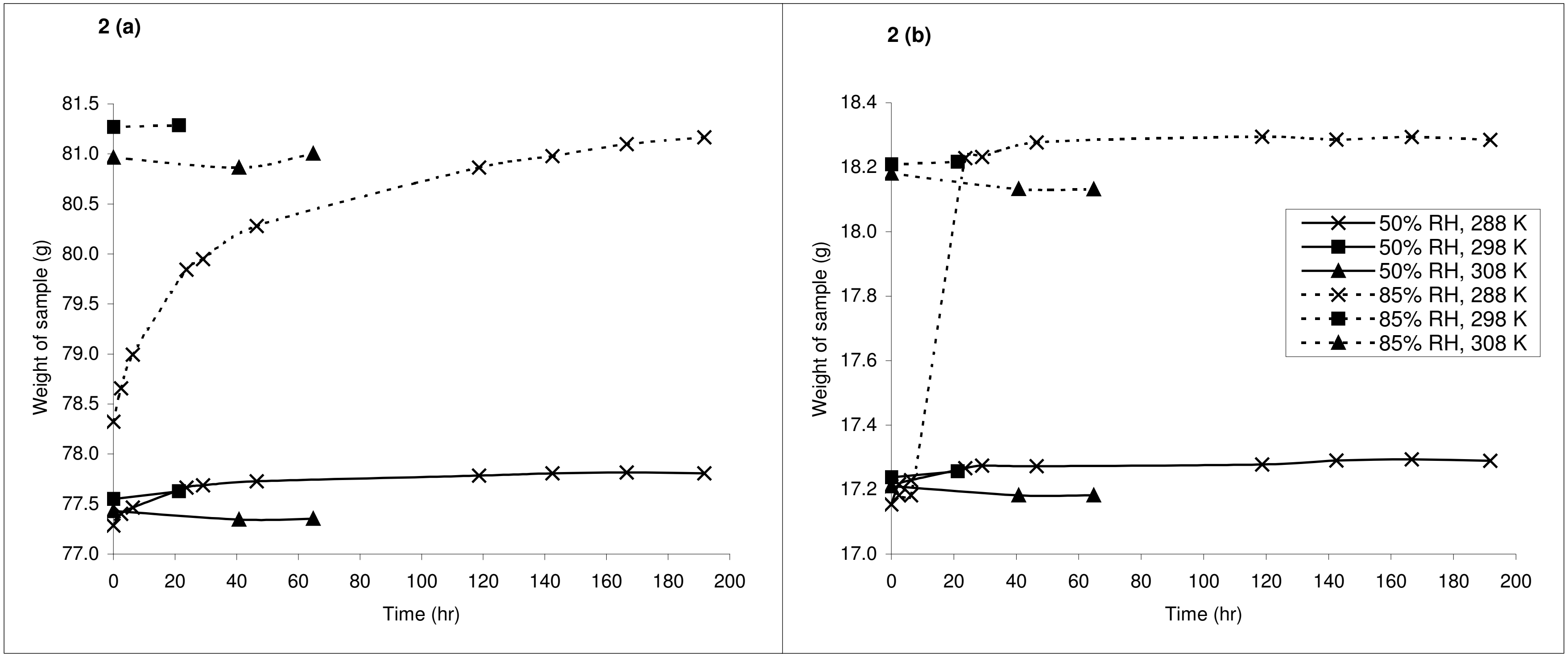
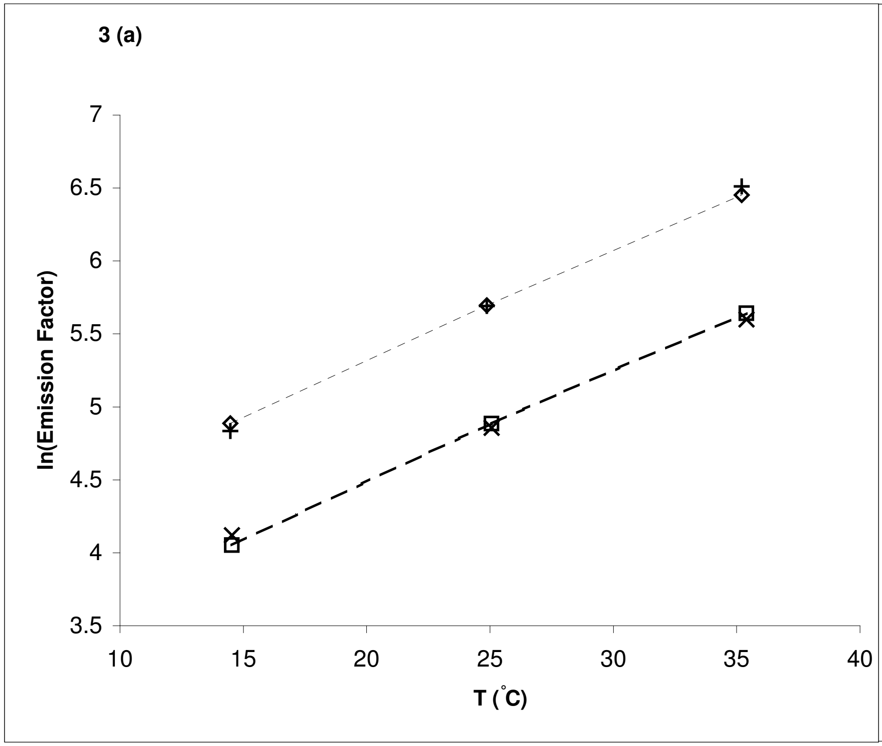


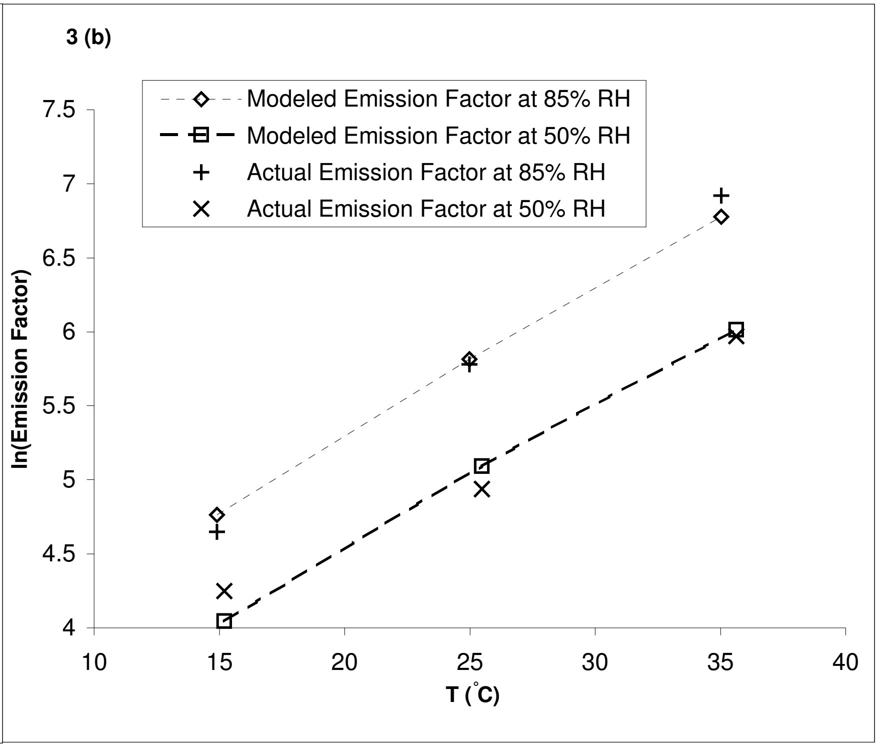
Figure 2. Results from Humidity Equilibration Experiments: (a) Cabinet Wall sample (b) Subfloor sample

3 (a)



ln(Emission Factor)
7
6.5
6
5.5
5
4.5
4
3.5
10
15
20
25
30
35
40
T (˚C)

3 (b)



Modeled Emission Factor at 85% RH
Modeled Emission Factor at 50% RH
Actual Emission Factor at 85% RH
Actual Emission Factor at 50% RH
ln(Emission Factor)
7.5
7
6.5
6
5.5
5
4.5
4
10
15
20
25
30
35
40
T (˚C)

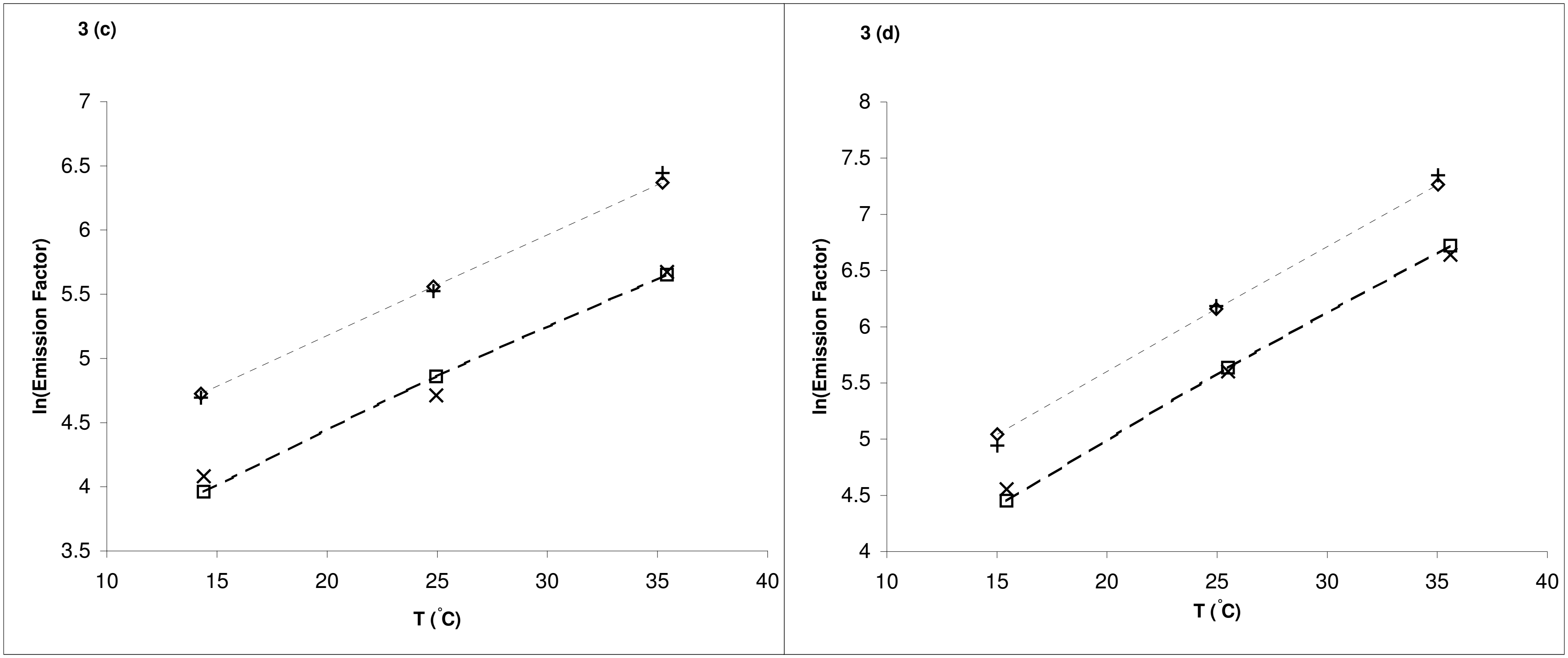


Figure 3. Arrhenius plot of emission factors as a function of temperature and relative humidity: (a) Benchseat sample (b) Cabinet sample (c) Cabinet Wall sample (d) Subfloor sample

FIGURE CAPTIONS

1. Schematic diagram of experimental set-up Humidity Equilibration Experiments
2. Results from Humidity Equilibration Experiments: (a) Cabinet Wall sample (b) Subfloor sample
3. Arrhenius plot of emission factors as a function of temperature and relative humidity: (a) Benchseat sample (b) Cabinet sample (c) Cabinet Wall sample (d) Subfloor sample