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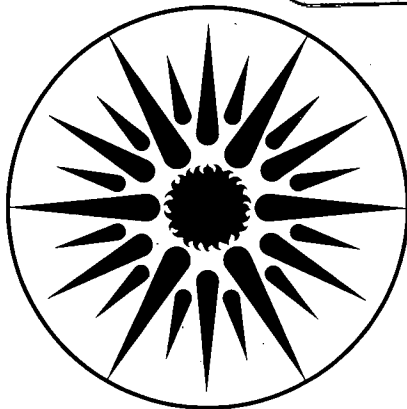
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SOURCES AND CONCENTRATIONS OF FORMALDEHYDE IN INDOOR ENVIRONMENTS

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June 1983

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ABSTRACT

Formaldehyde is used as an inexpensive chemical component of resins commonly used in many building materials. As is now well known, the formaldehyde in these resins can be released from building materials into the indoor environment. In addition, unvented combustion appliances such as gas ranges and heaters emit formaldehyde as a product of incomplete combustion. In this study, we summarize formaldehyde emission rates from a variety of combustion appliances and the formaldehyde concentrations observed in forty residential indoor environments. If the combustion appliance is well tuned, it appears that high formaldehyde concentrations are not produced under most conditions of use. Energy-efficient houses were generally found to have higher concentrations than those observed in weatherized houses, with about a third being above the ASHRAE guideline, 100 ppb. The ventilation rate was shown to have a strong effect on formaldehyde concentrations in about half the houses studied when the ventilation was varied but had little effect in other cases. This anomaly should be studied further.

Keywords: combustion appliance, emission rate, energy-efficiency, formaldehyde, gas range, heat exchanger, kerosene heater, ventilation, weatherization.

INTRODUCTION

Formaldehyde is an inexpensive chemical used extensively in a variety of products, principally in urea, phenolic, melamine and acetal resins. These resins are widely used in building materials such as insulation, particleboard, plywood, textiles, and adhesives. Use of products containing formaldehyde-containing resins can result in the release of formaldehyde into the indoor environment. In addition, unvented combustion appliances, such as gas-fired ranges and heaters, and tobacco smoking emit formaldehyde as a product of incomplete combustion.

Several studies have indicated that exposure to even relatively low concentrations of formaldehyde can produce allergenic symptoms such as burning of the eyes and irritation of the upper respiratory passages¹. Individual response to formaldehyde varies widely; some individuals become increasingly sensitive to it as a result of continued exposure. In addition, concern exists that formaldehyde may have serious long-term health effects. As a result, several foreign countries and states in the United States are moving to establish formaldehyde standards for indoor air.²

In this paper we review the formaldehyde emission rates obtained from a variety of combustion appliances in laboratory studies at Lawrence Berkeley Laboratory (LBL) and report on formaldehyde concentrations we have observed in a variety of residential indoor environments.

COMBUSTION APPLIANCES

We have conducted several studies, in the laboratory and in the field, to measure pollutant emissions from unvented combustion appliances. In Table 1, the formaldehyde emission rates determined from laboratory testing of a gas stove, unvented gas-fired space heaters, and portable kerosene-fired space heaters are listed.^{3,4,5}

Gas stoves are the most common unvented combustion appliance in general use and the emission rates for the oven and top burner are the highest of all rates listed in Table 1. However, the contribution of gas stoves to total formaldehyde exposures in houses may not be as large as that from some other appliances. The formaldehyde source strengths (the mass of pollutant emitted per unit time) from gas stoves, both oven and top burner, are not very large because the fuel consumption rates are fairly low. In addition, gas stove use is intermittent which further decreases their contribution to indoor formaldehyde concentrations.

Unvented gas-fired space heaters are most commonly used in the southern and southwestern states. The average formaldehyde emission rate, 0.81 $\mu\text{g}/\text{kJ}$, from these heaters is lower than those associated with gas ranges but, as is indicated by the range of emission rates observed for individual heaters (0.43 to 4.2 $\mu\text{g}/\text{kJ}$), the emission rates of individual heaters can be much higher. Moreover, the fuel consumption rates for these space heaters are generally higher than those of a gas stove so the source strength can also be much higher. In addition, it is likely that they are operated for much longer periods of time. The combination of high source strengths and duration of use suggest that the

use of this type of appliance is a more important source of formaldehyde.

Calculations based upon these emission rates have indicated that formaldehyde concentrations resulting from the steady-state operation of unvented gas-fired space heaters would generally be less than 100 ppb in a well ventilated (one air change per hour), average-sized house (1400 ft²).

However, formaldehyde emissions from these space heaters are sensitive to the air/fuel ratio as controlled by the air shutter adjustment. The previous estimate assumes that the unvented heater is well tuned with respect to the air shutter adjustment. A poorly tuned unvented heater can emit significant amounts of formaldehyde. Under conditions similar to those outlined above, a poorly tuned unvented gas-fired space heater produced formaldehyde concentrations in excess of 1 ppm in a controlled field study.⁶ (At this time, however, the frequency at which poor tuning of combustion appliances occurs is unknown.)

As also indicated in Table 1, formaldehyde emission rates from portable kerosene space heaters are lower than those of the other combustion appliances tested and the fuel consumption rates are low as well, indicating that formaldehyde concentrations from these kerosene heaters are not expected to occur at problem levels.

In summary, formaldehyde source strengths from well tuned, unvented combustion appliances appear of a size that can exacerbate an existing formaldehyde problem or in combination with other sources, contribute to high formaldehyde concentrations. However, formaldehyde emissions from

these appliances, in the absence of other strong sources, are not likely to be a problem under most conditions of use unless the appliance is poorly tuned or malfunctioning. (This is not to imply that emissions of pollutants other than formaldehyde do not occur at problem levels from these appliances.)^{3,4,5,6}

RESIDENTIAL CONCENTRATIONS

Several field studies have been conducted by the Indoor Air Quality Group at LBL to measure concentrations of pollutants in residences in several areas of the U.S. A summary of the formaldehyde concentrations measured in these studies is given in Table 2.2,7-16. These data were obtained through the use of refrigerated pump/bubbler samplers.¹⁷ The limited data reflects, in part, the expense and difficulty of obtaining formaldehyde concentrations by employing refrigerated pump/bubbler samplers in the field. (This difficulty and expense will soon be overcome by the use of the recently developed passive samplers for formaldehyde.)¹⁶ With the exception of two studies of office buildings, the buildings sampled were either relatively new energy-efficient houses or older houses that were weatherized. As well as using large amounts of insulation, energy-efficient houses usually incorporate measures that seal the building envelope, such as vapor barriers, improved window and door seals, and caulking of the plumbing and wiring penetrations of the envelope. These measures reduce the infiltration rates of houses, and thereby reduce space heating and cooling requirements. Houses that are weatherized are typically older houses that are retrofitted, usually with increased insulation and with measures reducing infiltration such as caulking and weatherstripping. In several of these studies involving

energy-efficient houses, mechanical ventilation incorporating air-to-air heat exchangers was used to increase the ventilation rates while minimizing energy costs.

Figure 1 shows a frequency distribution of the maximum formaldehyde concentrations observed in 40 energy-efficient or weatherized houses. The formaldehyde concentrations reported are time-weighted averages (weekly, 12-, or 24-hour averages). The distribution is approximately lognormal with a geometric mean of 49.5 ppb and a geometric standard deviation of 2.09. Reported maximum concentrations are used in this distribution since most proposed or promulgated standards for formaldehyde in indoor air are maximum or ceiling values.

Figure 2 compares the formaldehyde frequency distributions for weatherized (geometric mean, 28 ppb; geometric standard deviation, 2.00) houses and for energy-efficient houses (geometric mean, 64 ppb; geometric standard deviation, 2.18). The range of formaldehyde concentrations in weatherized houses extends to just below the most stringent proposed or promulgated standard for indoor formaldehyde (100 ppb),¹⁸ whereas, about one third of the energy-efficient houses exceed this limit. Because of the small sample size, any conclusions drawn from this distribution can only be tentative. However, this figure suggests that the actual distributions may differ, with the energy-efficient houses having higher formaldehyde concentrations. This is consistent with the observations that (1) the energy-efficient houses are not as old as the weatherized houses, being for the most part less than two years old (and formaldehyde emissions from building materials decrease with aging of the materials)¹⁹ and (2) they are generally tighter than

weatherized houses so that less fresh air is available to dilute and flush out formaldehyde.

In general, as shown in Table 3, it appears that energy-efficient and weatherized houses, as a group, may have lower formaldehyde concentrations than has been observed in other classes of houses with potential for high formaldehyde concentrations, e.g., mobile homes and urea-formaldehyde - foam insulated houses. We suggest that this may be due to two reasons. Much of the sampling done within these other classes of housing has been performed in response to occupant complaints and sampling may be predominately occurring in houses with higher formaldehyde concentrations. A second reason may be that energy-efficient houses and almost certainly weatherized houses have lower formaldehyde source strengths in relationship to other factors affecting concentrations (such as surface-to-volume ratio and ventilation rate).

Looking at the range of concentrations observed in groups of houses can provide a certain type of information, but examining the variation of concentrations occurring in a particular house can provide detail in understanding the contributions of specific sources to formaldehyde concentrations. Table 4, for example, presents the data from an energy-efficient house in Mission Viejo, California.⁹ Initially the house was unoccupied and unfurnished. Under these conditions, the formaldehyde concentration was moderate, indicating that the contribution of building materials to indoor formaldehyde concentrations was low. When furniture was brought into the house, the formaldehyde concentration almost tripled. A further increase may have occurred when the house was occupied, and this increase may have been directly due to occupant activities such

as using a gas stove, or indirectly by increasing the humidity and thereby increasing formaldehyde offgassing from building materials.²⁰ However, even if this increase due to occupant activity were real, it is small compared to the increase observed with the addition of furniture.

The effect of ventilation on formaldehyde concentrations is shown in Table 4. The formaldehyde concentration in this house appeared to be strongly affected by changes in ventilation caused by opening windows. However, diurnal variations in temperature and humidity rather than opening windows may have caused the changes observed in the formaldehyde concentration, since, for example, in one of the houses sampled in Eugene, Oregon, the formaldehyde concentration did not seem to respond to any ventilation changes caused by opening windows and doors.¹⁴

The effect of ventilation on formaldehyde concentrations is worth examining in more detail. In several studies where formaldehyde was sampled, the ventilation rate of houses changed, either decreasing because the house was weatherized or increasing because mechanical ventilation was employed.^{11,12,15} In these studies, formaldehyde concentrations were measured while the house was in the initial state, i.e., prior to weatherization or prior to the use of mechanical ventilation. After weatherization or while mechanical ventilation was in continuous use, the final formaldehyde concentrations were measured. Table 5 lists the initial and final formaldehyde concentrations observed along with the initial and final ventilation rates.

To assess the effect of ventilation on formaldehyde concentrations, we calculated a "predicted" final formaldehyde concentration based upon the initial formaldehyde concentrations measured, the initial and final

air-exchange rates and a reactive decay constant for formaldehyde of 0.4 h^{-1} . The value of 0.4 h^{-1} was measured in studies conducted at LBL using a 27-m^3 chamber.³ For the purpose of these calculations, we will assume it applies to other environments as well. Implicit in these calculations is the assumption that the formaldehyde source strength is constant during the initial and final sampling periods. The results of these calculations are listed in Table 5 and illustrated in Figure 3.

In about half the studies listed, the predicted and observed final concentrations agree well (especially in view of the fact that the measurement technique has a precision of about 10-15%), and it appears that ventilation has a significant effect on formaldehyde concentrations. In the other studies listed, however, it is just as obvious that ventilation is not the dominant factor, and perhaps changes in other factors such as relative humidity or temperature are more important. In these studies the observed changes are considerably different from the predicted changes. It should also be noted that in three cases where the disagreement between predicted and observed concentrations occurs, it is because the final concentration differed very little from the initial concentration. Reasons for this lack of change should be examined in future studies.

SUMMARY

We have presented a summary of formaldehyde emission rates measured in our laboratory studies of unvented combustion appliances, and we have reviewed the formaldehyde concentrations actually measured in indoor environments, predominately in energy-efficient and weatherized houses. It appears that as long as the unvented combustion appliance is well

tuned, formaldehyde emissions, under most conditions of use, are of such a size that operation of these appliances would not create high formaldehyde concentrations. However, high concentrations can be produced by poorly tuned appliances.

The formaldehyde concentrations observed in weatherized houses were below the most stringent proposed or promulgated indoor standard of 100 ppb. In energy-efficient houses, however, formaldehyde concentrations were generally higher -- in many cases exceeding 100 ppb. The sample size in both groups of houses was small, and firm conclusions regarding concentration distributions cannot be drawn. However, studies with sufficient data to establish formaldehyde concentration distributions by housing type will become practical through the use of recently developed passive samplers for formaldehyde.

While in some studies, the ventilation rate has been shown to be a strong factor affecting formaldehyde concentrations, in others, it is not a dominant factor. This anomaly demonstrates the need for detailed studies where formaldehyde is measured continuously over short-term intervals in occupied houses. Such studies should investigate the effects of temperature, relative humidity, ventilation, and occupant activities on formaldehyde concentrations and produce a comprehensive inventory of potential formaldehyde sources. Studies of this type should be conducted over sufficient periods of time so that variations due to aging of building materials and those due to seasonal changes can also be evaluated.

ACKNOWLEDGEMENT

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Table 1. Formaldehyde Emission Rates from Unvented Combustion Appliances.

Appliance	No. of Appliances Tested	Average Emission Rate ($\mu\text{g}/\text{kJ}$)	Range of Emission Rates ($\mu\text{g}/\text{kJ}$)	Fuel Consumption (kJ/h)
Gas Stove ^a	1			
Oven		2.7	2.4-3.4	8,400
Top Burner		1.7	0.86-2.5	9,200
Gas Space Heater ^b				
Well Tuned	8	0.81	0.43-4.2	10,100-44,700
Poorly Tuned	2	1.5	0.46-20.3	33,600-43,900
Kerosene Space Heater ^c	4	0.47	0.01-1.22	4,230-8,250

^aTraynor et al. [3]

^bGirman et al. [4]

^cTraynor et al. [5]

Table 2. Summary of Formaldehyde Concentrations in Indoor Environments.

Location	Building Type ^a	No. Buildings	Formaldehyde Range (ppb)	Text Reference
Ames, IA	energy-efficient	1	28-61	7
Carroll Co. MD	energy-efficient	1	44-148	7
Berkeley, CA	office trailers	3	20-185	8
Mission Viejo, CA	energy-efficient	1	66-214	9
San Francisco, CA	office-building	1	41	10
Medford, OR	conventional, retrofitted	2	51-68	11
Midway, WA	conventional, retrofitted	12	<5-79	2
Northfield, MN	energy-efficient, heat exchanger	1	69-73	12
Dundas, MN	energy-efficient, mechanical ventilation	1	64-80	12
Rio, WI	conventional, retrofitted	1	53	12
Cranbury, NJ	>100 yrs, retrofitted	1	19-22	13
Eugene, OR	energy-efficient	2	37-73	14
	energy-efficient, passive solar	2	82-112	
Rochester, NY	energy-efficient, mechanical ventilation	10	<5-64	15
Sacramento, CA	energy-efficient, passive solar	5	98-127	16 ^b

^aAll buildings are houses unless otherwise indicated.

^bSamples S-6, S-10, S-15, S-16, and S-17.

Table 3. Summary of Formaldehyde Measurements in Various Indoor Environments.

Sampling Site	Concentration (ppm)	
	Range	Mean
Two mobile homes in Pittsburg, Pennsylvania	0.1-0.8 ^a	0.36
Mobile homes registering complaints in State of Washington	0-1.77	0.1-0.44
Mobile homes registering complaints in Minnesota	0-3.0	0.4
Mobile homes registering complaints in Wisconsin	0.02-4.2	0.88
Public buildings and energy-efficient homes, occupied and unoccupied	0-0.21 0-0.23 ^a	— —

^aTotal aliphatic aldehydes.

Source: National Research Council, National Academy of Sciences [1].

Table 4. Indoor Formaldehyde Concentrations in a New Residential Building in Mission Viejo, CA

Condition	Formaldehyde (ppb)
Unoccupied, without furniture ^a	67 ± 9%
Unoccupied, with furniture ^a	186 ± 7%
Occupied, day ^a	218 ± 10%
Occupied, night ^b	117 ± 31%
Outdoor Air	<17

^aAir exchange rate $\cong 0.4 \text{ h}^{-1}$

^bWindows open part of time; air exchange rate significantly greater than 0.4 h^{-1} and variable.

Source: Berk et al. [9].

Table 5. Predicted and Observed Indoor Formaldehyde Concentrations in Residential Environments with Changed Ventilation Rates.

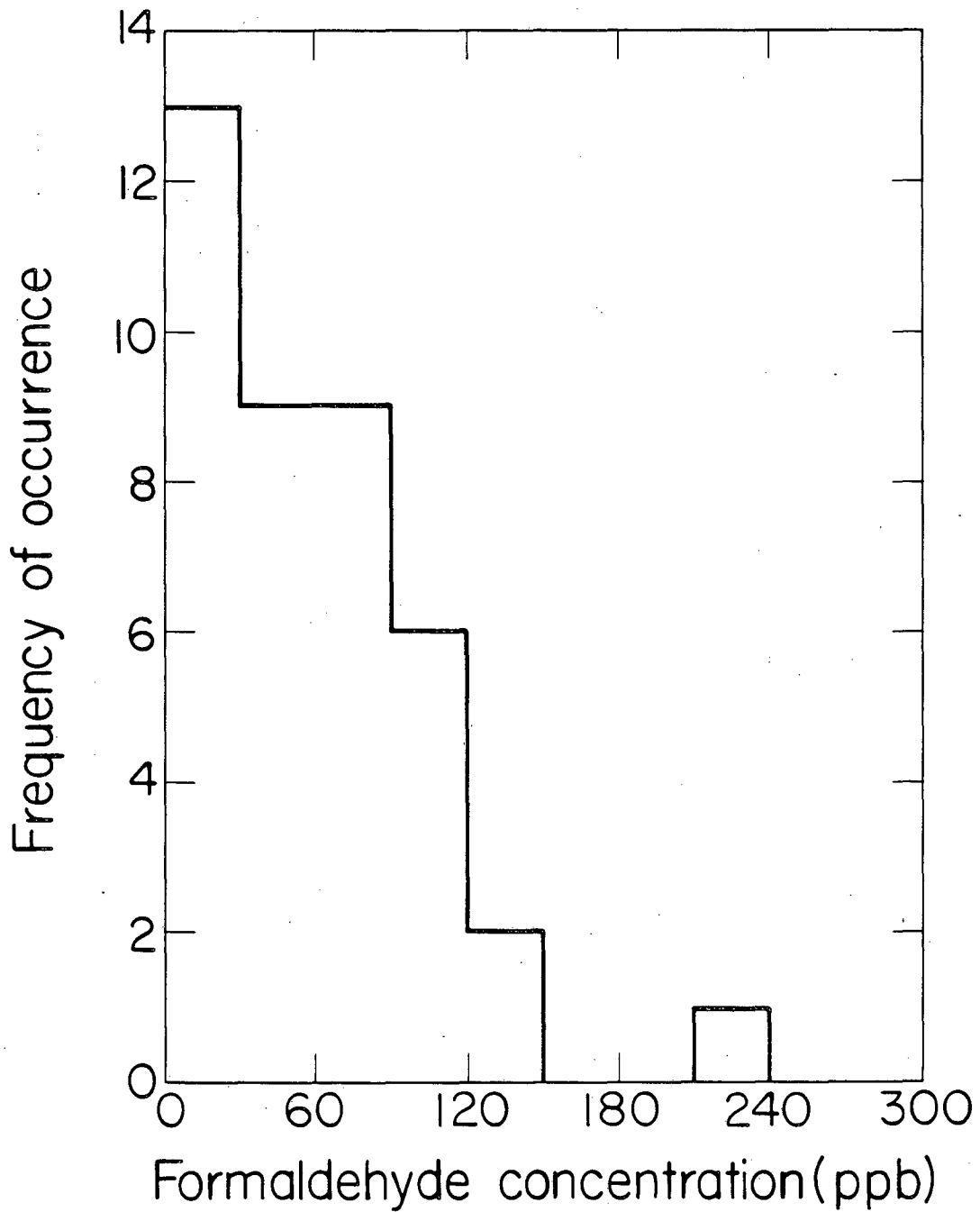
Location [Ref.] ^a	Ventilation Rate (ach)		Formaldehyde Concentration (ppb)			Concentration Change (%)	
	Initial	Final	Initial	Predicted ^b Final	Observed Final	Predicted	Observed
Medford, OR [11]	0.48 ^c	0.35 ^c	55	65	53 ^d	18	-4 ^d
	0.57 ^c	0.41 ^c	68	81	51 ^d	19	-25 ^d
Northfield, MN [12]	0.1	0.3	69	52	73 ^d	-24	6 ^d
Dundas, MN [12]	0.1	0.3	80	57	64	-29	-20
Rochester, NY [15]	0.22	0.47	36	26	19	-28	-47
	0.38	0.66	29	21	22	-28	-24
	0.30	0.61	7	5	<5	-29	>29
	0.38	0.78	33	22	19	-33	-42
	0.42	0.64	30	24	29	-20	-3
	0.33	0.52	57	45	42	-21	-26

^aReferences keyed to text.

^bCalculated assuming a constant formaldehyde source strength, zero outside formaldehyde concentration, and a reactive decay constant of 0.4 h^{-1} .

^cAverage of infiltration rates measured with furnace fan on and off.

^dObserved formaldehyde concentration change was opposite predicted direction.



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Figure 1. Frequency distribution of maximum formaldehyde concentrations in energy-efficient and weatherized houses.

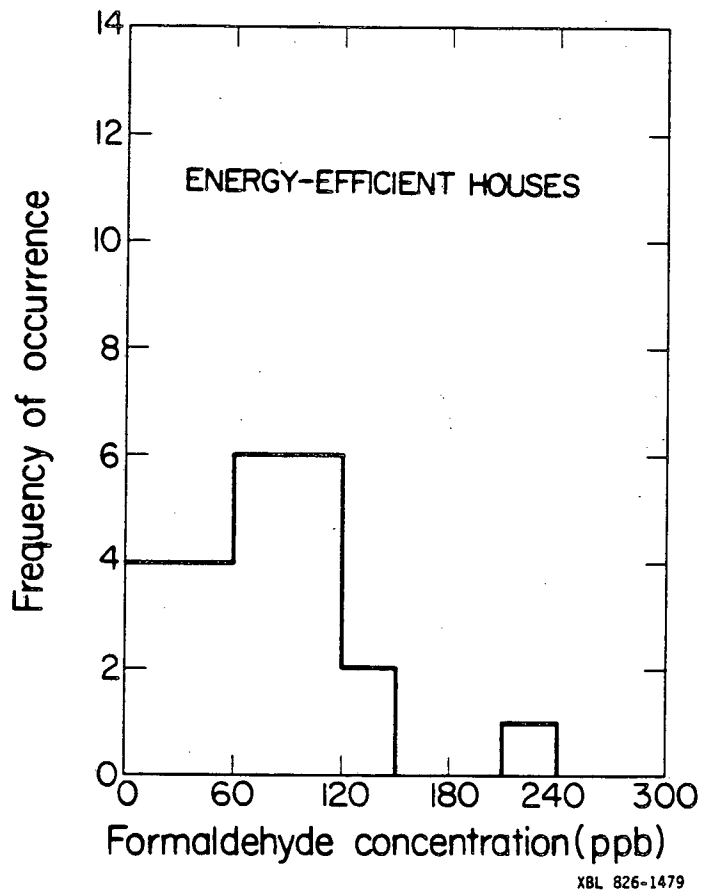
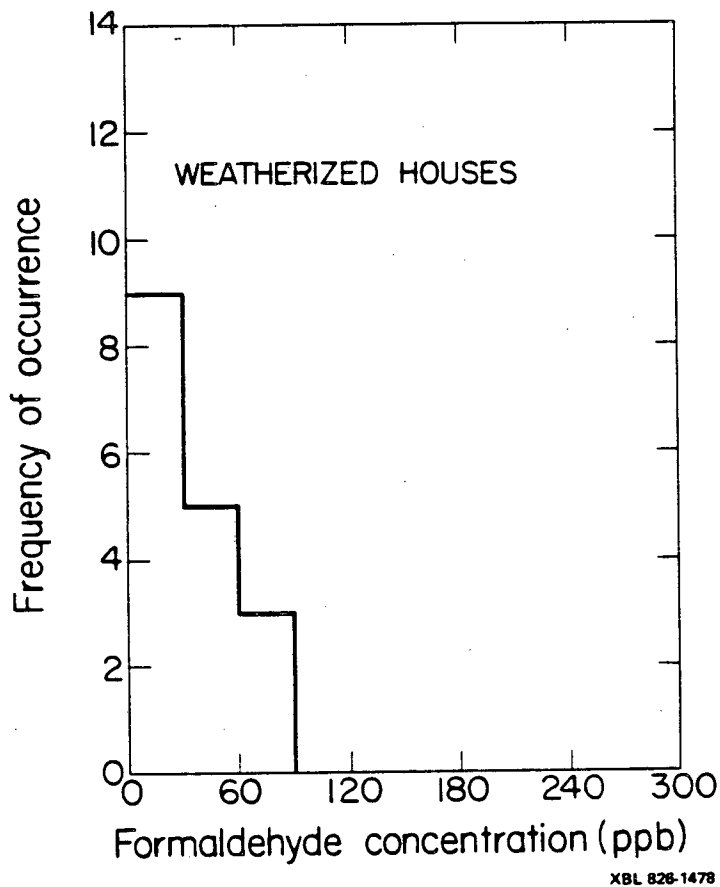


Figure 2. Comparison of frequency distributions of maximum formaldehyde concentrations between weatherized and energy-efficient houses.

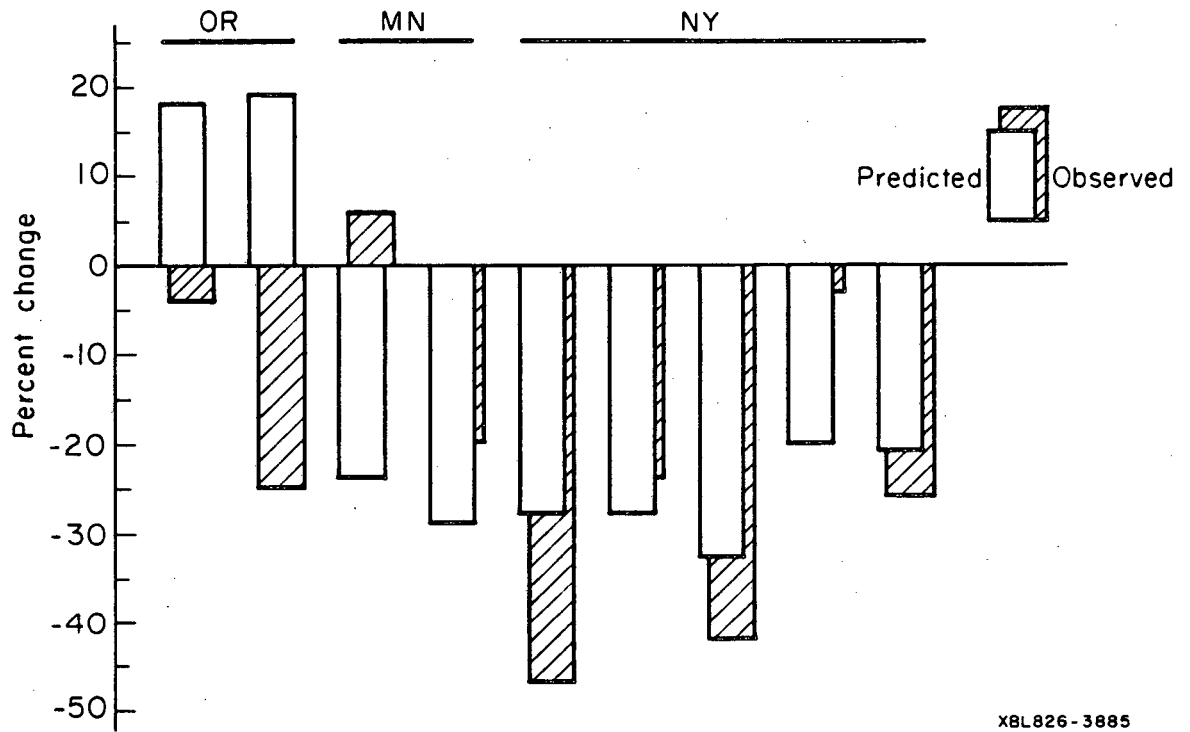


Figure 3. Predicted and observed changes in formaldehyde concentrations with changes in ventilation.

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