FORMALDEHYDE EXPOSURE IN RESIDENTIAL SETTINGS: SOURCES, LEVELS AND EFFECTIVENESS OF CONTROL OPTIONS (INTERIM FINAL REPORT) (EPA CONTRACT #68-02-3968)

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Interim Final Report

Formaldehyde Exposure in Residential Settings: Sources, Levels, and Effectiveness of Control Options

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EXECUTIVE SUMMARY

The U.S. Environmental Protection Agency (EPA) is reexamining existing information on residential exposure to formaldehyde released from pressed-wood products containing urea-formaldehyde (UF) resins. This report addresses two general topic areas:

Current levels of exposure to formaldehyde in housing, the source of that exposure, and factors that affect these levels.

Reduction in exposure levels that could result from implementation of measures to control formaldehyde emissions from pressed wood products.

Pressed Wood Products Containing UF Resins

Formaldehyde is released from all pressed-wood products containing UF resin. The three types of pressed-wood products formulated with UF resin are particleboard, medium density fiberboard (MDF), and hardwood plywood.

Particleboard is composition board comprised of 6 to 10 percent resin (by weight), and small wood particles; additives are also a small fraction of the board. UF is the resin used for the vast majority of particleboard, though producers accounting for 10 percent of total production in 1983 used other resins (phenol-formaldehyde, 6 percent; isocyanate, 4 percent). Manufacture entails mixing these components and pressing the mixture at elevated temperatures. The 1983 production of particleboard was over 3 billion square feet, of which 70 percent was used in furniture, fixtures, cabinets, and similar products. The remaining 30 percent was used for construction purposes, including mobile home manufacture (common uses are as decking or flooring underlayment). Particleboard is used increasingly as a substitute for whole-wood products. Particleboard is commonly used in mobile home construction at a loading rate of 0.5 square meters per 1 cubic meter of indoor air volume. The results of a recent survey of homebuilders indicate that particleboard, when used in new conventional home construction, is used at an average loading rate of 0.17 m^2/m^3.

Medium-density fiberboard (MDF) is currently made only with UF resin. The resin accounts for 7 to 9 percent of the board weight. Ten companies with 11 plants produced over 600 million square feet in 1983. Approximately 95 percent of that total was turned into doors, furniture, fixtures, and cabinetry; the other 5 percent went into miscellaneous products. MDF differs from particleboard mainly in the character of its wood particles (known as "furnish"). As the name implies, the wood is separated by cooking or shredding into fibers smaller than 1 mm. The resulting pressed-wood product is more homogeneous in texture, appears more like wood, and can be machined. The extent to which MDF is used in housing is uncertain and is probably highly variable.

Unlike the two composition boards discussed above, hardwood plywood is a laminated product; the resin is used as a glue to hold thin layers of wood and veneers together. It contains only 2.5 percent resin (by weight). Nearly 2 billion square feet were produced in 1983; consumption is estimated as 55 percent to indoor paneling, 30 percent to furniture and cabinets, and 15 percent to doors and laminated flooring. A large part of the hardwood plywood used in the U.S. is imported lauan plywood, which is prefinished in the U.S. by a variety of decorative processes.

Causes of Formaldehyde Release from Pressed Wood Products

An understanding of the exposure to formaldehyde releases from pressed-wood products must be based on at least a rudimentary understanding of the chemistry of UF resins. Urea-formaldehyde resins are prepolymers that result from the reaction of excess formaldehyde with urea; additives such as catalysts and waxes can be added to the resin mixture. The product is adjusted to suit its specific end-use (resins used in different products can vary in formaldehyde:urea ratio, among other variables). UF resin is a thermosetting resin, implying that the resin undergoes crosslinking and other changes when subjected to heat

during product manufacture. The heating is called curing in the pressed-wood product business, and curing does change the nature of the resin. It is speculated that ten major types of organic compounds and organocellulose complexes are formed, and each is a potential source of formaldehyde release to the atmosphere.

There are two basic sources of formaldehyde that can be released from pressed-wood products:

(1) Free (unreacted) formaldehyde present as a result of incomplete crosslinking during resin cure.

(2) Decomposition of unstable UF resin or resin-wood chemical species as a result of their intrinsic instability and/or due to hydrolysis.

Free formaldehyde, which is present in cured resin at low levels (<1 percent) is the most significant source of formaldehyde release from pressed-wood products in the initial period after they are manufactured. The specific time period in which free formaldehyde dominates releases is not known.

The second source, decomposition and hydrolysis, pertains to the large proportion of formaldehyde-bearing species like methylene ureas, urea methylene ethers, and cellulose-crosslinked species that may release formaldehyde for a much longer period of time. These species differ in their susceptibility to hydrolytic attack and decomposition, and their relative rates and durations of release can only be hypothesized at this time.

Factors Affecting Formaldehyde Release from Pressed Wood Products

A variety of factors affect the amount of each formaldehyde-releasing species present in the finished product. The resin formulation has a direct effect on release; resins with a low formaldehyde:urea ratio have, when cured, a lower level of free formaldehyde but may be less stable and more susceptible to hydrolysis. Other additives to the resin, such as

acid catalysts, change the resin chemistry and influence the release profile. The conditions under which the resin is cured affect bond strength, determining to some extent the stability of the resin components. The character of the wood itself also affects formaldehyde release; the more acidic the wood, the greater the tendency for acid hydrolysis and formaldehyde release.

Many other product-specific factors influence release. The more porous composition boards (particleboard and medium-density fiberboard) generally release more formaldehyde than laminated plywood. Emissions are a function of the surface roughness of the product as well, and a diffusion-theory approach that links boundary layer thickness and surface velocity has been experimentally validated. Formaldehyde emission rates are controlled by an equilibrium process that lowers the emission rate as the formaldehyde level in the air rises; that effect is more pronounced in smooth-surfaced products like fiberboard and plain plywood.

Environmental and architectural conditions also affect releases. Numerous investigators have evaluated the effect of temperature and, to a lesser extent, humidity on formaldehyde emission from pressed-wood products. These studies indicate that formaldehyde emission depends strongly on temperature and moderately on humidity. Experimentally-determined correction factors are generally used to correct monitoring data to a standard temperature and relative humidity. The temperature effect is exponential and is better understood than the humidity factor. Pressed-wood products respond to humidity changes by taking up some of the atmospheric moisture. Depending on the chemical moieties present in the resin and their susceptibility to hydrolysis, varying levels of formaldehyde may then be released.

As mentioned earlier, formaldehyde emission is an inverse function of the background concentration of formaldehyde in the air surrounding the board. Unlike other chemicals that are removed with ventilation air in a home, the formaldehyde concentration is not a direct function of ventilation or air exchange rate. Though an increase in ventilation does reduce levels by dilution of formaldehyde with clean air, any concentration reduction is followed by an increase in the emission rate. Doubling the ventilation rate may achieve only a one-third reduction or less in atmospheric formaldehyde levels. If the outside air has elevated levels, air exchange can become a source in homes.

Measures to Control Formaldehyde Emissions from Pressed Wood Products

Each control option under consideration by EPA is based on controlling one or more of the above-mentioned factors affecting emissions. As there are essentially two types of emissions -- long-term hydrolysis and decomposition and short-term release of free formaldehyde -- a control may reduce one type while either not affecting or, in some cases, actually increasing the other.

Reduction in the formaldehyde-urea ratio is a control already practiced by much of industry. In recent years, particleboard manufacturers have been using resins with a ratio of 1.2 or 1.3 parts formaldehyde per part urea, down from the resins with ratios of 1.6 or higher used prior to 1982. Resins with ratios of less than 1.2 have been developed and are being evaluated further. The use of lower mole ratio resins has been attributed with the demonstrated decline in emissions over recent years. All testing to date has, however, focused on the short-term emissions of free formaldehyde that are measured by the commonly-used emissions test methods (described in this report). It has been shown that the free formaldehyde emissions are lowered by the switch to low mole ratio resins in a proportion approximately equal to the

degree of ratio reduction. As measured by the two-hour desiccator test, emissions from particleboard with a ratio of <1.1 are in the range of 0.4 to 0.8 ug/ml, while conventional resins with ratios of ~1.3 have emissions of 1.2 to 2.0 ug/ml. Reducing the ratio in resins made for medium-density fiberboard is more difficult because a loss in essential properties is highly possible, but a reduction from 1.6 (current) to 1.2 can reduce emissions from 3.8 ug/ml to 0.6 to 1.4 ug/ml. It has been postulated, however, that lower mole ratio resins are less stable and more likely to hydrolyze; only repeated emissions testing, designed to detect hydrolyzing moieties, can resolve this question.

Formulation of scavengers into UF resin/wood systems also provides short-term reductions in emissions by adding reactive chemicals that bind the free formaldehyde in the resin, forming more stable complexes. Reactive scavengers added to the resin/wood system are designed to control formaldehyde that is unreacted or is released during the curing process; if scavengers are present in excess, they may affect emissions of decomposing or hydrolyzing products as well. The additives are generally ammonium compounds, urea, or sulfites. The long-term stability of these complexes has not been demonstrated; it is unlikely that they would be totally inert over years of product use in various environmental conditions. Short-term measurements indicate that various scavengers can reduce emissions by 50 to 75 percent.

Post-cure treatments with formaldehyde scavengers may be accomplished by placing finished products in the presence of a reactive gas (ammonia) or painting or spraying the board surface with the scavenger. The treatments control free formaldehyde in the short term, and can control long term releases if an excess of the scavenging agent is maintained in place.

There are a number of ammonia treatment processes that have been patented for formaldehyde control -- the Verkor FD-EX, the RYAB, the Swedspan, the BASF, and the Weyerhauser processes. The process variations that make these different are largely the actual method of ammonia application and whether the application takes place under pressure. In a test of the effectiveness of the Verkor method, performed three months after treatment, the process had reduced a particleboard's emission rate from 174 mg/100g wood to 5.5 mg/100g (perforator method). It is similarly effective on plywood paneling. The RYAB and Swedspan methods have been shown to be slightly less effective. Some of these treatment methods, including Verkor, involve removing excess ammonia as a final step; only the presence of excess, unreacted ammonia would ensure the long-term ability of these processes to reduce formaldehyde emissions. Another ammonia treatment method is an in-home fumigation, which could be used to reduce formaldehyde emissions. The long-term effectiveness of that method has not been well documented.

Coatings that contain reactive scavengers are also considered viable control options. All tests to date have demonstrated short-term effectiveness but the option would appear to have longer term possibilities because the coating would inhibit diffusion of water vapor and formaldehyde across the wood-air boundary layer. Tests on particleboard coatings include vinyl-toluene, which resulted in a 1.5 to 3-fold reduction in room air levels; a melamine coating that was 90 to 98 percent effective; and Falima-F, the active ingredient of which is unknown, which reduced emissions to <0.1 ug/m^2/hr. A urea-containing coating, Valspar, was applied to plywood and found to be 90 percent effective in short-term dynamic chamber tests, reducing chamber levels from 3 to 0.3 ppm.

Non-scavenger emission barriers perform two functions: they inhibit the ability of a pressed-wood product to absorb water vapor from the air, which speeds hydrolytic formaldehyde production and release; and they present a barrier to the formaldehyde diffusing out of the product. Paints, coatings, vinyl veneers, and other decorative overlays inhibit formaldehyde release; the effectiveness of the barrier is a function of the degree to which it inhibits permeability and porosity. Effectiveness ranges from over 30 percent for wallpapers on plywood paneling to 98 percent for particleboard coated on all its edges with nitrocellulose-based paint. Nonscavenger emission barriers would be expected to be less efficient than scavenger coatings because of the lack of reactive chemicals to actually bind formaldehyde to prevent its release.

Resin substitution, involving use of either isocyanate binders or phenol-formaldehyde resins in place of urea-formaldehyde resin, would virtually eliminate release of formaldehyde from pressed-wood products. Isocyanate resin products contain no formaldehyde per se, though some incidential release as a result of decomposition of cellulose might occur. Phenol-formaldehyde (PF) resins do contain formaldehyde, but are so stable that they emit relatively low levels of formaldehyde. The disadvantages to these resins, besides increased costs, are that they cannot be universally substituted. PF can be used in particleboard and in hardwood plywoods except those with light-colored veneers (the resin is dark and can discolor light wood); its ability to be used in MDF is not known. Isocyanate may not be suitable for plywood but is currently used successfully in particleboard and MDF manufacture.

Substitute wood products are available for all pressed-wood products that currently contain UF resins. Formaldehyde release from products like hardboard and softwood plywood does occur, but at very low levels. Gypsum board is another possible substitute, and it contains no formaldehyde, so only incidential release would be expected.

Increased ventilation does lower formaldehyde levels, though not in direct proportion to the increase in air exchange rate. It is the only option listed here that is not an emission control; it is, rather, an exposure control measure. As mentioned earlier, a reduction is followed closely by an increase in emission rate from a pressed-wood product. Increasing ventilation is effective both in the short term and in the long term, and this control will be effective for other pollutants in the indoor air environment. Increased ventilation, unlike the other controls listed, is effective on all residential sources of formaldehyde, not only pressed-wood products.

Other Residential Sources of Formaldehyde

There are numerous other sources of formaldehyde in homes: construction products containing PF resins (e.g., fibrous glass ceiling tiles and softwood plywood); appliances that incompletely burn hydrocarbon fuels, releasing formaldehyde and other aldehydes; smoke from cigarettes and other tobacco products; upholstered furniture and draperies with UF resin permanent press finishes; urea-formaldehyde foam insulation; and outdoor air used in ventilation. The significance of these other sources relative to pressed-wood products with UF resins varies widely with the occurrence of the sources in homes.

Though no residential sources of formaldehyde have been as well-studied as urea-formaldehyde foam insulation (UFFI) and pressed-wood products made from UF resins, there are enough data on many of these other sources to enable estimates to be made of their probable impacts on residential air levels of formaldehyde.

Emission rate information for fibrous glass insulation and ceiling tile containing PF resins indicates that these products are not likely to cause increases in indoor formaldehyde levels greater than 0.02 ppm even when subjected to elevated temperatures and relative humidities.

Similarly, emission rate testing of pressed wood products manufactured with PF resins indicates that these products will contribute less than 0.1 ppm to indoor air even when used at high loadings; monitoring conducted in three new mobile homes constructed with only PF resin wood products showed formaldehyde levels ranging from 0.02 to 0.07 ppm.

The data on combustion appliances show that formaldehyde release is a function of whether the appliance is tuned and functioning properly. Gas ovens and ranges may emit less than 2 to nearly 30 mg formaldehyde per hour of use; gas space heaters can emit less than 5 to over 60 mg/hour, depending on the efficiency of burning; and new kerosene space heaters emit up to 6 mg/hr of formaldehyde.

The emissions data on sidestream cigarette smoke range from 20 ug per cigarette to nearly 1.5 mg/cigarette. Several studies, however, concur on an emission rate of 1.0 to 1.2 mg/cigarette. The importance of this source is obviously related to use patterns. Studies where numerous persons chain-smoked in a poorly ventilated room did indeed show that formaldehyde levels were elevated after a short period of time, but other studies in the homes of smokers indicated that, at a smoking rate of 10 cigarettes per day, formaldehyde levels were not elevated over controls with similar loading rates of other sources.

Available data on drapery and upholstery fabrics indicate that, with emission rates only as high as 15 ug/m^2/hr, these could cause indoor air levels to increase by greater than 0.01 ppm only under very high loading situations. Although emission rates for new unwashed apparel have been reported a high as 31 ug/m^2/hr, the impact of apparel on indoor air levels is expected to be negligible because laundering will significantly decrease the emission rate. A modeling exercise discussed in Section 3.6 of this report was intentionally designed to estimate the relative importance of numerous sources of residential formaldehyde in a

model mobile home and model conventional home. Reasonable estimates of the loadings of the sources were made and it was conservatively assumed that the emission rate from each source was independent of emissions from other sources. A simplified ranking follows; setting the most important sources as 1 and scaling the other appropriately:

Model Mobile Home		Model Conventional Home	
Hardwood paneling	= 1.0	Industrial particleboard	= 1.0
Particleboard flooring	= 0.55	Gas space heater	= 0.84
Industrial particleboard	= 0.22	MDF	= 0.76
Gas space heater	= 0.19	Particleboard flooring	= 0.73
MDF	= 0.17	Hardwood paneling	= 0.56
Other combustion sources	= 0.12	Other combustion sources	= 0.56
All other sources	=<0.10	All other sources	=<0.40

The "all others" category includes textiles, carpeting, fiberglass insulation and ceiling tiles with PF resins, and other sources.

Current Levels of Exposure

Because of the changing nature of pressed-wood products with UF resins and the constant evolution and improvement in monitoring techniques, the universe of residential monitoring data is not the most appropriate data base for describing formaldehyde exposure in homes. Many data sets are based on investigation of homes from which complaints of formaldehyde symptoms have been filed; these data sets may not be representative of average exposure because of bias toward high concentrations. Homes studied before 1980 were built with products made of high F:U ratio resins that are no longer on the market; they cannot be considered as baseline exposures for that reason. The most appropriate data for describing current exposures in mobile and conventional homes are therefore those generated by random sampling of noncomplaint homes after 1980, preferably after 1982 (when manufacturers began using resins with mole ratios of 1.5 or less). These restrictions on the "appropriate" data base still leaves a considerable volume of monitoring data on levels in homes.

Numerous studies on conventional homes are discussed in this report. Studies performed by the Lawrence Berkeley Laboratories, the Consumer Product Safety Commission, the government of Canada, and state and academic officials in Washington, Iowa, and Indiana are the most recent and representative of average exposure in conventional homes, per the criteria discussed above. These studies indicate that the average level of formaldehyde in conventional homes is approximately 0.05 ppm, and that the age and construction of the home are the major determinants of the concentration. Newer homes and energy-efficient homes with low air exchange rates tend to have higher formaldehyde levels (around 0.1 to 0.2 ppm) than older (over five year old) homes, with average levels of 0.005 to 0.08 ppm. Comparison of these data with data collected prior to 1980 indicates that there has been little change in conventional home levels since 1978, the data of the earliest comprehensive survey of home levels.

The average level in mobile homes appears, however, to have declined in recent years. Average levels in the existing stock of mobile homes are now around 0.2 to 0.5 ppm, with mean levels in individual homes (including complaint homes) ranging from <0.1 to over 1.0 ppm. An aggregated data set of two well-conducted studies (the 1980-1982 Wisconsin study and the 1980-1981 Clayton study described in this report) has nearly 1,200 data points. The mean of that data set is 0.43 ppm, with a median of 0.31. This aggregated data set also contain home age values for every mobile home sampled. The correlation coefficient of the log-transformation exponential function describing the data is 0.4, indicating that 40 percent of the variability in home levels is attributable to home age, while other factors control 60 percent of the variability. The 0.43 ppm mean for the data set corresponds to a home age of 246 days. The predicted average concentration of formaldehyde in a mobile home over the first ten years of its use is 0.19 ppm. A more recent study, University of Texas (1982-1983) showed average levels of

about 0.2 ppm in mobile homes built primarily during the 1980s but showed little significant variation by home age. The data indicate that recently-built mobile homes may have lower initial formaldehyde levels than homes built prior to 1980, but that the concentrations may not significantly decline over time.

Potential Reductions in Exposure Levels

The exposure reduction that could result from the implementation of the control measures described in this report is difficult to determine. The factors controlling formaldehyde concentrations in mobile and conventional homes are complicated, interdependant, and not well understood. The available data on emissions reductions that can be accomplished by various controls are sparse; the relatively few data are often not comparable because of differences in measurement techniques. Finally, the long-term effectiveness of the control options is not known, and can only be speculated on.

There are, however, some simple tools that may indicate the exposure that may result from control of pressed-wood products containing UF resins. Review of monitoring data for older homes in which formaldehyde emission from pressed-wood products is probably limited to low levels of hydrolysis products may be representative of situations in which other sources predominate. Measured levels in conventional homes greater than 15 years old averaged 0.03 ppm in one study (Hawthorne et al. 1984). In a study of various formaldehyde sources in homes (Traynor and Nitschke 1984), the control homes that had no identified sources of formaldehyde and the homes without pressed-wood products but with combustion sources had formaldehyde levels ranging from 0.007 to 0.077 ppm. These are in the reported range of levels in homes with pressed-wood products and are not different from the average reported levels in conventional homes (~0.05 ppm).

and the homes without pressed-wood products but with combustion sources had formaldehyde levels ranging from 0.007 to 0.077 ppm. These are in the reported range of levels in homes with pressed-wood products and are not different from the average reported levels in conventional homes (~0.05 ppm).

The levels in mobile homes without pressed-wood products are more difficult to determine, since virtually all mobile homes currently constructed contain particleboard, MDF, and/or paneling as major structural components. The decay function for the Clayton and Wisconsin data, previously described, can be used to project levels into the future, when emissions from pressed-wood products may be relatively low. This highly speculative approach to predicting exposure reduction is not specific to any particular control. The decay function predicts that a concentration of 0.047 ppm would be present in a mobile home ten years after construction assuming the initial concentration in the new home was 0.50 ppm. This level might correspond to the levels that would be reached by controlling pressed-wood product emissions in some fairly effective manner. An error inherent in using the decay function as described is that sources that would not release less over time (e.g., gas appliances and cigarettes) are decayed in the same manner as pressed-wood products. This error can be corrected by adding a constant to the decay function representing a background level attributable to outdoor air, combustion products, etc.

Another simple approach to predicting exposure reduction is to perform simple modeling calculations of indoor air levels in homes, factoring in emissions from all sources except pressed-wood products with UF resins. A simple steady-state model developed at Oak Ridge National Laboratory is described in this report. Using that model, and emission factors for new residential sources except UF resin bonded pressed-wood products, yields an estimated steady-state concentration of approximately 0.07 ppm (at a typical mobile home air exchange rate of 0.35 ACH in a volume of 175 m^3).

If controls on pressed wood products containing UF resins were 100 percent effective, these simple assumptions indicate that levels in mobile homes would drop from a current average of 0.2 to 0.5 ppm to less than 0.1 ppm. Implementation of one or more controls with an effectiveness of less than 100 percent would result in incremental improvement between the current average and the projected levels.

There is a high degree of uncertainty surrounding this prediction. Current tools and data do not allow refinement at this stage. Ongoing work on a more sophisticated model sponsored by CPSC and EPA should produce a more reasonable approach to exposure prediction in the fall of 1985. This work includes compilation of emissions data, study of the factors affecting formaldehyde concentrations, and preparation and validation of an indoor air model applicable to this situation. The model is described in this report, and will provide many of the answers to questions that are now addressed by educated speculation. The current work is, however, limited; still lacking are emissions characterization for pressed-wood products that have been treated with specific control options. There are no data that can be utilized in that model on emissions from boards with known, low molar ratio resins; no data on boards with specific scavengers; there are only emissions data on boards characterized by industry as either typical or low-emitting. These data do include emissions from PF resin wood products, so that analysis of exposure reduction resulting from product substitutions will be possible. Further data development will, however, be required to demonstrate exposure reduction from other control options scenarios.

1. INTRODUCTION

1.1 Background

On May 23, 1984, the U.S. Environmental Protection Agency (EPA) decided that residents of manufactured and conventional housing could be subject to a significant risk of cancer from exposure to formaldehyde. A major source of formaldehyde in these homes is construction material in which urea-formaldehyde resins are used. EPA at that time issued an Advanced Notice of Proposed Rulemaking (ANPR) in which initiation of a full regulatory investigation was announced; the purpose of that investigation is to determine whether reasonable control options exist for reducing formaldehyde exposure to this population.

As part of this regulatory investigation, EPA is reexamining existing information and is gathering and reviewing additional information on two general topic areas: (1) current levels of exposure to formaldehyde in housing and the sources and factors that affect these levels, and (2) reduction of exposure levels that could result if control measures are implemented. The purpose of this report is to summarize current knowledge regarding these topic areas.

1.2 Report Organization

There are seven major sections to this report, which are summarized below:

Section 2 provides a background discussion on formaldehyde emissions from pressed-wood products containing urea-formaldehyde (UF) resins. Included in this discussion are descriptions of the major pressed-wood products and their uses in residential settings, the mechanisms of formaldehyde release, and the factors that affect the rate of release. Section 2 also presents a background discussion on formaldehyde emission rate testing methods. All methods used by researchers to generate data discussed within this report are described. Correlations between results generated by different methods are, where applicable, discussed.

Section 3 discusses the residential sources of formaldehyde (other than pressed-wood products formulated with UF resins). Described are urea-formaldehyde foam insulation (UFFI), products made with phenol-formaldehyde (PF) resins, consumer products made with UF resins (textiles), indoor combustion, and infiltration

of outdoor air. This section is concluded with a presentation of data on the comparable strengths of various formaldehyde sources in residential settings.

Section 4 presents a summary of monitoring data for formaldehyde in homes. The results of a number of large-scale studies are tabulated, and ongoing research projects are discussed. Also included is a brief discussion of monitoring studies that were designed to examine the factors that affect indoor air levels of formaldehyde in homes.

Section 5 describes the control options currently under consideration by EPA for reduction of exposure to formaldehyde emitted from pressed-wood products. Four types of controls are described in terms of their projected short- and long-term effectiveness: changes in UF resin formulation, post-cure board treatments, use of substitute resins, and use of substitute products. Other potential controls, such as increased room ventilation, are discussed briefly. Section 5 also presents a summary of quantitative data on formaldehyde emissions and exposure levels resulting from the application of these control options.

Section 6 presents a summary of existing formaldehyde emissions and exposure standards in the U.S. and in other countries.

Section 7 describes efforts to predict residential levels through modeling. The Consumer Product Safety Commission (CPSC) and EPA are supporting the development of a sophisticated formaldehyde model; the status of model development and validation is a subsection in Section 7. Simplified models or algorithms are also briefly discussed.

2. PRESSED-WOOD PRODUCTS CONTAINING UF RESINS

2.1 Product Descriptions

Pressed-wood products that utilize urea-formaldehyde resin as a thermosetting binder are used in flooring, interior walls and doors, cabinetry, and furniture construction; these relatively inexpensive pressed-wood products are a growing market share of the construction products industry (Meyer and Hermanns 1984a). The three major classes of pressed-wood products containing UF resins are particleboard, medium-density fiberboard (MDF), and hardwood plywood.

2.1.1. Particleboard

The National Particleboard Association (NPA 1984) states that 28 firms, operating 45 plants, manufactured over 3 billion square feet of particleboard in 1983 by the platen-press, or mat-forming, process. The NPA (1984) defines the mat-forming process as one where resin-coated wood particles are formed into mats, which are pressed in a heated press (platen) at elevated temperatures. These 45 plants account for an estimated 96 percent of U.S. particleboard production capacity (NPA 1984). An estimated 10 plants with a total annual capacity of approximately 50 million square feet manufacture particleboard by extrusion of resin and wood into mounted platens that serve as a die (NPA 1984). An additional 75 million square feet may have been produced by the Mende Process (NPA 1984), which forms thinner particleboard by pressing a ribbon of resin-coated wood particles. The average capacity of each of the 45 plants is 81 million square feet (NPA 1984).

Though three types of resin (UF, PF, and isocyanate resins) are suitable for use in particleboard, UF resin is the primary adhesive used in 41 of the 45 mat-form process plants in the U.S. (NPA 1984). The four remaining plants, three of which use phenol-formaldehyde (PF) resin and one of which uses isocyanate resin, constitute less than 10 percent of total U.S. particleboard production capacity (NPA 1984, ICF 1984). Most current F:U mole ratios for UF resins used in particleboard are claimed

to be in the range of 1.15 to 1.3 (i.e., 1.15 to 1.3 moles of formaldehyde per mole of urea), down from about 1.6 in the late 1970s (Podall 1984).

Particleboard produced with UF resin uses about 5 to 10 percent resin by weight (Podall 1984). George (1977) describes the variations in UF production techniques for resins with different end uses. The UF resin he describes is a standard UF resin that is closely controlled for viscosity; UF resin for particleboard is essentially monomeric, for enhanced solubility and ease of application (George 1977). Approximately 5 percent urea (by weight) is added to the resin mixture to control polymerization; the final resin, in liquid form, is 59 to 65 percent solids (George 1977, Podall 1984).

NPA (1984) describes three major types of particleboard: underlayment, mobile home decking, and industrial board. Underlayment is the least expensive type of particleboard and is typically used in floor systems and for general applications. Mobile home decking, whose name implies its use, is more expensive because it must be manufactured to higher specifications of strength and stability. Industrial particleboard, the most expensive, is often used as the base material in cabinets and furniture and is the highest grade manufactured. The NPA estimates that, in 1983, approximately 70 percent of particleboard was used in furniture, fixtures, cabinets, etc., and that the remaining 30 percent was used for construction purposes (NPA 1984). Table 1 summarizes available information on loading rates of pressed wood products, including particleboard, in residences. The average loading rate of particleboard (underlayment, kitchen cabinet, and shelving) in new conventional U.S. homes containing particleboard is reported to range from 0.112 to 0.167 m^2/m^3 (m^2 of product surface area/m^3 of indoor air volume) (NPA 1984), while the average reported loading rate in mobile homes is 0.5 m^2/m^3.

Table 1. Use of Pressed-Wood Products in Home Construction

Category	Type of home[f]			
	SFD	TH	MF	MH
New Homes (U.S.)[a,b]				
Percent units containing				
Hardwood plywood paneling	7.6	9.3	8.5	most
Particleboard underlayment	30.5	9.2	1.7	most
Average loading rates,[c] (m^2/m^3)				
Hardwood plywood paneling	0.066	0.059	0.049	1.0
Particleboard underlayment	0.118	0.092	0.033	-
Particleboard shelving	0.010	0.016	0.020	-
Particleboard kitchen cabinets	0.039	0.052	0.059	-
Total particleboard	0.167	0.160	0.112	0.5
New Homes (Canada)[d]				
Percent units containing				
Particleboard	100	100	100	100
Average loading rates (m^2/m^3)				
Total particleboard	0.145	0.100	0.079	0.479
Existing Homes (U.S.)[e]				
Percent units containing				
Hardwood plywood paneling	35.5	—	—	most
Particleboard	90.3	—	—	most
Average loading rate (m^2/m^3)				
Hardwood plywood paneling	0.098	—	—	1.0
Particleboard	0.058	—	—	0.5

Note

Data reflect only interior uses of UF pressed wood products. Loading rates are for those homes containing these products.

[a]Source: NPA (1984) and HPMA (1984) for conventional homes - Based on interpretation of the results of a survey of 900 home builders (103 responses) regarding the extent of use of particleboard and hardwood plywood paneling in new homes containing these products (NAHB 1984).

[b]Source: Meyer and Hermanns (1984a), NAHB (1984), MHI (1984) for mobile homes.

(Footnotes continued on next page)

Table 1. Footnotes (continued)

[c] m^2 of produce surface area/m^3 of indoor air volume.

[d] Source: InterArt (1983) - based on in-home surveys at 9 SFD, 1 TH, 1 MF and 1 MH. Total loading includes underlayment, shelving and cabinets. SFD loadings ranged from 0.028 to 0.491 m^2/m^3.

[e] Source: Schutte (1981) - Based on in-home surveys at 31 SFD. Average loadings based on homes containing these products.

[f] SFD = Single family dwelling
TH = Townhouse
MF = Multifamily dwelling
MH = Mobile home

2.1.2 Medium-Density Fiberboard (MDF)

The NPA also represents manufacturers of medium-density fiberboard, which is composed of resin and sawmill residue fibers. Resin is added to comprise approximately 7 to 9 percent by weight of the mixture; the wood is in the form of 0.2 to 0.8 mm fibers that are created by cooking or shredding the raw wood material (Podall 1984, NPA 1984, Meyer 1979). Essentially all medium-density fiberboard is manufactured by platen-pressing (mat-forming) (NPA 1984).

Ten companies with eleven plants currently produce MDF; the total 1983 production was 604 million square feet, with a total capacity of 760 million square feet (an average capacity per plant of 69 million square feet) (NPA 1984).

Urea formaldehyde is the only resin currently used in MDF production, according to NPA (1984), with a typical mole ratio of F:U of 1.65 (Podall 1984). MDF requires a higher mole ratio resin because strong adhesion is more difficult to obtain than in particleboard manufacture (Podall 1984). Two factors contribute to this: (1) the lower moisture content of the wood fibers (4 percent in MDF as opposed to 7 to 11 percent in particleboard), and (2) there is less lignin and hemicellulose in the wood fibers, which normally aid the bonding process (Podall 1984). NPA mentions experiments with PF and isocyanate resins (NPA 1984), and Forss and Fuhrmann (1980) discuss the use of lignin as a fiberboard adhesive (and compare the performance of lignin-based boards to those manufactured with PF). It appears that the exclusive use of UF in medium density fiberboard is not a result of technical necessity of UF, but is probably because of cost considerations. It is widely recognized that UF is the least expensive, most readily available resin for most pressed-wood product applications.

Medium-density fiberboard panels are homogeneous in texture and color, and appear more like lumber when finished than other pressed-wood products (NPA 1984). About 95 percent of production is directed to

furniture, fixtures, doors, and cabinets, while 5 percent is formulated into miscellaneous wood products (NPA 1984). In mobile homes, MDF is also used as a decorative molding around acoustical ceilings (Meyer 1979). The differences in use patterns between MDF and particleboard are largely attributable to the ability of MDF to accept machining of its edges, allowing it to be used directly as a finished product.

No data are available on the precise extent of MDF's use in either mobile or conventional homes. The National Association of Home Builders (NAHB 1984) survey of new conventional homes revealed that 9.5 percent of the components of kitchen cabinets are composed of MDF and that MDF accounts for 0.7 percent of shelving in new homes. The use of MDF in home construction is probably highly variable, and it is likely used to a lesser extent than is particleboard.

2.1.3. Hardwood Plywood

Hardwood plywood is a laminated product, unlike particleboard and fiberboard, and contains only 2.5 percent UF resin by weight (Meyer and Hermanns 1984a). It is manufactured by cross-stacking three to five layers of veneers, with UF resin and fillers between the layers (Meyer 1979). In some boards, veneers are applied to a core substrate of particleboard or MDF (Smith 1982, 1983). The stack is then pressed at temperatures up to 100°C and pressures up to 300 psi (Meyer 1979). Nearly 2 billion square feet were manufactured in 1983 (HPMA 1984). It is used for interior wall paneling (55 percent of production), furniture and cabinets (30 percent of production), and door skins and laminated flooring (15 percent of production) (HPMA 1984).

George (1977) describes the manufacture of UF resin for use in plywoods. The major difference between UF resin designed for plywood adhesion and resin for use in fiberboard or particleboard is that the plywood resin is of higher average molecular weight (more completely polymerized). The final resin can be a spray-dried powder or a syrup of about 66 percent solid content (George 1977). Extenders (starch or protein) may comprise up to 25 percent of the resin (George 1977).

A substantial quantity of the hardwood plywood consumed in the U.S. (about 75 percent of total consumption) is imported Asian "lauan" plywood that is prefinished by U.S. firms (Smith 1982). The faces of imported plywoods are either printed, stained, papered, or covered with a domestic hardwood veneer to produce the finished product. Several varieties of plywood are commercially available; all can be used in mobile or conventional home interiors. Printed paneling is inked in a decorative pattern and accounts for 35 percent of plywood use; papered paneling is covered by wallpaper (40 percent of plywood use) or vinyl (7 percent of plywood use); natural hardwood or domestic paneling is plywood covered with a hardwood veneer, and accounts for 18 percent of all panels (HPMA 1984). The NAHB survey indicates that plywood is used for 15.6 percent of kitchen cabinetry and 7.2 percent of shelving in conventional homes.

Table 1 provides additional statistics on use of plywood paneling in new home construction. Meyer and Hermanns (1984a) state that the average loading rate in mobile homes is 1.0 m^2/m^3, which is appreciably higher than that found in the NAHB survey of new conventional homes. However, it should be realized that because sales of paneling for remodeling and repair applications generally account for more than 20 times the sales of paneling for new home construction, the actual loading rates of paneling in conventional homes may be higher than those listed in Table 1. Unfortunately, the average paneling use for remodeling or repairs in homes is not available (Matthews et al. 1983b).

UF resin is the overwhelming choice of plywood manufacturers with current formaldehyde:urea mole ratios reported to range from 1.2 to 1.5 or higher (Podall 1984, HPMA 1984). The Hardwood Plywood Manufacturers Association (HPMA 1984) says that over 90 percent of hardwood plywood produced uses UF resin and that phenol-formaldehyde resin is used to manufacture the balance (ICF 1984). Forss and Fuhrmann (1980) describe the use of lignin in Finnish production plants, but it seems unlikely that it is currently used in U.S. plywood production.

2.2 Sources and Mechanisms of Formaldehyde Formation and Release

This section is excerpted from a recent draft report (Podall 1984) by H. Podall of EPA's Office of Toxic Substances entitled "A Review of the State-of-the-Art on Urea Formaldehyde Resins for Wood and Causes of Formaldehyde Release." In addition to discussing sources and release mechanisms, this report examines in depth: resin production; resin composition and chemistry; resin use by board type and wood species; and recent advances in UF resins.

The formation and subsequent release of formaldehyde from a UF-bonded pressed-wood product is due to two basic sources of latent formaldehyde:

1. "Free" formaldehyde arising from the UF resin prior to or during the curing of the resin in the board.

2. Chemical species containing bound formaldehyde which liberate formaldehyde as a result of their intrinsic instability (and do not require, stoichiometrically, water, for the formation of formaldehyde) and/or due to hydrolysis.

In order to understand the short- and long-term significance of these formaldehyde emission sources, as well as to understand the effects various control measures may have on reducing or eliminating these sources, it is important to:

- Define the actual species capable of producing formaldehyde.

- Assess their relative importance as sources of formaldehyde by identifying the key reactions involved in the formation of formaldehyde.

2.2.1 Chemical Species Capable of Producing Formaldehyde

The "free" formaldehyde in a pressed-wood product presumably exists as methylene glycol, low molecular weight formaldehyde oligomers (e.g., $HO-(CH_2-O)_n-H$, where $n = 2$ to 4), and possibly some paraformaldehyde. These chemical forms of formaldehyde may be extensively hydrogen-bonded to the cellulose, hemicelluloses, lignin, and

to the UF resin. In addition, they may be dissolved in the water contained in the pores of the wood. The following components of the cured UF resin can undergo hydrolytic degradation to form formaldehyde: N-methylol urea, methylene ether urea, substituted urea, and methylene urea moieties. Also, reaction of N-methyol urea moieties with the wood cellulose and with formaldehyde (formed during cure) may produce latent formaldehyde moieties.

A list of the principal potential formaldehyde releasing moieties believed to be present in a board bonded with a commercial 1.3:1 F/U mole ratio resin and an estimate of the relative amounts of formaldehyde present are given in Table 2. The reactions and assumed distribution of products are given in the footnotes to Table 2. Although the assumptions made are believed to be reasonable, it is important to recognize that they represent major extrapolations of a given set of results for a particular cured resin in the absence of wood.

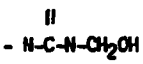
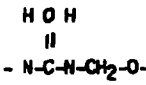
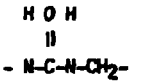
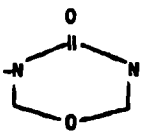
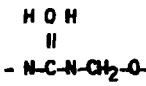
2.2.2 Relative Importance of Chemical Species as Sources of Formaldehyde

There appears to be available in the literature two sets of kinetic data pertaining to the hydrolysis of structural moieties or components present in UF resins. They are (1) dilute solution kinetics of relatively simple model compounds, such as N,N'-dimethylol urea, and (2) more limited data on the hydrolysis of UF-crosslinked cellulose and of crosslinked UF resins, generally at very low pH and high temperatures, and in certain cases employing a questionable analytical method for formaldehyde.

Based on the available kinetic data and considerations of structure reactivity, estimates were derived of the relative reactivities for the structural species given in Table 2. These are given in Table 3, together with estimated relative durations of formaldehyde releases.

A picture that emerges from the values estimated in Table 3 is that (1) next to the "free" formaldehyde, the formaldehyde bound to the wood

Table 2. A Rough Estimate of the Relative Amounts of the Various Formaldehyde Derived Special in a Cured Board with 1.3:1 F/U Mole Ratio Resin

Structural species	Percent formaldehyde		
	Prior to cure[a]	Cured (neat resin)	Cured[b,e] (in board)
CH_2O dissolved in pores	0	0	4.4(e2)→2.2(g) → 0.5(h)
CH_2 - hydrogen bonded to wood cellulose, etc.	0	0	4.4(e2)→2.2(g) → 1.7(h)
CH_2O in resin	0.5(c)	0	—
cell-O-CH_2OH	—	—	2.2(g) → 1.8(i)
cell-O-CH_2-O-cell	—	—	2.2(g) → 0.4(i)
	43.8(c)	2.8	1.1(e3)
	29.0	44.3(d)	42(f1)
	26.6	52.8(d)	48(f2)
	—	—	4.4(e1)
	—	—	
	—	—	

Source: Podall (1984).

[a]Borden data (Williams 1984).

[b]Should vary with board type (hardwood plywood, medium density fiberboard, or particleboard) and with furnish in particleboard.

[c]Percent available N-methylolurea (NMU) for reaction during cure = 0.5 + 43.8 = 44.3%.

Table 2. Footnotes (continued)

[d]Percent conversion available NMU to methylene ether urea = (44.3 - 29.0)x100/(44.3 - 2.8) = 15.3 x 100/41 5 = 37 percent. Percent conversion of available NMU to methylene urea = (52.8 - 26.6) x 100/41.5 = 26.2 x 100/41.5 = 63 percent.

[e]Assumptions regarding converstion of NMU:
e1 = 10 percent of the available NMU reacts with wood = 4.4 percent.
e2 = 10 percent of the available NMU hydrolyzes to CH_2O = 4.4 percent.
e3 = 2.5 percent of the available NMU does not react = 1.1 percent.
e4 = 77.5 percent x 0.443 of NMU converts to 34.3 percent methylene ether ureas + methylene ureas

[f] f1 = Net increase in methylene ether ureas = 34.3 x .37 = 12.7 percent.
f2 = Net increase in methylene ureas = 34.3 x .63 = 21.6 percent.

[g]Assume 50 percent of CH_2O from NMU reacts with cellulose = 0.5 x 4.4 = 2.2 percent and 50 percent remains as CH_2O.

[h]Assume 75 percent of residual CH_2O (1.7%) becomes H - bonded to wood and 25 percent is in pores (0.5 percent).

[i]Assumes 80 percent of CH_2O which reacts with wood (1.8%) is converted to cellulose hemiformals and 20 percent is converted to cellulose formals (0.4 percent).

Table 3. Relative Rates of Formation of Formaldehyde and Duration of Release from a Cured UF Board

Structural moiety	% as HCHO[a]	Relative reactivity[b]	Initial relative rate[c]	Estimated relative duration of release[d]
CH_2O dissolved in pores	0.5	instantaneous (4,000)	2,000	0.0004 (instantaneous)
CH_2O H-bonded to wood	1.7	very fast (400)	680	0.007 (instantaneous)
cell-O-CH_2-O-H	1.8	fast (20)	36	0.14 (short term)
	1.1	moderate (1.00)	1.1	2.4 (intermed. term)
cell-O-CH_2-O-cell	0.4	moderate-slow (0.5)	2	2.8 (intermed. term)
	42	slow (0.1)	4.2	60 (life of board)
	4.4	slower (0.03)	0.13	126 (life of board)
	48	very slow (.002)	0.10	3087 (life of board)

Source: Podall (1984).

[a]Based on estimates given in Table 2.

[b]Proportional to pseudo 1st order rate constants in sec^{-1}, pH 4 to 5, 25°C.

[c]Initial relative rate ~ % as HCHO x relative reactivity.

[d]Estimated duration = $t_{99.9\%} \sim \frac{2.303 \log C_i + 2.303}{k_{rel.}}$; C_i = % HCHO:

k_{rel} = relative reactivity (see Podall 1984 for derivation).

as hemiformals constitutes the most important source of formaldehyde from a rate standpoint, (2) over the long term, the formation of formaldehyde appears to be largely determined by the concentration of methylene ether urea functionalities, in spite of the greater reactivity expected for N-methylol urea functionalities, and (3) the primary form of the resin, viz., crosslinked methylene ureas, would appear to contribute very little to the release of formaldehyde, even at pHs (of 4 to 5) conducive to hydrolysis.

Thus, this picture appears consistent with a water transport mechanism as being rate-determining for the immediate through short term release, and with the hydrolysis mechanism as rate-determining for the intermediate to long term release. The board may thus be viewed as functioning as a tight reservoir for the formaldehyde formed from the hemiformals of cellulose and related species, such as the hemiformal of N-methylol ureas. Following the release of the "free" formaldehyde, initially present in the board after curing and from the facile hydrolysis of the hemiformals, the formation of formaldehyde from such sources as methylene ether ureas, N-methylol ureas, and the formals of cellulose, become rate determining for the release of formaldehyde.

2.3 Factors Affecting Formaldehyde Release from Pressed Wood Products

2.3.1 Product-Specific Factors

Many of the factors that dictate whether release occurs (and if so, at what rate) are functions of the wood or resin and the manufacturing processes used. Each factor discussed below is important under at least some circumstances; researchers have met with only limited success in defining the controlling factor under circumstances of use in mobile or conventional homes.

(1) Material Structure and Porosity. Christensen et al. (1981) state that board porosity is a "major controlling factor in formaldehyde emission" and that "the rate of formaldehyde release from particleboard is a diffusion controlled process." Meyer and Hermanns (1984a) agree

that the formaldehyde emission rate is strongly influenced by the structure and porosity of the pressed-wood product. This statement refers to diffusion of unreacted formaldehyde from the core of the product, though these parameters could conceivably affect resin hydrolysis as well. Structure and porosity are closely related parameters.

Structure refers to the inherent differences between a laminate, such as hardwood plywood, and the true composite woods (fiberboard and particleboard). Some researchers have found formaldehyde emissions from the laminated plywood to be lower than emissions from the same amount of particleboard or fiberboard. This is easily explained by the fact that the UF resin in plywood is segregated in the glue layer between intact wood sheets, with little if any direct contact with air. Particleboard and fiberboard are, however, mixtures of wood and resin throughout; though the residual formaldehyde is concentrated in the center by the manufacturing process, free formaldehyde levels at the surface of these pressed wood products can be half the elevated level in the center (Meyer and Hermanns 1984a). Thus, some free formaldehyde is available for release from fiberboard and particleboard immediately.

Plywood is also less porous than fiberboard or particleboard. The speed of formaldehyde diffusion through pressed wood products has been studied. Meyer and Hermanns (1984a) report rate constants (in meters per hour) for the three types of products:

- 0.4 ± 0.3 plywood
- 0.5 ± 0.2 medium density fiberboard
- 0.8 ± 0.2 particleboard

The tests designed and performed by Christensen et al. (1981) generated data that compare (1) formaldehyde emissions from particleboard surfaces and edges and (2) emissions from board surfaces of different structure. The test involved a chamber containing the board sample,

into which was introduced a calibrated flow of heated, water saturated air. After the air exited the chamber, it was analyzed via the chromotropic acid method. The purpose of creating the test was to determine the extent to which the increased edge area of board samples commonly tested (increased relative to surface area) dominate emissions test data. Results are reported in units of mg formaldehyde/ft^2 board. The test was run by sealing either the edges or the surface with a thick layer of epoxy resin.

Christensen et al. found that the more porous particleboard edges emitted formaldehyde at a rate of 2.1 times the surface emission rate. A three-layered hardwood plywood was also tested, and the average ratio of edge emissions to surface emissions was 4.9. These researchers also found a correlation between density and surface emissions, with the most dense (least porous) board composition emitting far less formaldehyde than the two less-dense boards tested. Actual porosity (as measured by resistance to air flow through the board under vacuum) also showed a strong positive correlation with emission of free formaldehyde.

Matthews et al., in their work for ORNL, developed a diffusion-theory approach that predicts formaldehyde emission rates as a function of ambient concentration and a product's surface structure (Matthews et al. 1982, Report IV). They found that the macro-level surface characteristics that affect boundary layer thickness and velocity of air across the face have a direct, predictable effect on emission rate. Products with relatively smooth surfaces produce, when tested, a linear plot of emission rate versus concentration with a high negative value for slope. Products with irregular surfaces generally show linear plots with low negative slope values.

The diffusion theory was borne out by tests and calculations, which confirmed that:

- Fiberboard and plain plywood, with smooth surfaces, will have lower emission rates as concentration increases.

- Paneling, with alternating smooth and rough areas, will have a range of emission rate/concentration relationships.
- The relationship between emission rate and concentration for particleboard, with its diverse mixture of surfaces, is not linear over all ranges of emission rate and concentration.

(2) Type and Quantity of Resin Used As discussed in Section 2.2, the F:U ratio in the resin strongly affects the presence and potential release of formaldehyde. Other formaldehyde resin formulations (phenol-, melamine-, and tannin-formaldehyde are good examples) generally emit less formaldehyde, either as unreacted residual or as a hydrolytic product. Figure 1 shows the low emission profile of PF particleboard relative to UF particleboard. Myers (1984a) presents a review of the literature on the effect of mole ratio on formaldehyde emission rate. It describes the relationships between F:U mole ratio, resin free formaldehyde (as percent), and formaldehyde emission rate. Data from several investigators, as presented by Myers (1984a), clearly show the positive relationships between mole ratio and free formaldehyde and emission rate. Myers states that the exact relationship is uncertain; other variables (board manufacturing and aging) can affect the slope of these lines. It is clear, however, that the type of UF resin (as defined by mole ratio) strongly affects emission rates of formaldehyde.

Some inferences can be drawn from existing data on the effect of resin quantity on formaldehyde emission rates. Researchers have shown (Matthews et al. at ORNL and others) that emission rates for UF pressed wood products can be generally described as:

MDF > particleboard >> plywood

Numerous differences between these product types affect emission rate; among those differences may be resin quantity. Recall from Section 2.1 that the weight percents of resin in the three product types are:

MDF - 7 to 9% w/w
Particleboard - 5 to 10% w/w
Plywood - 2.5% w/w

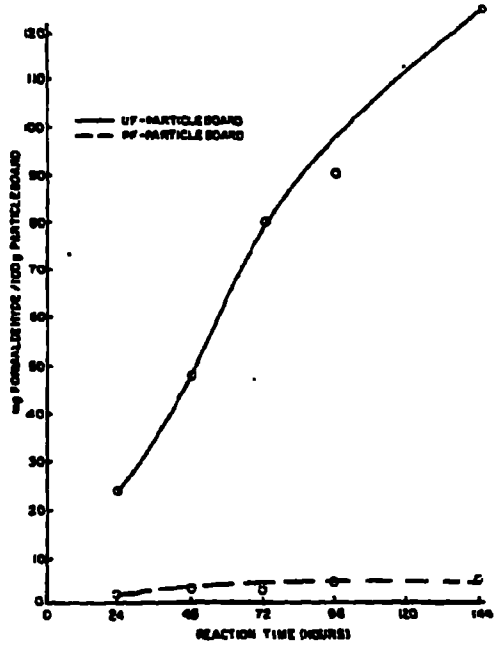


Figure 1. Formaldehyde Release from UF and PF Particleboards Measured by the WKI Method

Source: Roffael (1978).

Plywood, with the lowest resin quantity, also has generally the lowest emission rate; the distinction between particleboard and MDF is less clear.

(3) Manufacturing Conditions. A variety of manufacturing conditions can affect the degree and uniformity of resin polymerization. Incomplete polymerization leaves an excess of unreacted formaldehyde in the pressed wood product, as well as increased amounts of moieties that can readily hydrolyze to release formaldehyde.

Meyer and Hermanns (1984a) and Meyer (1979) present discussions of UF resin manufacture and formulation into pressed wood products. Most potential manufacturing variables that can affect formaldehyde release are related to the complex resin chemistry:

- Use of non-uniform wood particles can lead to areas of improper resin:wood ratios and incomplete cross-linking of the polymer (this refers largely to particleboard).
- Use of non-uniform wood particles, with spatially-varying water content and pH, can also cause incomplete polymerization in particleboard manufacture.
- Use of a UF resin with another chemical that may change the pH of the wood-adhesive mixture can prevent proper polymerization. Manufacturers are urged to test any change in catalyst or reactant fully before changing the entire manufacturing process (Meyer 1979).
- The length and magnitude of temperature and pressure during manufacture affect polymerization and, therefore, free formaldehyde levels. Myers (1984a) shows that increasing press time and/or temperature lowers emissions, especially for high F:U ratio resins.
- Manufacture may include a final step designed to mitigate formaldehyde off-gassing, such as treatment with scavengers or improved/extended curing.

The variables listed above are generalizations; Meyer (1979) lists the following as manufacturing parameters affecting formaldehyde release:

resin components
resin component ratio
resin application
hardener components
hardener quantity
press temperature
press duration
wood species
wood moisture
resin concentration
resin viscosity
wood chip size
wood chip diameter

Wood chip geometry was specifically studied by Christensen et al. (1981) as a factor in emission rate. Formaldehyde emission from particleboard surfaces decreased in direct proportion to an increase in wood chip particle size.

Not specifically listed above is a parameter Myers (1982a) discusses: the curing process, a combination of the catalyst (or pH), press time, press temperature, and other manufacturing conditions. Myers' review of the data was performed in an effort to relate cure with releases by resin hydrolysis. Quadrupling curing time (from 5 to 20 minutes) was found to decrease emissions approximately two-fold, by resulting in 2 to 5 times stronger bonds in the cured resin. Decreasing cure temperature from 40°C to 23°C resulted in a two-fold increase in bond stability with a corresponding six-fold decrease in formaldehyde emission under test conditions. Increasing the pH from 3.0 to 6.5 caused an increase in bond strength of a factor of 10; the cured resin showed a two-fold reduction in emissions.

(4) Age of the Product. Under normal use conditions, the release of formaldehyde decreases with time, as discussed previously. Emission reductions linked to product aging relate to a decrease over time in both the formaldehyde present in the board as a residual from manufacturing and the latent formaldehyde present in the board in hydrolytically labile resin and wood components. The emission rate decay curve for a board is

apparently exponential with time; the residual formaldehyde is emitted at relatively high rates followed by a slow release of latent formaldehyde. Although the short-term emission rate behavior of boards has been reported in numerous studies, little quantitative information is available on the long-term emission rates, as was discussed in Section 2.2.

Meyer and Hermanns (1984a) state that the emission rate may slow by a factor of two within the first three weeks after a product is manufactured (if allowed to aerate properly). They present Figure 2 to support this. The data upon which this figure is based were not presented by the authors; the long-term emission rate behavior may well be mathematically predicted.

ORNL is currently conducting experimental chamber studies for CPSC designed to measure the decay of formaldehyde emissions from a combination of particleboard, hardwood plywood paneling, and MDF under controlled environmental conditions (23°C and 50% relative humidity) over a period of one year. The board locations and loadings in the chambers are designed to approximate consumer use conditions.

One study, the "slow" decay study, is being conducted with a relatively low air exchange rate in the chamber (0.4 air changes per hour). The elevated formaldehyde concentrations that are anticipated with low ventilation should reduce the emission rates of the pressed wood products and thus lengthen their decay period. (The effect of background formaldehyde concentration on emission rates is explained in the following section). Preliminary results indicate that the decay period to 1/e of the original chamber formaldehyde concentration (i.e., 0.37) is greater than one year (Matthews et al. 1982-1984).

The second study, the "fast" decay study, is being conducted with relatively high air exchange rates designed to keep the chamber formaldehyde concentration at or below 0.1 ppm. The low background formaldehyde concentration should increase the emission rates and thus

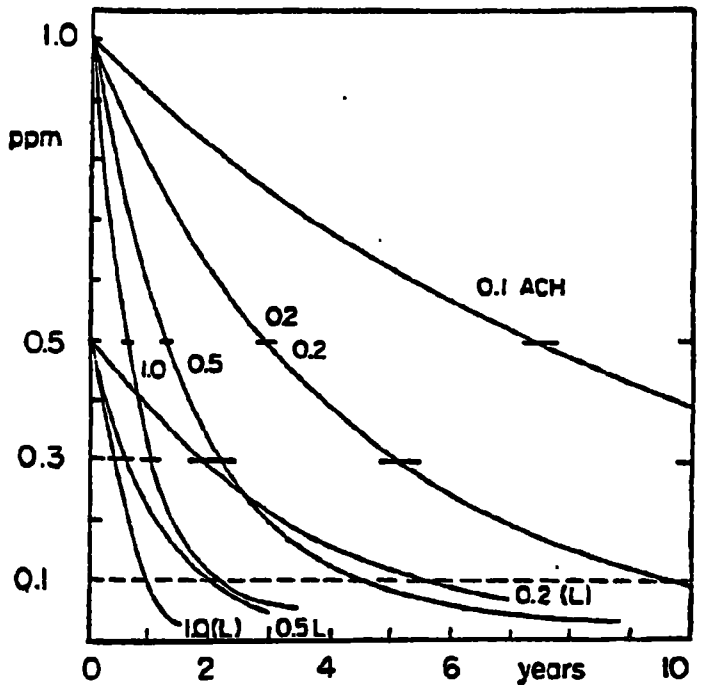


Figure 2. Predicted Formaldehyde Release as a Function of Age and Ventilation Rate for Two Particleboards, One with an Initial Emission Rate of 1.0 ppm and One with an Initial Emission Rate of 0.5 ppm

Source: Meyer and Hermanns (1984a).

shorten the decay periods relative to those observed in the "slow" decay study. Preliminary results indicate that the decay periods to 1/e of the original emission rates (i.e., 0.37) are less than one year for most boards tested (Matthews et al. 1982-1984).

2.3.2 Environmental and Architectural Factors

Factors not specific to the type of pressed-wood product may also affect formaldehyde release and may often be the overriding determinants of release.

(1) Temperature and Humidity. The effect of temperature and, to a lesser extent, the effect of humidity on formaldehyde emission from pressed wood products have been investigated by numerous researchers. The results of these studies indicate that formaldehyde emission depends strongly on temperature and moderately on humidity. Most of the investigations have involved emission rate testing of products within laboratory chambers, although a few have involved measurements of temperature effects in homes. Myers (1984b) recently reviewed all available data that had been reported during the period 1960 through May 1984. The results of his review and analysis are summarized below. The effects of temperature are addressed first, followed by the effects of humidity.

Researchers, most notably Berge et al. (1980), have described the temperature dependence of formaldehyde emission in exponential terms. Myers (1984b) concluded that an exponential Arrhenius type expression does provide adequate representation of this strong temperature dependence.

$$C_m = C_c \times e^{-R\left(\frac{1}{T_m} - \frac{1}{T_c}\right)}$$

or by rearrangement

$$C_c = C_m \times e^{-R\left(\frac{1}{T_c} - \frac{1}{T_m}\right)}$$

where

C_m = measured formaldehyde concentration
C_c = corrected formaldehyde concentration
R = temperature coefficient
T_m = measurement temperature (°K)
T_c = new temperature (°K)

Because of this strong temperature dependence, Myers (1984b) concluded that meaningful comparisons of emission rate data or home air levels necessitate that the measurements be made at a standard temperature (preferably 25°C) or that the measured values be corrected to the standard temperature using the equation above. Except for those laboratories that have consistently observed a particular temperature coefficient (i.e., the R value) from board tests, Myers (1984b) recommends that a temperature coefficient of 8930 be used for correcting measured home air levels and emission rate chamber data to a standard temperature of 25°C. This value was obtained by a statistical analysis of all chamber test data as a composite set (normalized to unity at 25°C), with ~ 95 percent confidence interval of 8390 to 9470 ($\pm$ 6 percent relative error). Berge et al. (1980) reported a temperature coefficient of 9799, which has been used by other researchers.

The statistical analysis also indicated that, although the temperature response of different boards can differ significantly, it is not clear if there are significant differences in temperature response between board types (i.e., particleboard versus hardwood plywood paneling), between "low" and "high" emission boards, or between chamber tests and homes.

Researchers have found that the humidity dependence of formaldehyde emission is much weaker than the temperature dependence and, at present, can best be described in linear terms. Myers (1984b) concluded that the linear expression below does describe the humidity dependence based on the limited data available and that a more complex model is not warranted at this time because of the uncertainties surrounding measurements of humidity dependence.

$$C_m = C_c \times [1 + A (H_m - H_c)]$$

or by rearrangement

$$C_c = C_m [1 + A (H_c - H_m)]$$

where

C_m = measured formaldehyde concentration
C_c = corrected formaldehyde concentration
A = humidity coefficient
H_m = measured relative humidity (%)
H_c = new relative humidity (%)

The response of board formaldehyde emission to humidity changes is more complex and less well understood than board response to temperature change. Fewer investigators have studied this dependence and Myers (1984b) states that the tested boards may not have achieved equilibrium or steady state by the time the concentration measurements were made, because of the very slow (sometimes weeks or more) and erratic response of boards to humidity change. This may explain, in part, the wide variation (almost tenfold) in the humidity coefficients (i.e., A values) measured in the various studies reviewed by Myers (1984b).

Similar to the statistical analysis performed on the temperature dependence data, Myers (1984b) performed an analysis on the humidity dependence data (normalized to unity at 50% relative humidity) that yielded a composite humidity coefficient of 0.0195 with a 95 percent confidence interval of 0.014 to 0.025 ($\pm$ 28 percent relative error). However, because of the large variations in the humidity coefficients measured by different investigators and between different boards, Myers (1984b) expressed less confidence in the use of this humidity coefficient to correct emission rate and in-home air measurements to standard conditions than in the use of the composite temperature coefficient. Berge et al. (1980) reported a humidity coefficient of 0.0175, which has been used by other researchers.

Table 4 provides an indication of the variability in formaldehyde levels that potentially could result from changes in temperature and relative humidity.

(2) Barriers. Paints and coatings have long been used on fiberboard and particleboard for decorating purposes and to render them somewhat water-resistant; those substances are said to be effective barriers to formaldehyde release (Meyer 1979, NPA 1984). Painting or coating a surface effectively lowers the porosity of the material, hinders diffusion of formaldehyde out of the wood, and slows moisture accumulation in the wood (which may cause hydrolysis or transport of formaldehyde with water vapor).

Meyer's 1979 publication also lists waxes; gypsum board; and paper, plastic, and metal laminates as effective barriers to formaldehyde emissions. These barriers may manifest themselves in homes as tile flooring or simulated wood counter or furniture surfaces. In fact, almost every overlayment (including carpet) or surface treatment affects formaldehyde release. Only in a very few cases has that effect been quantified; systematic, complete data are not available.

Pickrell et al. (1984) do provide some quantitative data on the effect of carpet and insulation as barriers. The release rate of carpeting over particleboard was 73 percent of the rate for particleboard alone. Similar results were obtained for other product/barrier combinations.

(3) Background formaldehyde concentration. The background level of formaldehyde has been found to be a major factor affecting emission rates. The primary factors controlling the background level are (1) ventilation rates and (2) interrelationships among numerous sources (and sinks) of formaldehyde.

Mobile homes generally have low air exchange rates relative to conventional homes, which exacerbates formaldehyde exposure. The average exchange rate in mobile homes is 0.35 changes per hour (University of

Table 4. Potential Effects of Temperature and Relative Humidity Changes on Formaldehyde Air Concentrations (ppm)*

Temperature	Relative humidity				
	30%	40%	50%	60%	70%
59°F (15°C)	0.08	0.11	0.14	0.17	0.19
68°F (20°C)	0.15	0.19	0.24	0.29	0.33
77°F (25°C)	0.24	0.32	0.40	0.48	0.56
86°F (30°C)	0.40	0.53	0.66	0.79	0.92

*Calculated using equations in Section 2.3.2(1) which were developed primarily from data on new pressed wood products and new homes. Assumes a temperature coefficient of 8,930 and a humidity coefficient of 0.0195. Assumes a base formaldehyde measurement of 0.40 ppm at 25°C and 50 percent relative humidity.

Texas 1983), while the rate in conventional homes may range from less than 1 change to 10 changes per hour. Lower rates in localized areas with poor mixing (such as closets) may lead to higher localized formaldehyde levels.

Myers (1984c) and Myers and Nagaoka (1981a) discuss the effect of ventilation rate on formaldehyde air levels. Myers and Nagaoka experimentally validated some predictive equations that show an exponential decline in concentration with increase in ventilation rate. Myers' (1984c) literature review points out that existing data must be regarded as semi-empirical because most data are from controlled chamber tests, and extrapolation to dwellings is not reliable. Moreover, test results are almost exclusively for single products; actual homes will have a complicating array of pressed-wood and other formaldehyde sources.

The interrelationship between numerous formaldehyde sources in homes is not well-understood. Meyer and Hermanns (1984a) state that the strongest-emitting product may be essentially the only active source of emissions at some point in time, and that other UF-bonded products may act as sinks by absorbing excess formaldehyde. If that source were removed, then theoretically the next strongest formaldehyde emitter would become the source rather than a sink. This interrelationship is one subject of an ongoing research effort by the Consumer Product Safety Commission. Products other than UF-containing materials also can act as formaldehyde absorbers or sinks. Formaldehyde is such a reactive chemical that an almost limitless variety of reactions with structural components, consumer products, or indoor air pollutants can be imagined.

2.4 Formaldehyde Emission Rate Testing Methods

2.4.1 Background

There are five basic types of formaldehyde collection methods that have been developed to measure formaldehyde emissions from pressed wood products. These methods can be categorized as equilibrium, static, dynamic air flow, distillation/extraction, and passive. The Hardwood

Plywood Manufacturers Association (HPMA), National Particleboard Association (NPA), and the Formaldehyde Institute (FI) identified 34 potential testing techniques within these 5 basic method types in 1979 (HPMA, NPA, FI 1979). It is the purpose of this section of the report to review the test methods most widely used today with regard to formaldehyde collection and analytical procedures utilized and advantages/disadvantages of the methods. Also, correlations between methods, if any, will be discussed.

2.4.2 Methods

(1) Static test method. The static test method, or desiccator test, is a test method that has no air passed through for collection of formaldehyde, but utilizes an aqueous medium for the collection of formaldehyde. It is a destructive method in that it requires that small samples be removed from large formaldehyde-bearing materials for testing. In this test, formaldehyde is continuously emitted from the samples and absorbed into the aqueous medium. This type of test, in particular the Japanese Industrial Standard (JIS) and a version of the JIS (FTM-1), has been used extensively in the U.S. as a quality control test method for wood products containing formaldehyde.

The desiccator test apparatus consists of a glass desiccator with a secure, close fitting top; a sample rack to hold samples in the desiccator; and a beaker or desiccator plate to contain the aqueous collection medium (distilled water). The formaldehyde-containing samples are placed in the rack above a specified quantity of distilled water. Once the cover is placed in position, the test is underway. Figure 3 presents a view of the JIS desiccator test apparatus, which is generally representative of desiccator systems. The temperature of the system, typically about 75°F, is maintained for the duration of the test period, which may vary from 2 to 24 hours. Data are reported in units of concentration, such as micrograms of formaldehyde per milliliter of distilled water. The total volume of water and mass of product tested

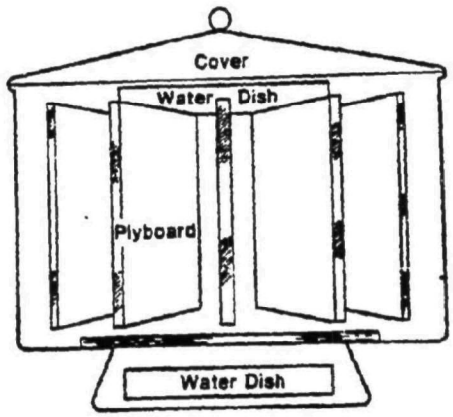


Figure 3. JIS Desiccator Test Apparatus

Source: Pickrell et al. (1982).

must be known to convert results to mass formaldehyde per mass of pressed-wood products.

Another static method, developed by Roffael (1978) and used widely in Europe, is known as the WKI method. The WKI method apparatus is a simple one, consisting of a polyethylene bottle; a tight fitting bottle cap; and an assembly with which to hold the material samples containing formaldehyde in suspension within the sample bottle (see Figure 4). Two samples of formaldehyde-bearing material are attached to the suspension assembly and placed above 50 ml of distilled water contained within the bottle. The distance between the suspended samples and the water surface has not been deemed to be a significant factor for the experiment. The samples should be cubes, with 2.5 cm (1") sides. When all apparatus are in place, the bottle is tightly sealed with the bottle cap. The test is performed at 40°C for different time intervals. Roffael presented his results in units of milligrams of formaldehyde per 100 grams of sample.

Advantages in using a static air test method include ease of operation, since neither air flow control nor actual experiment observation is required. Manpower is needed only for apparatus set-up and analysis of data. Low levels of formaldehyde can be detected by increasing the duration of the test.

Disadvantages of this type of test include the problem of induced high humidity in the sample chamber due to a stagnant atmosphere. This may alter the moisture content of the sample, thereby causing a variation in formaldehyde emissions and the condensation of water vapor on the walls of the chamber; test samples may also act as a formaldehyde sink during the test. Care must be taken to ensure that particles from the sample do not fall into the collection medium, which would lead to abnormally high formaldehyde concentrations in the collecting medium. Because of the small size of the wood samples tested, an atypically large amount of board edge surface is exposed; unless the edges are sealed, the edge releases will dominate the test results. Finally, the test must be

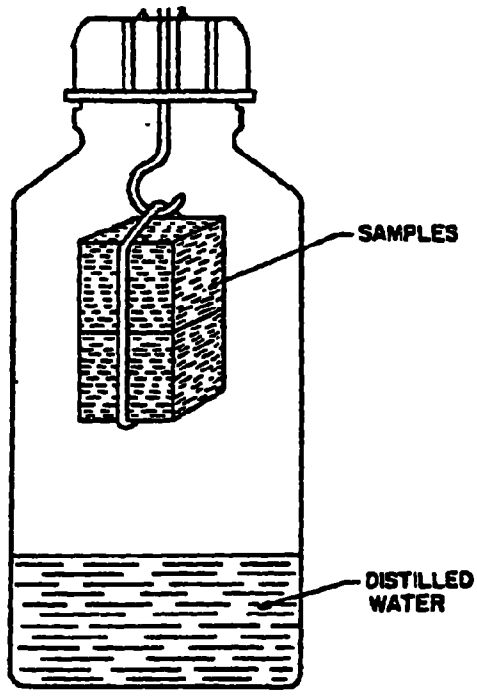


Figure 4. WKI Test Apparatus

Source: Roffael (1978).

timed accurately to obtain proper results because formaldehyde concentration in the collecting medium is a direct function of time.

The quantity of formaldehyde emitted from the sample, which would eventually be absorbed in water medium, can be determined by one of four analytical methods:

(1) Chromotropic acid procedure

(2) Purpald procedure

(3) Acetylacetone procedure

(4) Pararosaniline procedure

For static tests, depending on the application, any of the four tests can be used. According to data presented by the Hardwood Plywood Manufacturers Association (HPMA) in their July 1984 submission to EPA, the chromotropic acid and acetylacetone methods are used most in static tests. All four tests are valid analytical techniques for formaldehyde, though with different common detection limits and prone to different interferences. The chromotropic acid method is subject to interferences from NO_2, alkenes, acrolein, acetaldehyde, and phenol; pararsoaniline has few (if any) interferences; the purpald test may record higher aliphatic aldehydes; and the acetylacetone test is specific to formaldehyde.

Formaldehyde data obtained from the desiccator and WKI tests may be presented in units of formaldehyde concentration in solution or as an average emission rate per mass of particleboard.

(2) Dynamic test method. The dynamic air flow test method uses a system that passes air at a controlled rate through a chamber containing the samples to be tested. The released formaldehyde is carried by the air out of the chamber. The dynamic test models the actual home air contamination process by measuring the formaldehyde concentration in the test chamber atmosphere. Therefore, this test constitutes a primary characterization of formaldehyde emissions from formaldehyde-containing

materials. Dynamic chamber tests may be conducted with large chambers, capable of holding a full size sample of particleboard, or with small scale-laboratory scale apparatus. Since many of the test procedures are laboratory scale, this discussion will focus on the small-scale laboratory apparatus and results obtained from a small-scale unit.

The apparatus for the dynamic chamber test consists basically of a large standard desiccator within which the formaldehyde-bearing samples are contained (see Figure 5). The desiccator is fitted with a lid equipped with inlet and outlet nozzles for the circulation of air. During the experiment, air is pulled from the outside atmosphere into the chamber. This air mixes with the air in the chamber and exits the chamber through the exit nozzle. The rate at which air is circulated through the chamber is controlled to within $\pm$ 5 percent. A small fan or stirring bar can be used to circulate air within the chamber. Dynamic experiments are generally conducted under conditions of constant temperature and humidity. Assuming that good gas mixing occurs within the chamber, the effluent gas will be representative of the chamber gas. At various time intervals, the effluent gas is scrubbed through a prescribed quantity of distilled water and the absorbed formaldehyde is analyzed. Test results are reported in parts per million of formaldehyde in chamber air.

A primary advantage of this type of test is that, unlike the desiccator test, it can be used to simulate real world conditions. The dominant factors affecting the emission rate of formaldehyde from a given board (i.e., temperature, humidity, product loading, and ventilation rates) can all be varied using this method.

Disadvantages of this test include the need for precise measurement of air flow rate. A constant prescribed air flow rate must be maintained; erroneous test results will otherwise be obtained. The environmental conditions within the system must be maintained constant throughout the test, thus making this test more manpower-intensive than the static test.

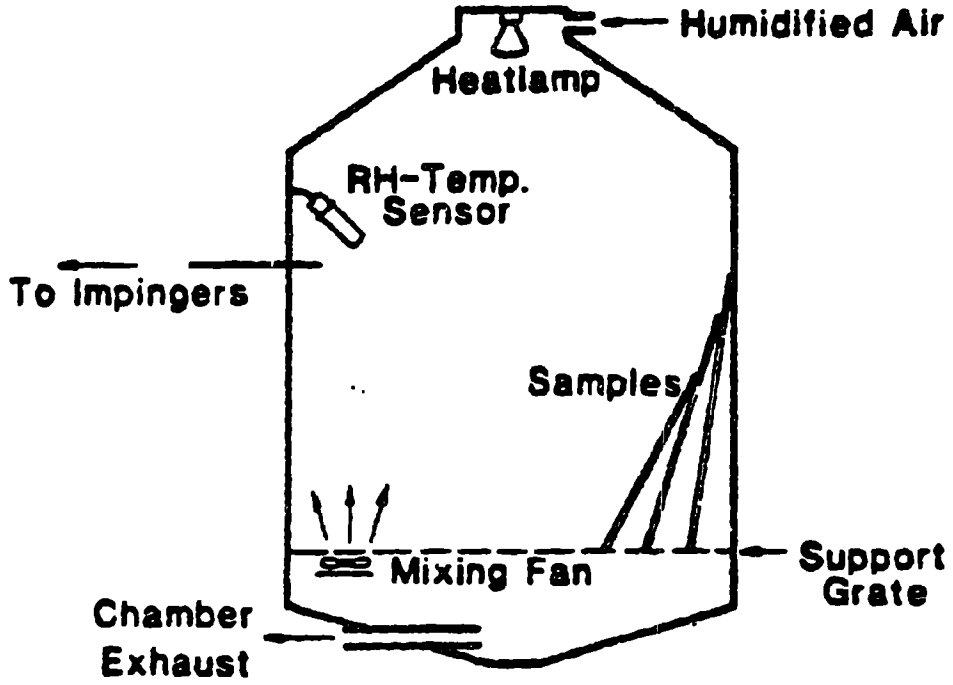


Figure 5. Dynamic Chamber Test Apparatus

Source: Pickrell et al. (1982).

The impinged solution can be analyzed using any of the four analytical methods mentioned previously. According to data presented by the HPMA, the chromotropic and purpald methods appear to be the most widely used. Again, as with the static tests, the chromotropic acid method has been recommended by NIOSH for the detection of formaldehyde.

(3) Distillation/extraction test methods. The distillation/extraction test method, also known as the perforator test method, is unlike either of the two previous test methods. Rather than using air as a transport or collection medium, a boiling solvent such as toluene is used to capture formaldehyde. The system, as defined by Myers (1983), involves a 2-hour reflux in boiling toluene with a prescribed mass of samples. The samples are placed in the boiling toluene, and the reflux, which contains the formaldehyde, is bubbled through distilled water. The distilled water extracts any formaldehyde in the toluene reflux. The toluene vapor, which is free of formaldehyde, is then condensed and returned to the boiling toluene pot. Final analysis of the water leads to the perforator value, generally expressed as milligrams of free formaldehyde per 100 g of dry board.

Advantages to this type of test include the fact that test results are generally reproducible, the test is fast, and it requires no preconditioning of the samples and no temperature or relative humidity control. Also, considerable work has been done by the Europeans using this test as a quality control and regulatory method.

Disadvantages in using this type of test include the potential generation of false formaldehyde results. Due to the rigorous test conditions and high temperatures involved, formaldehyde that under normal ventilation situations would not be emitted may be released. Thus, erroneous formaldehyde readings may be obtained. This was substantiated by Roffael (1978) when he attempted to correlate his WKI method with the perforator test. He stated that even in cases where no formaldehyde binder was present in the sample, the perforator test still reported, or

detected, formaldehyde in the sample. This fact, plus the inability to vary test procedures, are chief drawbacks to this test.

The water samples obtained with the perforator test can be tested by any one of the four analytical methods. In his experiments, Myers (1983) used acetylacetone to obtain test results. He found that trioxane emitted by the boards through the distillation process would be analyzed as formaldehyde by the chromotropic acid method, but not by the acetylacetone method.

(4) Formaldehyde surface emission monitor (FSEM). The formaldehyde surface emission monitor (FSEM) is a device that allows for the non-destructive measurement of formaldehyde emission rates from formaldehyde-bearing materials. The FSEM has been developed by Matthews et al. (1983, 1984) to address a need for a semi-quantitative, non-destructive measurement of formaldehyde emission rates from full-scale formaldehyde bearing objects.

The main components of the FSEM (Figure 6) are a 20 cm brass mechanical sieve, (No. 20 mesh- 0.0331 inch openings) and a brass cover. It is within this compartment that the formaldehyde sorbent is contained when it is in operation. A circular flange, consisting of plexiglass and neoprene, attached to the sieve provides for a seal between the outside atmosphere and the test atmosphere. The separation distance between the mesh surface and the emitting material is about 2.3 cm. The brass cover is mechanically clamped to the sieve to ensure that contamination of the sorbent from the outside atmosphere is avoided. In order to use the FSEM, the solid sorbent material is sprinkled in a uniform manner on the screen of the mechanical sieve assembly. The sieve is then sealed for the duration of the two-hour test period. At the conclusion of the test, the sorbent is washed with distilled water and the solution is filtered and assayed for formaldehyde content using a pararosaniline method. Results from this stage of the test are presented in grams of formaldehyde per milliliter of solution tested. Matthews, concerned

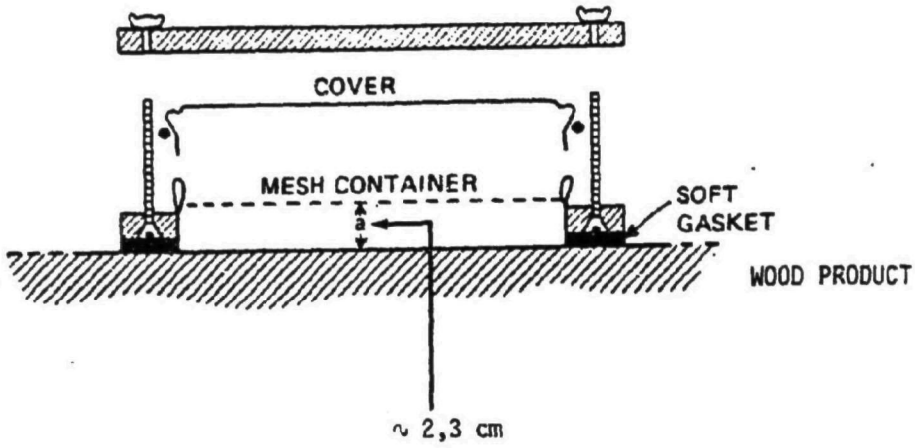


Figure 6. FSEM Test Apparatus

Source: Matthews et al. (1984).

primarily with emission rates, provided an algebraic relationship to determine the emission rate:

$$\text{Formaldehyde emission rate } (mg/m^2/h) = \frac{\text{Concentration of formaldehyde found } (mg/ml) \times \text{Rinse volume } (ml)}{\text{Sampling period } (h) \times \text{Sample area } (m^2)}$$

The major advantages to this test method are that it is non-destructive and portable, thus potentially enabling emission rate testing to be conducted on-site (e.g., in the home). The major disadvantage to this method is that it has not yet been fully validated. The effects of equipment design and environmental conditions on operation results have not been completely resolved.

2.4.3 Inter-Method Correlations

Correlations or agreement between data generated by various test methods are presented in this subsection. It should be noted in reviewing the data that most correlations apply to specific test conditions and specific sets of products. The exact correlation between various test methods is complex and not yet fully established (Meyer et al. 1983) The same correlation may not be valid when, for instance, the resin formulation is changed, the wood species differs, the board finish or top coat is modified, the pre-test conditioning of the boards is altered, or boards have been manufactured by different facilities (even in similar manners) (Meyer et al. 1983).

In addition, it should be realized that the various test methods may very well be measuring formaldehyde emission generated by different mechanisms (Myers 1983). The perforator method, according to Myers, measures free formaldehyde but may also be measuring formaldehyde generated by hydrolysis as a result of the testing. Desiccator tests measure only the easily liberated portion of the free formaldehyde, although the test, if sufficiently prolonged, may also cause resin

hydrolysis. The dynamic tests measure varying combinations of initial free formaldehyde and hydrolytically produced formaldehyde depending upon exposure time and conditions (Myers 1983).

Although approximate linear correlations with relatively high correlation coefficients have been found between various methods, large deviations occured from the linear regressions. Myers (1983) concluded that without large safety factors and/or much testing to clarify those deviations neither the desiccator nor perforator test should be used as product standard test methods; product standards should be based on dynamic chamber tests.

Myers and Nagaoka (1980) summarized inter-method correlations for six formaldehyde emission tests: two desiccator tests (NPA and JIS), the perforator test, the jar (WKI) test, a paper sorption test, and a dynamic chamber method. They found nearly perfect cartesian correlation (r^2 of 0.99) between results obtained via the two desiccator test methods. Close agreement between different static tests is not unexpected; these investigators also found a close correlation ($r^2 > .96$) for results obtained via jar and NPA 2-hour desiccator tests. An interesting correlation is found with the results of the seldom-used paper sorption test and dynamic chamber test. The paper test involved stacking filter paper between wood samples, then extracting the formaldehyde from the paper and using any of the common analytical methods. With ten data points, a correlation coefficient of 0.99 was reported, with only 2 percent error; this was the closest correlation found by Myers and Nagaoka (1980).

Data correlations between static and other formaldehyde test methods have been developed by many scientists. Myers and Nagaoka (1980, 1981b) have produced a relationship from data obtained in a JIS test and dynamic test. Presented in Figure 7, these tests were conducted with fresh particleboard at conditions of 25°C and 75 percent relative humidity (RH) and displayed excellent agreement ($r^2 = 0.965$) (Myers and Nagaoka

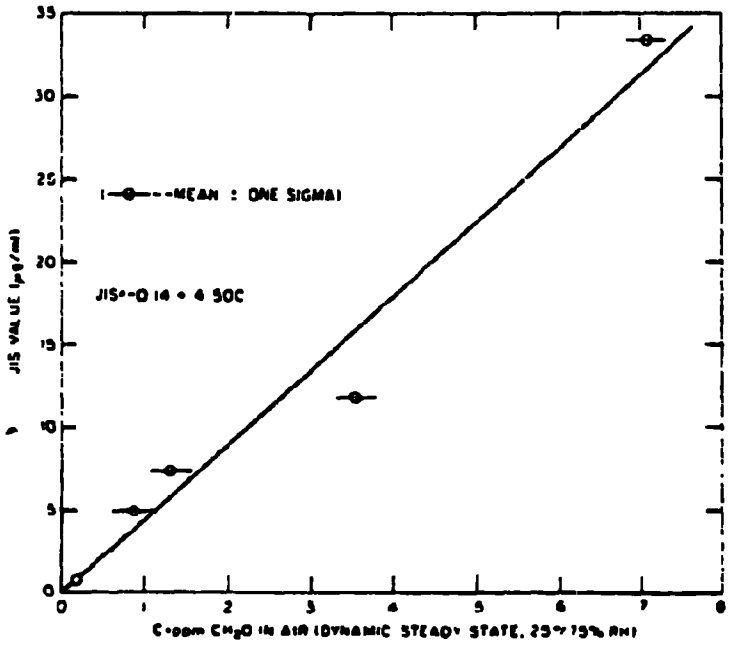


Figure 7. Desiccator Test and Dynamic Test Data Correlation

Source: Myers and Nagaoka (1981b).

1980). A similar correlation developed by Myers (1983) shows that data obtained from a 2-hour desiccator test and a dynamic chamber test are in close agreement (r^2 = 0.87). This correlation is given in Figure 8. Test conditions were 25°C and 50 percent RH.

Roffael (1978) was able to correlate data obtained from his WKI method and that of a dynamic chamber. Again, various particleboards under similar conditions (23°C/45% RH) were tested. The results are graphically presented in Figure 9.

Roffael also found that a correlation exists between data obtained by the WKI test method and the extraction/distillation (perforator) method. Using similar formaldehyde samples and testing for periods of 24- and 48-hours, two correlations were found to exist. They are given in Figures 10 and 11.

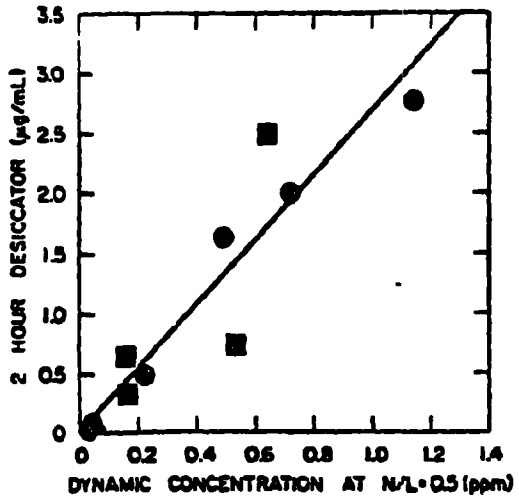
A data correlation between the dynamic test method and the extraction/distillation (perforator) method was also obtained by Myers (1983). At conditions of 25°C, 50% RH, and a board loading factor (N/L) of 0.5, a near linear correlation was obtained and is presented in Figure 12.

Matthews (1984) identified a correlation between his FSEM method and that of a dynamic test chamber. Test conditions were set at 23°C and 50 percent RH. Graphed data results are given in Figure 13.

2.5 Formaldehyde Emission from Conventional Pressed-Wood Products

Numerous investigators have measured formaldehyde emissions from pressed-wood products over the past ten years. The changing nature of pressed-wood products, however, renders data obtained prior to 1982 relatively obsolete in determining baseline formaldehyde emissions for currently-marketed products.

During the period 1980 through 1983, three major surveys were conducted to characterize formaldehyde emissions from pressed wood products. NPA conducted industry-wide surveys of particleboard and MDF

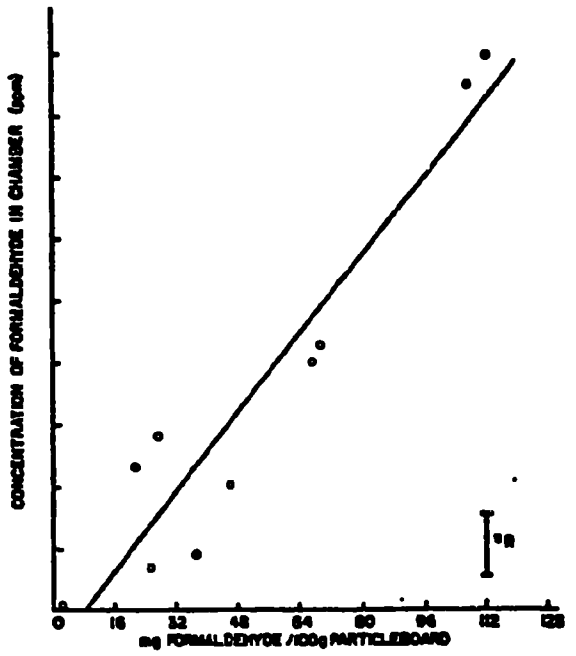


Relation between 2-hour desiccator values and dynamic concentration at 25° C/50% RH and $N/L = 0.5$ (Group 1 boards: O = plywood; □ = particleboard. Δ = PF particleboard.)

NOTE: N/L is loading factor - N is ventilation rate (HR^{-1}), L is board loading (M^2 board area/M^3 chamber volume).

Figure 8. 2-Hour Desiccator Test and Dynamic Chamber Test Data Correlation

Source: Myers (1983).



NOTE: Values on the Y-AXIS are not given. Roffael stated that he was unable to provide these values due to an agreement between himself and the German Particleboard Association.

Figure 9. WKI - Dynamic Chamber Correlation

Source: Roffael (1978).

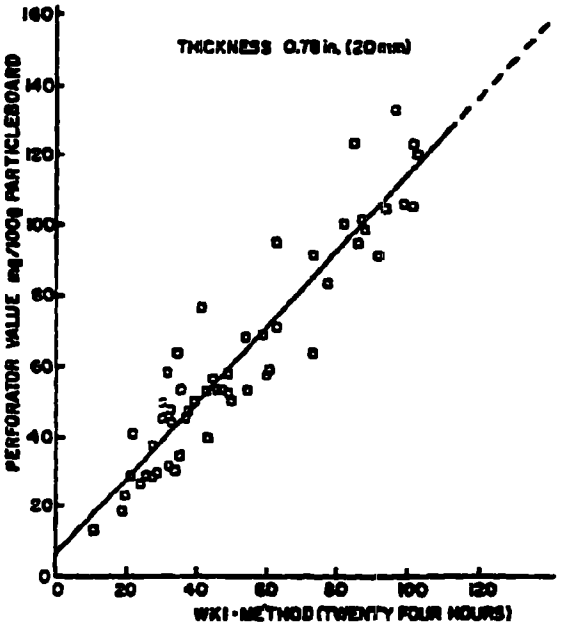


Figure 10. WKI Method and Distillation/Extraction Method Data Correlation (24 Hour Test Period)

Source: Roffael (1978).

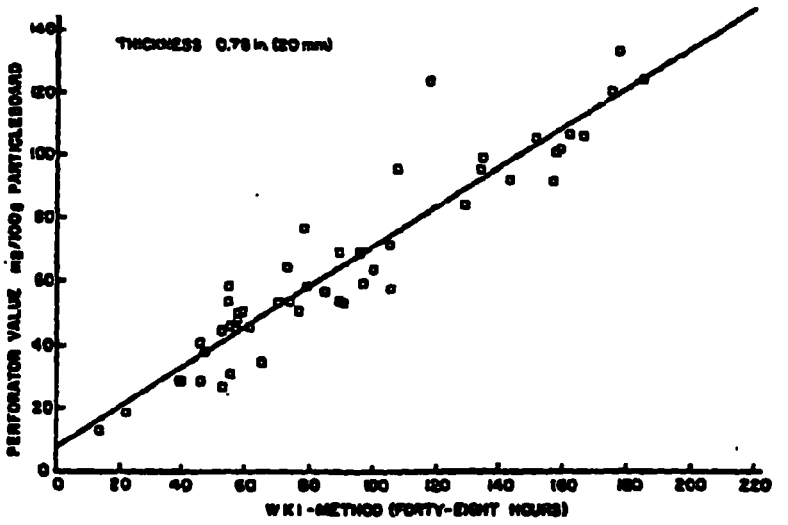
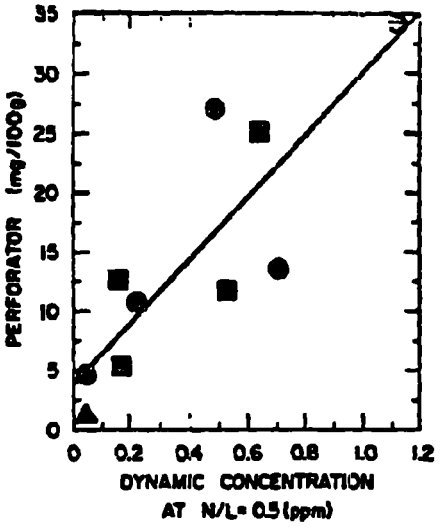


Figure 11. WKI Method and Distillation/Extraction (Perforator) Method Data Correlation (48 Hour Test)

Source: Roffael (1978).



Relation between perforator and dynamic concentration at N/L=0.50, 25°C/50% RH. (Group 1 boards. ○=plywood; □=particleboard; △=PF particleboard.)

Figure 12. Dynamic Test Method and Distillation/Extraction Test Method Data Correlation

Source: Myers (1983).

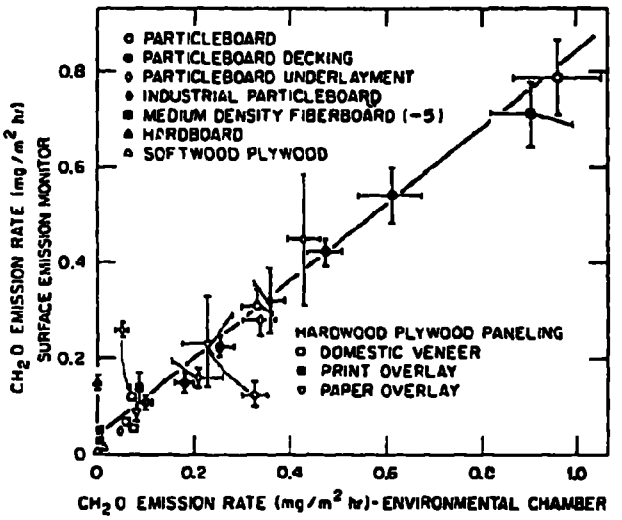


Figure 13. FSEM Test Method and Dynamic Test Method Data Correlation

Source: Matthews et al. (1984).

during 1980 and 1982. In 1983, CPSC conducted a survey of particleboard, MDF, and hardwood plywood paneling produced by the top three manufacturers of each product type. Although the three surveys used different emission rate test methods and, to a varying extent, tested boards produced by different (and unidentified) manufacturers, the results of these three surveys provide the best available information characterizing the extent of formaldehyde emissions from pressed wood products marketed during the 1980's. The survey designs and results are discussed below. Comparison of the NPA and CPSC results are made where appropriate.

2.5.1 1980 and 1982 NPA Surveys (NPA 1984)

During 1980 and 1982, NPA requested particleboard and MDF samples from members and non-members of NPA. Each participating plant was requested to supply two samples of each of the major product types produced by the plant. The samples were requested to be finished products ready for sale and of recent manufacture.

A total of 47 products submitted by 32 different plants were tested in the 1980 survey. A total of 62 products submitted by 38 different plants were tested in the 1982 survey. Table 5 presents additional information on the number of plants producing various product types that participated in the survey. The table also gives some indication of the extent of industry participation in the surveys by comparing the number of participants to the number of plants in the industry. Because the identities of the participants are confidential, it is not possible to determine what percentage of actual industry production volume or capacity was represented by the surveys.

The 1980 survey was performed using the 24-hour dessicator test, which was the standard test method for the industry at that time. The 1982 survey was performed using the 2-hour dessicator test which, by 1982, had become the industry standard test. In order to determine if a correlation could be obtained between the results of the two test

Table 5. Summary of Plant Participaton in NPA's 1980 and 1982 Surveys[a]

Product type	No. participating plants[b] 1980	1982	No. of repeats[b,c]	No. of plants in industry in 1983
Particleboard (combined)	(27)	(32)	(22)	(45)
- Mobile home decking	10	13	9	unknown
- Underlayment	15	14	9	unknown
- Industrial	16	27	15	unknown
MDF	3	4	2	11
Mende board[d]	2	2	2	8

[a]Source: NPA (1984).

[b]Combined number of particleboard plants is less than the sum of the individual product plants because many plants supplied more than one product type.

[c]Indicates the number of plants that definitely participated in both the 1980 and 1982 surveys.

[d]A type of particleboard defined by NPA (1984) as 1/8 to 1/4 inch thick board produced by pressing a continuously moving ribbon of resin-coated particles.

methods, 56 of the 62 products collected in the 1982 survey were tested using both the 2-hour and 24-hour test methods. A linear regression analysis of these data yielded the following equation with a correlation coefficient of 0.96:

(2-hour value) = 0.55 x (24-hour value) + 0.24

Using this equation, NPA predicted the 2-hour dessicator values for the products tested in the 1980 survey. Table 6 provides a summary of the average emission rates for each product type with all the manufacturers' data combined for the two surveys. As can be seen in the table, the average dessicator values, as well as the range of values, decreased for all product types between 1980 and 1982. The average decking, underlayment and industrial board test values decreased by 67 percent, 56 percent, and 61 percent, respectively. Figure 14 presents a profile of the dessicator test values for the combined particleboard subsets tested in the two surveys.

As indicated in Table 5, 37 of the same plants that supplied samples for the 1980 survey also supplied samples for the 1982 survey. The boards from 28 of these plants had reduced dessicator values in the 1982 survey. Values increased by 5 percent or less for 4 boards, between 11 and 18 percent for 3 boards, and by greater than 100 percent for 2 boards.

2.5.2 CPSC Pressed-Wood Product Survey (Matthews et al. 1982-1984)

During 1983, the CPSC collected samples of six types of pressed-wood products: particleboard underlayment, industrial particleboard, MDF, and hardwood plywood paneling (3 finishes - ink print, paper, and domestic veneer). Six boards were collected directly from one manufacturing plant of each of the three U.S. manufacturers with the largest volume of sales for each of the six product types. Thus, a total of 108 boards, 18 of each product type, were collected.

The companies whose products were tested were selected because they supply a large proportion of all the pressed wood products purchased by consumers directly or as components of consumer products (e.g.,

Table 6. NPA 1980 and 1982 Survey Summary Results

Product type	Survey	Number of samples	Test results (ug/al)* mean	Stnd. Dev.	Range
Mobile home decking	1980	10	3.61	3.09	0.57 - 9.03
	1982	13	1.18	0.45	0.41 - 2.01
Particleboard underlayment	1980	15	3.99	2.71	1.11 - 10.6
	1982	14	1.74	1.45	0.57 - 6.38
Industrial particleboard	1980	7	5.05	4.87	0.78 - 17.4
	1982	29	1.99	1.06	0.78 - 4.95
MDF	1980	3	7.56	--	1.49 - 13.4
	1982	4	3.77	--	1.36 - 4.92
Mende board	1980	2	5.64	--	5.53 - 5.75
	1982	2	3.87	--	3.22 - 4.52

Source: NPA (1984).

*2-hour dessicator test results. Values for 1980 survey are predicted; see Section 2.5.1 for details.

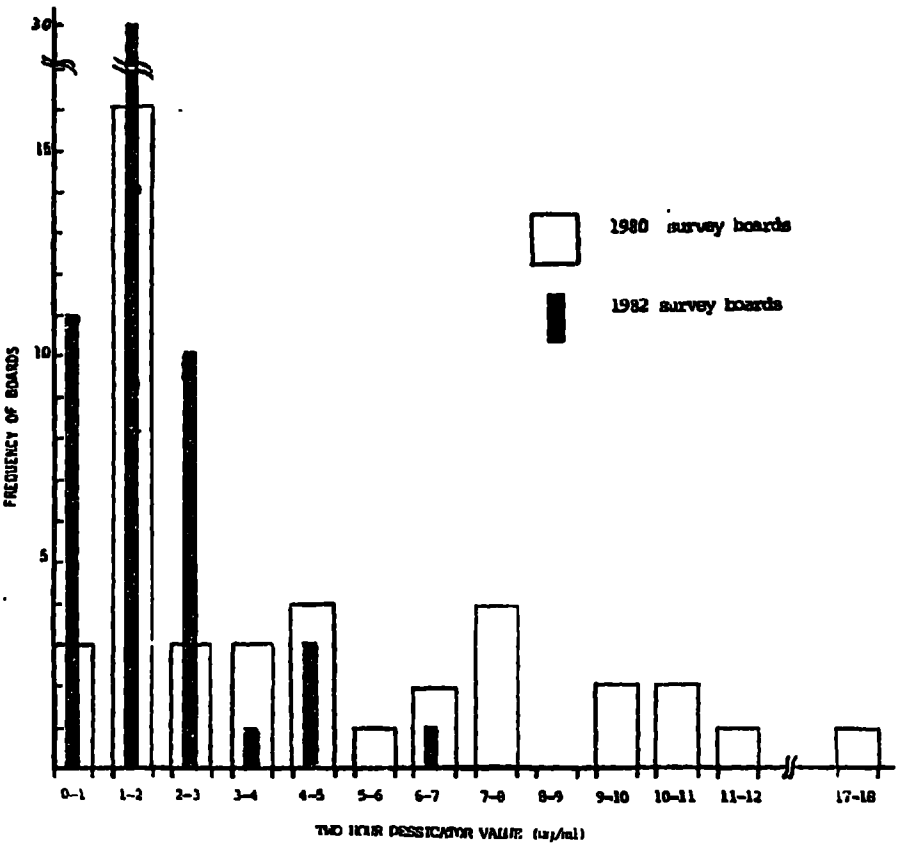


Figure 14. Profile of the Desiccator Values for the Particleboard Products Tested in The 1980 and 1982 NPA Surveys

Source: NPA (1982).

furniture). However, it was noted by CPSC that boards collected from the top manufacturers may represent the state-of-the-art in terms of low formaldehyde-emitting products rather than the actual range of formaldehyde-emitting products on the market.

In order to obtain a random selection of boards available at a plant, boards were selected from lots manufactured (or glued) on at least two different dates. In addition, boards were selected at widely different positions from a bundle of boards manufactured on a given date.

The samples were shipped to ORNL for emission rate testing with the FSEM (see Section 2.4.2 for more information about the FSEM). Prior to testing, the particleboard and paneling samples were conditioned for 2 weeks at approximately 22°C, 50 percent RH, and less than 0.15 ppm background formaldehyde concentration. The MDF samples were similarly preconditioned, except that the conditioning period lasted 3 to 4 weeks instead of 2 weeks. This extended conditioning time probably resulted in a 10 to 20 percent lower formaldehyde emission rate in comparison to what would have been measured after a 2 week conditioning period. Three FSEM measurements were made on each board because of anticipated intra-board variation in emission rates. The paneling samples were tested on the decorative side of the panel.

Table 7 presents a summary of the average emission rates for each product type with all the manufacturer's data combined. For particleboard, the mean emission rates of the underlayment and industrial subsets are very similar. In contrast, the mean emission rate of ink print paneling is more than twice that of both the paper and domestic veneer paneling products. On the average, the emission rates of uncoated MDF boards are about five times higher than the emission rates of the particleboard or paneling products.

Figure 15 presents a profile of the formaldehyde emission rates of the combined particleboard and hardwood plywood paneling boards tested in the survey. Figure 16 presents a comparative profile of the emission rates of the tested MDF, particleboard, and paneling products.

Table 7. CPSC Pressed-Wood Product Survey Emission Rate Summary Results

Product type	No. of Samples	Emission rate ($mg/m^2/hr$)* Mean	Stnd. Dev.	Range
Particleboard (combined)	(36)	(0.30)	(0.18)	(0.11 - 0.78)
- Underlayment	18	0.30	0.22	0.11 - 0.78
- Industrial	18	0.31	0.14	0.15 - 0.62
Paneling (combined)	(54)	(0.17)	(0.14)	(0.03 - 0.63)
- Ink print	18	0.28	0.20	0.05 - 0.63
- Paper	18	0.11	0.07	0.03 - 0.27
- Domestic veneer	18	0.12	0.04	0.07 - 0.24
MDF	18	1.56	0.50	0.57 - 2.30

Source: Matthews et al. (1982-1984).

*FSEM measurement results.

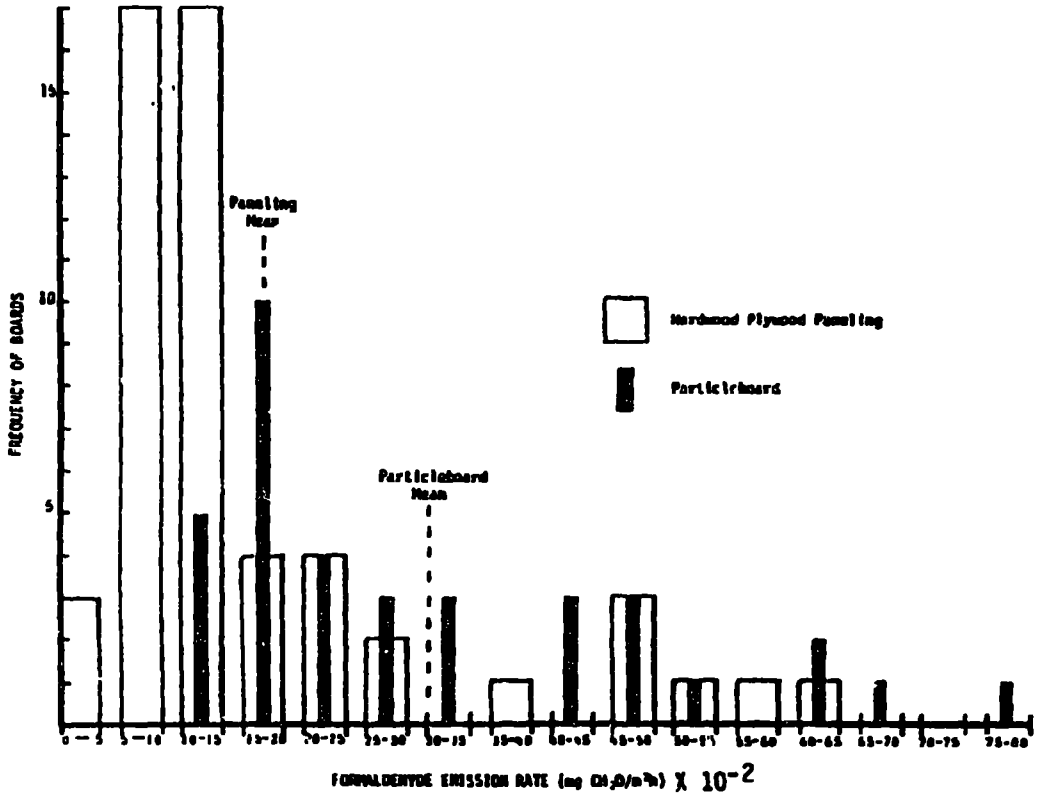


Figure 15. Profile of the Formaldehyde Emission Rates of the Particleboard and Hardwood Plywood Paneling Boards Tested in the CPSC Survey

Source: Matthews et al. (1980-1981).

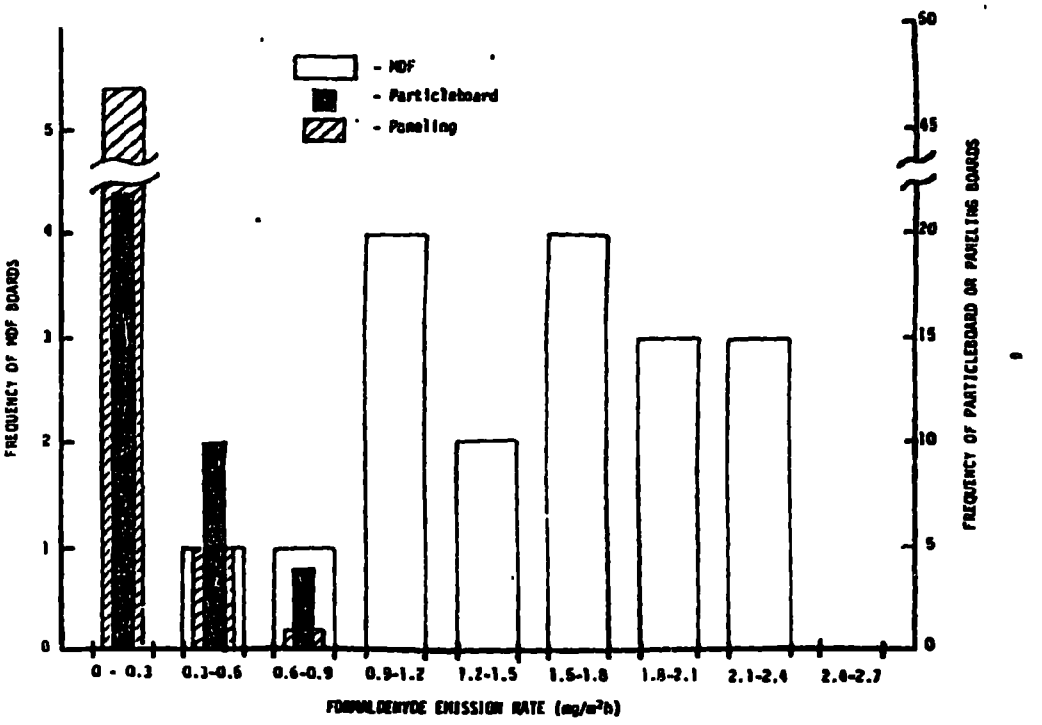


Figure 16. Profile of the Formaldehyde Emission Rates of the MDF, Particleboard and Hardwood Plywood Paneling Tested in the CPSC Survey

Source: Matthews et al. (1980-1984).

To evaluate the interboard variability of measured emission rates for products made by the same manufacturer, Matthews et al. (1982-1984) performed one-way analyses of variance on the emission rate data for each manufacturer (i.e., test results on six boards per manufacturer). The average coefficient of variation for interboard variation was 11 percent for particleboard (range of 0 to 22 percent), 43 percent for paneling (range of 17 to 94 percent), and 18 percent for MDF (range of 12 to 24 percent). These results, summarized in Figure 17, indicate that the manufacturing processes of each of the particleboard and MDF plants surveyed were reasonably consistent; however, the same was not true for the paneling plants.

Comparison of the CPSC and NPA Survey Results

A qualitative comparison between the results of the CPSC survey and the NPA surveys was made by Matthews (1982-1984) and is shown in Table 8. The interlaboratory comparison suffers from: (1) a semi-quantitative intermethod correlation between the 2-hour dessicator test and the FSEM*; (2) the change in formaldehyde emissions with newer products; and (3) the possible bias of the CPSC survey towards sampling of state-of-the-art boards rather than the entire range of products marketed in 1983. The results of the comparison may indicate significantly improved products in the 1983 CPSC survey in comparison to products tested in the 1982 NPA survey.

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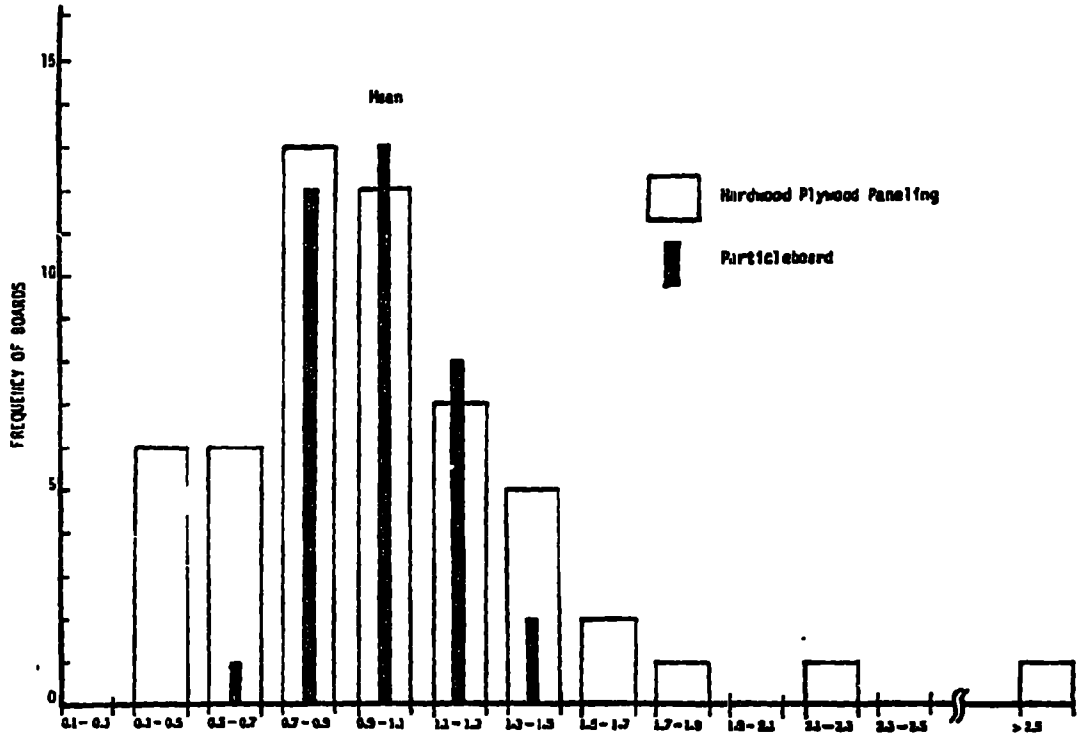
*An approximate 1 to 1 correlation between the results of the 2-hour dessicator test and the FSEM was found in testing at ORNL on nine different products (Matthews et al. 1983).



(Intra-Board Average Emission Rate)/(Inter-Board Average Emission Rate)

Source: Matthews et al. (1983).

Figure 17. Inter-Board Var ation in the CPSC Survey

Table 8. Comparison of 1980 CPSC and 1980, 1982 NPA Test Results

Product	NPA test results (ug/mL)[a] 1980	1982	CPSC test results (ug/mL)[b] 1983
Particleboard underlayment	4.0 ± 2.7	1.7 ± 1.5	0.9 ± 0.6
Industrial particleboard	5.1 ± 4.9	2.0 ± 1.1	0.9 ± 0.4
Mobile home decking	3.6 ± 3.1	1.2 ± 0.5	not tested

[a]2 hour dessicator test results: water sorbent, uncoated edges on test specimens.

[b]FSEM test results: molecular sieve sorbent. Results are selective to the CH_2O emission from the face of the product. An approximate 1 to 1 correlation between the results of the 2 hour dessicator test (units of ug CH_2O/mL H_2O) and the FSEM (ug CH_2O/mL H_2O in the sieve rinse solution) was found in ORNL tests.

3. OTHER RESIDENTIAL SOURCES

Two major sources of formaldehyde emission in a residential setting are pressed wood products and urea-formaldehyde foam insulation (UFFI). However, the formaldehyde concentrations in the home may be attributable to sources other than pressed-wood products containing urea-formaldehyde (UF) resin and UFFI. The other sources can be characterized in the following categories:

- Products with phenol formaldehyde (PF) resins
  - softwood plywood
  - hardboard
  - waferboard
  - oriented strand board
  - fibrous glass insulation
  - fibrous glass ceiling tiles
- Consumer products that may contain formaldehyde resins
  - carpeting
  - upholstery fabric
  - drapery fabric
  - other textiles
- Combustion products
  - unvented kerosene and gas appliances
  - smoke from tobacco products
  - combustion of wood or coal in fireplaces
- Outdoor air
  - ventilation system air exchange

The following sections will address these other residential formaldehyde sources.

3.1 Urea Formaldehyde Foam Insulation (UFFI)

UFFI was injected into the sidewalls of buildings for insulation primarily in the 1970s. Formaldehyde is released from the UF foam in varying concentrations and may arise from (1) excess

formaldehyde in the original UF resin, some of which may be present just after foaming, and (2) a continual generation and release due to hydrolysis of the UFFI (Hawthorne et al. 1981). The acid catalyst used to complete the polymerization of the UFFI generates an acidic environment. This circumstance tends to intensify hydrolytic decomposition and therefore produces a continual release of formaldehyde (Hawthorne et al. 1981.)

Levels of formaldehyde in the indoor environment of both mobile homes and conventional homes insulated with UFFI tend to decrease over time (Cohn et al. 1984). The formaldehyde levels can vary with changes in environmental conditions, such as the temperature and the relative humidity. The age of the UFFI and the air exchange rates of the building are also factors to be considered with the decrease in levels of concentration. According to Anderson et al. (1983), not all UFFI emits significant quantities of formaldehyde vapors. The concentration of formaldehyde is much higher immediately after installation, and with aging of the product less formaldehyde is emitted; the rate of decay of the formaldehyde is not, however, well defined.

Cohn et al. (1984) have gathered data from many studies of UFFI residences within the United States and Canada. They have accumulated 1,164 data points, each of which is the average formaldehyde level measured in a UFFI residence at the time of measurement; also listed is the age of the UFFI (with age defined as time lapsed between installation and measurement). Nearly all of the measurements were performed using the chromotropic acid method. The data were grouped into 10-week intervals by age of the UFFI home, and the average formaldehyde level for all homes falling within each 10-week interval was determined. The average formaldehyde measurement ranged from 0.210 ppm in the first 10-week (0 to 71 days) interval to 0.030 ppm in the last 10-week (3,011 to 3,080 days) interval. The average formaldehyde level showed a tendency to decline rapidly after the first 40 weeks and more slowly thereafter (see Table 9).

Table 9. Average Formaldehyde Measurement in UFFI Homes by Age

Days	Number of data points	Avg ppm HCHO
0-71	63	0.210
141-210	76	0.240
211-280	51	0.240
281-350	58	0.058
351-420	72	0.068
421-490	55	0.084
491-560	68	0.100
561-630	45	0.076
631-700	49	0.078
701-770	70	0.080
771-840	37	0.081
841-910	54	0.079
911-980	45	0.058
981-1050	44	0.082
1051-1120	66	0.072
1121-1190	30	0.050
1191-1260	46	0.040
1261-1330	29	0.072
1331-1400	22	0.054
1401-1470	22	0.074
1471-1540	8	0.063
1541-1610	6	0.047
1611-1680	15	0.032
1681-1750	9	0.021
1751-1820	14	0.067
1821-1890	4	0.102
1891-1960	5	0.039
1961-2030	4	0.080
2031-2100	5	0.054
2101-2170	4	0.050
2171-2240	4	0.078
2241-2310	1	0.040
2521-2590	1	0.117
3011-3080	2	0.030

Source: Cohn et al. (1984)

Limited measurements in other studies indicate that, for homes insulated with UFFI, the range of formaldehyde concentrations is 0.01 to 4.1 ppm with an average concentration of 0.14 ppm (Gupta et al. 1982).

The Consumer Product Safety Commission (CPSC) in 1982 prohibited the installation of UFFI in residential buildings and schools. Although it was later overturned by a Federal court, the CPSC ban of UFFI caused the virtual elimination of the UFFI industry (Formaldehyde Institute 1984). There is considerable debate between the regulatory agencies and the UFFI industry as to the extent of long-term formaldehyde emission from UFFI that is presently in place (Hawthorne et al. 1983).

Detailed information on UFFI can be obtained from the following references:

1. Consumer Product Safety Commission 1982. Urea-formaldehyde foam insulation. Fed. Reg. 16 CFR Part 1306 47(64):14366 (1982).

2. Cohn M. 1981. Revised carcinogenic risk assessment for urea-formaldehyde foam insulation. Washington, DC: Consumer Product Safety Commission.

3. Hawthorne AR, Gammage RB, Matthews TG, et al. 1981. Oak Ridge National Laboratory. An evaluation of formaldehyde emission potential from urea-formaldehyde foam insulation: panel measurements and modeling. Oak Ridge, TN: ORNL/TM-7959.

3.2 Construction Products Containing Phenol-Formaldehyde (PF) Resins

Most structural pressed-wood products used in construction are manufactured using PF resin adhesives instead of UF resins. These products include softwood plywood, waferboard, hardboard, oriented strand board, and structural phenolic particleboard. Other products that contain PF are fibrous glass ceiling tiles and insulation.

1. Structural Wood Panel Products

Structural wood panel products are basically used in construction and exterior applications that require water-proof boards. Common applications include roof and wall sheathing, subflooring, and siding. Small amounts are used for shelving, cabinets, indoor paneling, and fixtures (APA 1984). Although formaldehyde is used in the manufacture of phenolic resins for the phenolic adhesives, is is believed that virtually all the formaldehyde reacts to form PF polymers. The small amount of formaldehyde that is emitted from the panel products is the result of residual formaldehyde from the curing process; no release via resin hydrolysis is expected (APA 1984).

There are several published studies on formaldehyde emissions from PF pressed-wood panel products. The results of these studies, presented in detail in Section 5.3, indicate that PF pressed-wood products are not likely to contribute more than 0.1 ppm to indoor formaldehyde air levels regardless of the product loading or air exchange rate.

2. Fibrous Glass Insulation and Ceiling Tiles

Other generic product lines containing phenol formaldehyde that are used in construction applications are fibrous glass insulation and ceiling tiles. In 1983, as a result of a study on formaldehyde release from consumer products, CPSC recommended further evaluation of fibrous glass insulation and ceiling tiles. In this study, Pickrell et al. (1982) used dessicator tests to measure emission rates of 0.016 and 0.020 mg formaldehyde/m^2/hr for one sample each of fibrous glass insulation and ceiling panels, respectively. The CPSC provided the samples, presumably locally purchased. These products, when compared with other products tested, were among the highest group of emitters. Concern arose from the test results because of the high loading rates of these products in homes. Under normal use conditions (in attics), insulation would be subjected to temperatures much higher than normal room temperatures, thereby increasing potential formaldehyde emissions.

Further investigations on the emissions of formaldehyde from fibrous glass insulation and ceiling tiles were performed by Matthews and Westley (1983) using both the FSEM and a small scale environmental chamber test. FSEM measurements were performed on 9 insulation products and 11 ceiling tile products. Products from a total of 5 manufacturers were tested. Products with the highest FSEM results were tested in the environmental chamber. The samples were conditioned at 24°C, 50% RH and <50 ppb background formaldehyde for one month prior to testing. Tests were performed using 23°C/50% RH. The average formaldehyde emission rate for two insulation products tested in the chamber was 0.017 $mg/m^2/hr$ (range: 0.008 to 0.022 $mg/m^2/hr$) with an air exchange/loading ratio (N/L) of 0.8 to 1.7 m/hr. The average formaldehyde emission rate for three ceiling tiles tested in the chamber was 0.009 $mg/m^2/hr$ (range: 0.008 to 0.011 $mg/m^2/hr$) with an air exchange/loading ratio of 0.28 to 0.55 m/hr. The emission rates for insulation are similar to those of Pickrell et al. (1982); however, emission rates for ceiling tiles are much lower than those rep ·ted by Pickrell et al. (1982).

Matthews and Westley (1983) also modeled the formaldehyde air levels that could result in a room with an air exchange rate of 0.5 hr^{-1} containing a ceiling comprised entirely of ceiling tile overlaid with insulation. The insulation was assumed to be subjected to conditions that would lead to elevated emission rates (increased temperature (38°C) and relative humidity (68 percent)). This work led to a predicted maximum increase of 0.006 ppm in indoor formaldehyde level from use of new ceiling tiles and a maximum increase of 0.022 ppm from new insulation. As the products age, the formaldehyde emission rates and resulting indoor concentrations would be expected to decline significantly.

In a memorandum dated April 16, 1984, from Sandra Eberle of CPSC to the Commission, it is concluded that fibrous glass insulation and ceiling tiles will have little impact on in-home levels of formaldehyde.

3.3 Consumer Products Potentially Containing Formaldehyde Resins

Consumer products found in a residential setting that may contain formaldehyde resins include carpeting, fabric (apparel and non-apparel), and paper products. Pickrell et al. (1982) investigated formaldehyde emissions from 28 different samples of these consumer products using static dessicator tests. The individual sample emission rates are presented in Table 10. The emission rate values, representative of each product group and approximating the median of detectable and relevant values, are presented below. For comparison purposes, the median emission rate values for the pressed-wood products and insulation products tested by Pickrell are also listed.

Product	Emission rate ($ug/m^2/day$)
• Pressed-wood products	~15,000
• Wearing apparel (new, unwashed)	~400
• Insulation products	~400
• Paper products	~300
• Fabric (non-apparel)	~100
• Carpet	~ 15

Most fabrics that contain cotton are finished with a formaldehyde-containing crosslinking agent for durable press properties. Formaldehyde emission rates will, however, decrease with launderings. The first home laundering will greatly diminish the emission level.*

Although carpets are listed as possibly containing formaldehyde resins, an industry representative† states that formaldehyde-emitting resins have never been used by major carpet manufacturers, though small

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*Personal communication between R. Reinhardt, USDA Southern Regional Research Center, and P. Wood, Versar Inc., November 12, 1984.

†Personal communication between Dr. Donald Hayes, Burlington Industries and Gina Dixon, Versar Inc., January 2, 1985.

Table 10. Release of Formaldehyde from Specific Consumer Products[a]

Product	ug/g/day[b]	ug/m^2/day[b] (Range)	(Mean)
Drapery fabric:			
100% cotton, Sample 1	2.8 - 3.0	330 - 350	340
100% cotton, Sample 2	0.8 - 0.9	90 - 120	100
Blend (77% Rayon - 23% Cotton), Sample 1	0.3 - 0.3	50 - 50	50
Blend (77% Rayon - 23% Cotton), Sample 2	ND (0.01)	ND	ND
Upholstery fabric:			
100% Nylon, Sample 1	0.03 - 0.05	7 - 11	9
100% Nylon, Sample 2	0.02 - 0.02	6 - 7	7
100% Olefin, Sample 1	0 - 0.02	0 - 5	3
100% Olefin, Sample 2	ND (0.014)	ND	ND
100% Cotton, Sample 1	ND (0.014)	ND	ND
100% Cotton, Sample 2	ND (0.015)	ND	ND
Latex-backed fabric:			
Sample 1	0.5 - 0.6	90 - 100	100
Sample 2	ND (0.015)	ND	ND
Blend fabric:			
Sample 1	0.3 - 0.4	20 - 30	25
Sample 2	0.2 - 0.3	20 - 30	25
Carpet:			
Foam-backed, Sample 1	0.05 - 0.06	60 - 65	65
Foam-backed, Sample 2	0.006 - 0.01	8 - 13	10
Foam-backed, Sample 3	0 - 0.002	0 - 2	1
Sample 4	0.0005 - 0.0009	0 - 4	2
Sample 5	0.0007 - 0.0009	0 - 1	1
Sample 6	0 - 0.0009	0 - 1	1
Sample 7	ND (0.043)	ND	ND
Clothes (new, unwashed):			
Men's shirts (65% polyester cotton/35% cotton)	2.5 - 2.9	380 - 550	470
Ladies dresses	3.4 - 4.9	380 - 750	570
Girls' dresses (polyester/cotton)	0.9 - 1.1	120 - 140	130
Childrens' clothes (65% polyester cotton/35% cotton)	0.2 - 0.3	15 - 55	35
Paper products:			
Paper plates and cups, Sample 1	0.12 - 0.36	400 - 1000	680
Paper plates and cups, Sample 2	0.03 - 0.14	75 - 450	260
Paper plates and cups, Sample 3	0.10 - 0.15	330 - 335	330

[a]Preconditions = 25°C/100%RH, loading of 2 m^2/m^3.
[b]Range of 2 or more values.
ND = Not detected.
Source: Pickrell et al. (1982).

amounts of formaldehyde were added as a dye stabilizer prior to 1979. It was conjectured that glues used to attach carpet to flooring, or some other component of the flooring system (such as particleboard underlayment) may be responsible for emissions of formaldehyde attributed to carpet. A study conducted by the Ontario Research Foundation for the Canadian Carpet Institute reported that formaldehyde is used as a preservative in latex formulations used for foam backings on carpets (Canadian Carpet Institute 1982). The amount of formaldehyde typically used in commercial latexs was reported to be about 500 ppm. In order to determine if the latex backing could be the cause of reported formaldehyde emissions from carpeting, formaldehyde emission rates were measured for carpet samples prepared with foam latex backing containing 500, 1000 and 10,000 ppm of formaldehyde. Emission rates for carpets with latex backing containing 500 and 1,000 ppm of formaldehyde were less than 14 $ug/m^2/hr$. Emission rates for carpets with latex backing containing 10,000 ppm ranged from 56 to 162 $ug/m^2/hr$. These emission rates are comparable in magnitude to the emission rates measured by Pickrell et al. (1982) and Matthews et al. (1982-1984) for foam-backed carpeting. As shown in Table 9, Pickrell et al. (1982) measured significantly higher emission rates for foam-backed carpets than non-foam-backed carpets. Matthews et al. (1982-1984) measured emission rates of 14 $ug/m^2/hr$ for a urethane foam carpet cushion and 6 $ug/m^2/hr$ for a waffled sponge rubber carpet cushion.

3.4 Combustion

Unvented combustion appliances (such as gas ranges and heaters and kerosene heaters) and tobacco smoking emit formaldehyde as the result of incomplete combustion (Girman et al. 1983). Several controlled chamber studies have been conducted by Lawrence Berkeley Laboratory (LBL) to determine formaldehyde emission rates from gas- and kerosene-fueled

combustion appliances (Traynor et al. 1982, Girman et al. 1983, Traynor et al. 1983). Caceres et al. (1983) and Fortmann et al. (1984) have determined emission rates by sampling appliance exhaust gases. The results of these studies are summarized in Table 11. More recent studies have been conducted at LBL but the results are not yet available.*

The results listed in Table 11 indicate that emission rates vary considerably between different appliance types and are dependent, to a large degree, on whether the appliance is tuned and functioning properly. Gas stoves, water heaters and furnaces are the most common combustion appliances in general use. Formaldehyde was not detected in fugitive emissions from water heaters or furnaces. Although the emission rates for the oven and top burners of gas stoves can be relatively high, the intermittent use of gas stoves precludes them from typically being significant sources of formaldehyde emission in the home.

Gas-fueled space heaters, on the other hand, are likely to be used for longer durations of time and can have relatively high emission rates, particularly if not well-tuned. Caceres et al. (1983) measured a formaldehyde concentration of 0.24 ppm in a small room (21 m^3 with an air exchange rate of 0.5 hr^{-1}) containing a gas-fueled heater working at full strength and Girman et al. (1983) reported a concentration in excess of 1 ppm in a controlled field study with a poorly-tuned heater. The emission rates for kerosene-fueled space heaters are generally much lower than those from gas-fueled heaters.

Traynor and Nitschke (1984) surveyed indoor air levels of formaldehyde in 30 homes, stratified by presence o suspected emission sources. Results indicate no perceptible effect on indoor levels by combustion:

- Three homes with kerosene space heaters averaged 0.029 ppm.
- Three with wood-burning stoves averaged 0.026 ppm.

*Personal communication between J.R. Girman, Berkley Laboratories and P. Wood, Versar Inc. on November 15, 1984.

Table 11. Summary of Formaldehyde Emission Rates from Unvented Combustion Appliances

Appliance	No. of appliances tested	Emission factors (ug/kj) Average	Range	Fuel consumption (kj/h)	Emission rates (mg/hr) Average	Range
Gas stove (age unspecified)[a]	1					
Oven		2.7	2.4-?.4	8,400	22.7	20.1-28.6
Top burner		1.7	0.?o-2.5	9,200	15.6	7.9-23.0
Older gas stove, cast iron[c] burners	1	0.72[f]	NR	9,500[f]	6.8	-
Older oven[c]	1	0.19[f]	NR	6,030[f]	1.2	-
New gas stove, steel[c] burners	1	0.84[f]	NR	3,400[f]	2.9	-
New oven[c]	1	1.5[f]	NR	9,300[f]	14.0	-
Gas space heater						
Well tuned[b]	8	0.81	0.43-4.2	10,100-44,700	8.2-35.8	-
Poorly tuned[b]	2	1.5	0.46-20.3	33,600-43,900	50.4-65.8	-
Well tuned[e]	4	-	-	-	7.0	4.4-10.4
Kerosene space heater						
New convective[d]	1	0.17	0.01-0.42	4,230-7,980	1.0	0.08-1.8
New radiant[d]	2	0.52	0.10-0.80	6,640-8,250	4.0	0.66-5.7
Radiant[e]	2	-	-	-	1.0	0.9-1.0
Wick[e]	1	-	-	-	0.4	-
Gas furnace[c]	1	<0.003[f]	NR	137,000[f]	<0.4	-
Gas water heater[c]	1	<0.005[f]	NR	45,500[f]	<0.3	-

Source: [a]Traynor et al. (1982).
[b]Girman et al. (1983).
[c]Fortmann et al.(1984).
[d]Traynor et al. (1983)
[e]Caceres et al. (1983)
[f]Values originally reported in units of Kcal were converted to kj (1 Kcal=4.187 Kj).

NR = not reported

- One with a coal-burning stove averaged 0.028 ppm, and one with a coal-burning fireplace averaged 0.019 ppm.

- Two with gas-fired ranges averaged 0.046 ppm, and three with gas furnaces averaged 0.030 ppm.

- The four homes with no identified formaldehyde source averaged .036 ppm (range of 0.007 to 0.077).

The University of Texas (1983), in their indoor air quality study, evaluated the effect of emissions from a propane stove on formaldehyde levels in mobile homes with relatively high formaldehyde levels. The appliance was operated both with and without an exhaust fan. They found that levels did not change during and after stove use (mean levels remained at 0.31 ppm) and concluded that the propane stove was not a formaldehyde source in the two homes studied.

Leaderer et al. (1984) report similar results in their study of 55 homes in Connecticut. This study, conducted during the winter, found no significant difference in the formaldehyde levels measured in homes with gas stoves and/or kerosene heaters and in those homes without these combustion sources. The authors concluded that the low formaldehyde levels measured (average of 0.022 $\pm$ 0.014 ppm) were not associated with indoor combustion of fuels.

Another possible source of formaldehyde emission in the indoor environment is wood combustion in fireplaces and wood stoves. Lipari et al. (1984) measured aldehyde emissions from wood-burning fireplaces. Four different types of wood were tested, including jack pine, cedar, red oak, and ash. Formaldehyde was one of the major aldehydes emitted. Sampling was conducted using a freestanding fireplace installed in the laboratory. Samples were collected from the chimney port (not from the ambient air of the laboratory) using impingers containing 2,4-dinitrophenylhydrazine (DNPH) in acetonitrile. The aldehydes were analyzed by high performance liquid chromatography. Formaldehyde emissions ranged from 21 to 42 percent of the total aldehyde emission for

each wood type tested with one exception. For the red oak, formaldehyde emissions were 89 percent of the total aldehyde emission. The emissions reported ranged from 0.089 to 0.708 g/kg (grams of formaldehyde per kg of wood). There may be a relationship between wood type and formaldehyde emission; however, scarcity of data prevents any conclusions (Lipari et al. 1984).

The literature reviewed presented disparate estimates of formaldehyde emissions from burning cigarettes; published emission rates range from 20 to 1440 ug/cigarette.

Matthews et al. (1984), citing laboratory studies published by others, derived an emission rate of 1.2 mg/cigarette. They based their value on emission rates of 0.97 and 1.44 mg/hr published by other investigators. The value of 1.44 mg is described as a rate per unfiltered cigarette.

Egle and Hudgins (1974) measured 4.1 ug formaldehyde per 40 ml puff; assuming 30 puffs per cigarette, an emission rate of 1.2 mg/cigarette can be calculated. This independently-derived value agrees perfectly with Matthew's value of 1.2 mg/cigarette.

Data presented by Timm and Smith (1979) allow calculation of a formaldehyde emission rate of 0.74 mg/cigarette. This value is calculated from the measured level of 0.26 ppm in a 45.8 m^3 room in which 20 cigarettes had been smoked. It was assumed that, during the half-hour experiment, ventilation did not remove any formaldehyde.

Rickert et al. (1980) measured total aldehydes in sidestream smoke produced by a smoking machine, and calculated an emission rate of 0.912 mg total aldehydes per cigarette. The proportion of formaldehyde to total aldehydes is not known.

Bardana (1984) cites the Surgeon General's 1972 report on smoking in his derivation of a 0.57 mg/cigarette formaldehyde emission rate. The Surgeon General's 1972 and 1984 reports, however, list emission rates of

0.02 to 0.04 and 0.02 to 0.09 mg formaldehyde per cigarette; Bardana apparently cites a measured formaldehyde level in a room with cigarette smoke and other sources to back-calculate the 0.57 mg emission factor.

Pickrell et al. (1982) reported that cigarette smoke contains up to 40 ppm formaldehyde; Ayer and Yeager (1982) measured 90 to 110 ppm in sidestream smoke (a much more significant source of indoor formaldehyde than mainstream smoke). These levels are expected to diminish rapidly with distance from the source. Traynor and Nitschke (1984) measured levels in homes with smokers. Even with low air exchange rates (less than 0.2 ACH), formaldehyde levels in these homes did not exceed 0.06 ppm and were not significantly different from levels in homes with no formaldehyde source.

3.5 Outdoor Air

The levels of formaldehyde in outdoor air are generally lower than indoor levels. Thus, as the air exchange rate increases, there is a decrease in the level of formaldehyde indoors. However, in cases where formaldehyde levels outdoor are higher than levels indoors, the indoor air can potentially be further polluted with ventilation air exchanges. In outdoor air, formaldehyde can originate from industrial plants and from many combustion sources such as engine exhaust and incinerators.

Singh et al. (1982c) used the chromotropic acid method and the DNPH method to analyze for formaldehyde concentrations in six urban areas of the United States in 1980 and 1981. The reported concentrations are presented in Table 12. Altschuller (1983) has summarized other ambient air measurements for formaldehyde in urban areas in the United States. The average concentrations range from 7.0 ppb to 70 ppb. These data are also presented in Table 12. Measurements of formaldehyde in a nonurban site were in the range of 0.1 to 0.8 ppb (Altschuller 1983).

Table 12. Ambient Air Measurements of Formaldehyde at Urban Sites in the United States

Location	Time of Year	Time of Day	Concentrations (ppb) Average	Maximum
Downtown Los Angeles, CA	July, Nov. 1960	9 a.m.	45	130
S. Pasadena, CA	July, Nov. 1960	1 p.m.	30	70
Downtown Los Angeles, CA	Sept., Nov. 1961	7 a.m.	40	160
Huntington Park, CA	Oct. 1968	7:40 a.m.	70	135
El Monte, CA	Oct. 1968	5:45 a.m.	50	90
Los Angeles, CA (Cal. State Univ.)	June 1980	morning-evening	21, 20	35, 39
Los Angeles, CA (Cal. State Univ.)	June 1980	morning-eveing	44	71
Claremont, CA (Harvey Mudd College)	Oct. 1978	late morning-late evening	28	71
Claremont, CA (Harvey Mudd College)	Aug., Sept. 1979	morning-late evening	10	22
Claremont, CA (Harvey Mudd College)	Sept., Oct. 1980	morning-evening	24	48
Riverside, CA (U. CA. Riverside)	Oct. 1976	late morning-early evening	8	14
Riverside, CA (U. CA. Riverside)	June, Aug. Oct. 1977	late morning-evening	19	38
Houston, TX (Crawford)	Sept., Oct. 1978	morning-early evening	15	25
Houston, TX (Clinton, Dr.)	Sept., Oct. 1978	morning-noon	8	28

Table 12. (continued)

Location	Time of Year	Time of Day	Concentrations (ppb) Average	Maximum
Houston, TX (Parkhurst)	Sept., Oct. 1978	morning-early evening	7	15
Houston, TX (Fuqua)	Sept., Oct. 1978	morning-early evening	11	27
Columbus, OH (Fort Hayes)	Sept., Oct. 1980	early morning-early afternoon	8	23
Columbus, OH (WCVO)	Sept., Oct. 1980	early morning	10	12
Atlanta, GA (GA Tech)	July, Aug. 1981	6 a.m.	8	22

Source: Altshuller (1983)

Denver, CO	June 1980	-	12.3	28.7
St. Louis, MO	May 1980	-	11.3	18.7
Chicago, IL	April 1981	-	12.8	17.2
Pittsburgh, PA	April 1981	-	20.6	35.1
Staten Is., NY	April 1981	-	14.3	45.9
Riverside, CA	July 1980	-	19.0	41.0

Source: Singh et al. (1982c)

Monitoring studies of indoor residential levels of formaldehyde have frequently reported the results of formaldehyde measurements performed outside the residence. The results of several of these studies are summarized below.

Study	No. of Residential Sites	Mean Concentration (ppb)
Canada (UFFI/ICC 1981)	≤ 2,275	0.008
U.S. Nationwide (Singh et al. 1982a)	≤ 260	<0.02
United Kingdom (Everett 1983)	60	0.006
Indiana (Konopinski 1983)	47	0.005
Tennessee (Hawthorne et al. 1984)	40	<0.025
Iowa (Schutte et al. 1981)	27	0.002
Texas (University of Texas 1983)	≤ 164	<0.02

It has been suggested that the atmospheric levels of formaldehyde vary with seasonal influence. Tanner and Meng (1984) observed strong seasonal variations in the levels of formaldehyde. The maximum levels were observed in the summer. The formaldehyde samples were collected, at an unidentified northeast U.S. coastal site, using an impinger containing acetonitrile and DNPH; they were analyzed by high-pressure liquid chromatography. The concentrations ranged from 0.9 to 48 ppb with an overall mean of 7.5 ppb. The monthly averages of ambient levels were as follows:

Month	Concentration (ppb)
July - August 1982	15.8
October - November 1982	4.4
March 1983	3.8
April 1983	11.2
May 1983	12.2

Formaldehyde is known to be produced in the atmosphere from photolysis of hydrocarbons and ozone (NAS 1981). The increased intensity of sunlight in warmer months may account for much of this observed seasonal variation.

3.6 Relative Significance of Sources on Air Levels Indoors

In order to compare the relative impact of residential formaldehyde sources on indoor formaldehyde air concentrations, a simple steady-state indoor pollutant concentration model developed by Matthews et al. (1983) can be used. This model incorporates source emission algorithms for combustion sources and formaldehyde resin-containing products. The algorithms for resin-containing wood products assume a negative linear dependence of emission rate on background concentration of formaldehyde in air. The algorithms for combustion sources and resin-containing products with low emission rates (e.g., textiles) assume constant emission rates unaffected by background formaldehyde concentration. The model does not account for the effects of product aging and formaldehyde sinks and assumes constant environmental conditions of 23°C and 50 percent RH.

This model is a simplified version of a more complex model being developed by Matthews that incorporates algorithms to predict emission rates, absorption by formaldehyde sinks, the effects of numerous sources, the decay of emissions over time, and the effects of varying environmental conditions to describe dynamic formaldehyde levels in homes. This more complex model, as well as the algorithms for resin-containing wood product emissions used in the steady-state model, are currently undergoing validation testing at the National Bureau of Standards. Both models are discussed in more detail in Section 7 of this report.

Matthews' steady-state model has been used to predict the potential impact of individual emission sources on indoor formaldehyde levels in a dwelling with an interior volume of 175 m^3. This size corresponds to the the size of a single-wide mobile home or a small modular home or apartment. Pressed-wood products were assumed to be present at the loadings listed in Table 1 of this report for mobile homes and newly

constructed single family conventional homes. An additional 3 m^2 of industrial particleboard and 2 m^2 of MDF are assumed to be present in furniture. The loadings for other sources are based on assumed high usage rates. For example, carpeting and ceiling tile were assumed to cover the entire floor and ceilings, respectively. The air exchange rates in the 175 m^3 model mobile and conventional homes were assumed to be 0.35 and 0.50 air changes per hour, respectively. The emission rate data used for wood products and UFFI are based solely on average emission rate data presented in Matthews et al. (1983). The emission rate data for other sources are based on information presented previously in this section.

Tables 13 and 14 present the assumed source loadings and emission rates used and the modeled changes in the indoor formaldehyde concentration from an assumed background level of 0.024 ppm. It was conservatively assumed that the emission rate from each source was independent of emissions from other sources. The results indicate that, for both model homes under the assumed loading rates, UF resin-containing wood products and UFFI are the major potential contributors to indoor air levels. Gas space heaters could also cause elevated levels if used for long periods of time. A simplified ranking of the most significant sources can be derived by setting the most important source in each home equal to 1.0, and scaling the other appropriately:

Mobile home

Hardwood plywood paneling (avg. of 3 types)	= 1.0
Particleboard underlayment (carpet covered)	= 0.55
Industrial particleboard	= 0.22
Gas space heater	= 0.19
MDF	= 0.17
Other combustion sources (combined)	= 0.12
All other individual sources	= <0.10

Table 13. Potential Impact of PF Resin-Containing Products, Consumer Products, and Combustion on Indoor Formaldehyde Concentrations[a]

Product	Measured emission rates[b]	Assumed emission rate[b]	Assumed usage[c]	Modeled change in 24-Hr average CH_2O concentration	
				Mobile Home	Conventional Home
Construction products with PF resins	(units of mg/m^2 hr)	(units of mg/m^2 hr)	(units of m^2)		
PF plywood flooring (uncovered)	0.01 - 0.02	0.02	82	0.021	0.015
Fibrous glass insulation	0.008 - 0.022	0.017	82	0.018	0.013
Fibrous glass ceiling tiles	0.008 - 0.011	0 009	82	0.010	0 007
Consumer products with CH_2O resins	(units of ug/m^2 day)	(units of ug/m^2 day)	(units of m^2)		
Carpeting	ND - 65	13	82	<0.001	<0.001
Upholstery fabric	ND - 11	6	15	<0.001	<0.001
Drapery Fabric	ND - 350	170	15	0.001	<0.001
Apparel (unwashed)	15 - 550	300	5	0.001	<0.001
Combustion sources (unvented)	(units of mg/hr)	(units of mg/hr)	(units of hr/day)		
Gas stove burners	2.9 - 16	8.6	1.0	0.005	0.003
Gas oven	1.2 - 23	13	0.7	0.005	0.003
Kerosene space heater					
Convective (new)	0.08 - 1.8	1.0	8	0.004	0.003
Radiant (new)	0.66 - 4.0	4.0	8	0.017	0.012
Gas space heaters (well-tuned)	4.4 - 36	10	8	0.044	0.031
Cigarettes	0.02 - 1.44 mg/cig	1.2 mg/cig	10 cig/day	0.007	0.005

[a]Potential impacts estimated using Matthews et al. (1983) Simple-Steady State Model (see Sections 3.6 and 7.2 for more details).

[b]Emission rate data for PF resin products were obtained from Matthews et al. (1983), Matthews and Westley (1983). Emission rate data for consumer products and combustion sources were obtained from Tables 10 and 11, respectively. The assumed emission rates for consumer products were obtained by averaging the mean emission rates for those products with detected emission rates.

[c]See Section 3.6 for details.

[d]Air exchange rate assumed to be 0.35 hr^{-1} for a 175 m^3 mobile home and 0.5 hr^{-1} for a 175 m^3 conventional home.

Table 14. Potential Impact of UF Resin-Containing Wood Products and Insulation on Indoor Formaldehyde Concentrations[a]

Product	Measured emission rates[b] (mg/m^2 hr)	Assumed emission rate[b] (mg/m^2 hr)	Assumed usage[c], m^2		Modeled change in 24-Hr average CH_2O concentration (ppm)	
			Mobile Home	Conv. Home	Mobile Home	Conv. Home
Furniture, cabinetry, and shelving						
Industrial particleboard	0.15 - 0.62	0.31	12	12	0.051	0.037
MDF	0.57 - 2.3	1.5	2	2	0.040	0.028
Hardwood plywood paneling						
Ink print overlay	0.05 - 0.63	0.28	175	12	0.340	0.033
Paper overlay	0.03 - 0.27	0.11	175	12	0.188	0.014
Domestic veneer overlay	0.07 - 0.24	0.12	175	12	0.183	0.015
Particleboard underlayment						
No cover	0.11 - 0.78	0.30	82	21	0.222	0.061
Carpet and cushion cover	---	---	82	21	0.130	0.027
Tile cover	---	---	82	21	0.002	<0.001
UFFI (in one exterior wall)	0.05 - 0.80	0.23	0	20	---	0.054

[a]Potential impacts estimated using Matthews et al. (1983) Simple Steady-State Model (see Sections 3.6 and 7.2 for more details).

[b]Emission rate data as reported in Matthews et al. (1983) for products manufactured during 1983.

[c]See Section 3.6 for details.

[d]Air exchange rates assumed to be 0.35 hr^{-1} for a 175 m^3 mobile home and 0.5 hr^{-1} for a 175 m^3 conventional home.

Conventional home (excluding UFFI)

Industrial particleboard	= 1.0
Gas space heater	= 0.84
MDF	= 0.76
Particleboard underlayment (carpet covered)	= 0.73
Hardwood plywood paneling (avg. of 3 types)	= 0.56
Other combustion sources (combined)	= 0.56
All other individual sources	= <0.40

4.0 RESIDENTIAL MONITORING DATA

The purpose of this section is to summarize available formaldehyde indoor air monitoring data for domestic and foreign residences. Each data set is accompanied by an overview of the study or project from which it resulted. The format of each summary will include, when available, the following components: study name, applicable literature references, monitoring dates, survey design (including types of sampling and analysis), and the results. Summaries are appropriately located in one of the sections immediately following.

4.1 Major Studies of Residential Levels

Lawrence Berkeley Laboratory Study

Lawrence Berkeley Laboratory (LBL) has summarized formaldehyde emission rates from a variety of combustion appliances. LBL Indoor Air Quality Group has also collected data on the formaldehyde concentrations observed in 40 residential indoor environments in various studies since 1979 (Girman et al. 1983). The combined data set (see Table 15) has been tabulated from the results presented in several papers published in the past few years. The data were obtained through the use of refrigerated pump/bubbler samplers and a modified pararosaniline analytical method (Girman et al. 1983).

The data presented in Table 15 indicate that new energy-efficient houses were generally found to have higher concentrations than those observed in weatherized houses, with about a third being above the American Society of Heating, Refrigeration, and Air Conditioning Engineers (ASHRAE) guideline, 100 ppb. The effect of ventilation rate on

Table 15. Summary of Formaldehyde Concentrations in Indoor Environments Studied by the Lawrence Berkeley Laboratory

Location	Type house	No. buildings	Formaldehyde range (ppm)	Formaldehyde average (ppm)[a]
Ames, IA	Energy-efficient	1	0.028-0.061	NA
Carroll Co., MO	Energy-efficient	1	0.044-0.148	0.098
Mission Viejo, CA	Energy-efficient	1	0.066-0.214	NA
Medford, OR	Conventional, retrofitted for energy-efficiency	2 (2)[b]	0.051-0.068	NA
Midway, WA	Conventional, retrofitted for energy-efficiency	12	<0.005-0.079	NA
Northfield, MN	Energy-efficient, heat exchanger	1 (1)[b]	0.069-0.073	0.070
Dundas, MN	Energy-efficient, mechanical ventilation	1 (1)[b]	0.064-0.080	0.072
Rio, WI	Conventional, retrofitted for energy-efficiency	1	0.053	0.053
Cranbury, NJ	>100 yrs, retrofitted for energy-efficiency	1	0.019-0.022	0.021
Eugene, OR	Energy-efficient	2	0.037-0.073	NA
	Energy-efficent, passive solar	2	0.082-0.112	NA
Rochester, NY	Energy-efficient, mechanical ventilation	10 (6)[b]	<0.005-0.064	0.029
Sacramento, CA	Energy-efficient, passive solar	5	0.098-0.127	NA

[a]NA indicates that neither an average concentration nor a set of data to calculate an average was reported in the literature.

[b]Indicates homes in which the effects of ventilation were studied.

Sources: Girman et al. (1983), Hollowell et al. (1982), Offerman et al. (1982).

formaldehyde levels was examined in ten houses. Variation in the ventilation rate was shown to have a predictable effect on formaldehyde concentrations in seven of the houses studied, but had effects opposite to those predicted in the other three houses.

Geomet Study

As part of the development of an indoor air pollution model based on outdoor pollution and air exchange rates, Geomet, Inc. studied the patterns of indoor aldehyde levels monitored in 17 houses and 2 mobile homes in the U.S. These data can be useful if we assume formaldehyde constitutes 60 percent of total aldehydes, based on LBL data.

In each of three indoor locations, three 4-hour averages were measured on each of 14 days. Outdoor concentrations were also observed over a 24-hour period at one location per home.

The results in Moschandreas et. al. (1978) concluded that the 17 houses had an average aldehyde concentration of 0.09 ppm, and the average for the two mobile homes was 0.35 ppm. If we use the 60 percent factor, the average formaldehyde concentration for the houses would be 0.05 ppm, with 0.21 ppm for the mobile homes. The observed outdoor concentrations of aldehydes were consistently lower than the indoor levels, typically by a factor of 6 and quite often by one order of magnitude. The results are summarized in detail in Table 16.

University of Washington Study

The Department of Health, University of Washington (Breysse 1984), along with a number of commercial laboratories, has monitored formaldehyde in more than 1,000 conventional and mobile homes. For the most part, sampling and surveying of these homes was initiated by residents' complaints and/or formaldehyde exposure symptoms.

Table 16. Summary of Observed Aldehyde Concentrations in U.S. Homes Monitored by Geomet, Inc.

Residence	Concentration (ppm)	
	Average	Range
Denver conventioanl	0.20	0.07-0.50
Chicago Experimental I	0.16	0.11-0.24
Chicago Experimental II	0.26	0.20-0.45
Washington Conventional I	0.04	0.02-0.12
Baltimore Conventional II	0.06	0.03-0.12
Washington Experimental I	0.07	0.01-0.23
Baltimore Experimental I	0.06	0.01-0.13
Baltimore Experimental II	0.04	>0.01-0.10
Pittsburgh Low Rise I	0.07	0.04-0.12
Pittsburgh High Rise I	0.05	0.02-0.10
Chicago Conventional I	0.04	0.01-0.13
Chicago Conventional II	0.04	0.02-0.15
Pittsburgh Low Rise II	0.06	0.03-0.12
Baltimore Conventional I	0.12	0.01-0.24
Pittsburgh High Rie II	0.10	0.06-0.19
Pittsburgh High Rise III	0.12	0.05-0.19
Pittsburgh Low Rise II	0.09	0.02-0.08
Pittsburgh Mobile Home I	0.38	0.16-0.76
Pittsburgh Mobile Home II	0.31	0.11-0.75

Range in 4-hour concentrations taken 3 times/day over 14-day period.

Source: Moschandreas et al. (1978).

After various methods of monitoring were reviewed, it was decided to utilize the chromotropic acid method using one impinger instead of two. No corrections were made for the use of only one impinger. Temperature and humidity were also monitored. Whenever possible, home owners were requested to close all windows and doors and keep the temperature at 70 to 72°F the night before the survey was scheduled.

University of Washington sampled 244 homes insulated with UFFI, 430 mobile homes, and 59 conventional homes or apartments. Table 17 presents the number of samples in each of the formaldehyde concentration ranges found during the study. Overall, average concentrations of formaldehyde in mobile homes were 2 to 10 times higher than concentrations in conventional homes with UFFI.

In early 1983, three private Washington laboratories reported formaldehyde monitoring results for 380 homes (see Table 18). Approximately 52 percent of the samples exceeded 0.05 ppm with a maximum of 5.3 ppm noted in a mobile home.

MHI Mobile Home Study

In 1984, the Manufactured Housing Institute (MHI) had Conner Homes, Inc. construct a single-wide demonstration mobile home unit for the purposes of monitoring indoor ambient formaldehyde levels (primarily to see whether levels conformed to the new HUD target ambient formaldehyde level of approximately 0.4 ppm).

The demonstration home was constructed in a fashion not dissimilar from normally produced Conner mobile homes. This included the use of particleboard and hardwood plywood, and interior features such as cabinet doors comprised of medium density fiberboard. Specific loadings were not available for the individual board types but the researchers did caution that they were unsure as to whether the home included formaldehyde emitting products in a manner generally representative of the industry.

Table 17. Number of Samples in Formaldehyde Concentration Ranges Found by University of Washington

Formaldehyde conc. (ppm)	Mobile homes (430)	UFFI (244)	Homes & apts. (59)
≥ 1.0	37	15	2
≥0.5-0.99	147	10	2
≥0.1-0.49	522	125	41
<0.1	116	370	68
Total	822	520	113

Source: Breysse (1984).

Table 18. Number of Samples in Formaldehyde Concentration Ranges Found by Private Washington Laboratories

Lab I.D.	No. homes	Concentration ranges (ppm) >1.0	0.99-0.50	0.49-0.1	0.09-0.05	<0.05	Total
I[a]	215	2	2	34	112	228	378
II[a]	121	1	3	39	94	106	242
III-Mobile	19	4	3	31	1	4	43
-Conven.	25		2	9	45	20	76
Total	380	7	9	113	252	358	739

[a]Home Type (conventional or mobile) was not reported in the literature.

Source: Breysse (1984).

Tests were conducted approximately three months after the construction of the demonstration home. Readings were taken on five separate days, all within a one-month period of time. Sampling stations were located at 50-inch elevations, drawing air through 20 ml of 1 percent aqueous sodium bisulfite solution at a rate of 1 liter/minute. The chromotropic acid method was used for sample analysis.

The results for each of the five testing days are summarized in Table 19 (Conners 1984). The overall average concentration observed was 0.34 ppm. Test details and environmental conditions on each corresponding test day are also presented in Table 19.

Canadian National Testing Survey

The Urea Formaldehyde Foam Insulation Information and Coordination Centre (UFFI/ICC 1981) was established by the Canadian government in 1981 to handle all UFFI-related matters for the government. One of the objectives of the Centre was to carry out a national testing survey that would involve monitoring nearly 2,300 Canadian homes.

Four different categories of homes were established from which a total of 2,275 houses were selected: the first 100 represented houses where individuals had reported serious health problems or where residents were forced to move from their residences; from the Canadian houses insulated under the CHIP (Canadian Home Insulation Program) program, 1,146 houses with UFFI and 378 houses without UFFI were selected; and another 651 houses containing UFFI were selected from UFFI/ICC files and provincial records, apparently at random.

The sampling and analysis involved the NIOSH method (Lawrence Berkeley Laboratory modified) using a sorbent tube containing a molecular sieve. Air was collected at a rate of 2 l/min. for 15 minutes. For each home, a minimum of two room air samples and one outdoor ambient air sample was collected. The solid sorbent tubes were analyzed by the modified pararosaniline method. QA/QC was reportedly good.

Table 19. MHI Mobile Home Study Test Results and Test Details

	Date of Measurement (1984)				
	2/15	2/16	2/27	3/1	3/2
Formaldehyde conc. (ppm)					
Kitchen	0.41	0.41	0.26	0.28	0.31
First bedroom	0.43	0.43	0.24	0.26	0.30
End bedroom	0.46	0.45	0.28	0.30	0.32
Avg. conc. observed (ppm)	0.43	0.43	0.26	0.28	0.31
Average inside temp. (°F)	78	77	74	76	78
Average conc. (ppm) adjusted to 77°F and 50% RH*	0.45	0.39	0.35	0.34	0.36
Time test initiated	11:20am	10:26am	11:31am	9:55am	9:40am
Sampling duration (min.)	45	45	45	45	45
Inside RH	45%	55%	43%	42%	39%
Outside temp. (°F)	50	55	40	36	43
Outside conditions	rain	100% overcast	100% overcast	Sunny	Partly cloudy
Outside HCHO (ppm)	0.01	<0.01	0.03	<0.01	<0.01

Source: Conners (1984).

*Concentrations adjusted using equations presented in Section 2.3.2 and equation coefficients reported by Myers (1984b).

The results of the averaged concentrations found in each of the four categories are presented in Table 20. Results from three of the categories, the non-complaint UFFI and the non-UFFI homes, are broken out in more detail in Table 21.

Clayton Study

In 1980, Clayton Environmental Consultants, Inc. was contracted by the U.S. Department of Housing and Urban Development (HUD) to conduct several research projects related to formaldehyde contamination in mobile home indoor air (Singh et al. 1982a). Both occupied and unoccupied mobile homes were selected for this study (approximately 260), and all were voluntarily enrolled (i.e., they were primarily "non-complaint" homes). The testing involved homes in Florida, Georgia, Texas, California, Indiana, Michigan, and Minnesota, covering a spectrum of climatic conditions.

The testing took place between September 1980 and October 1981. Three measurements were typically taken in each single-wide home (usually in the kitchen, living room, and master bedroom). Four measurements were made in each double-wide home (usually the kitchen, living room, master bedroom, and second bedroom or den). Outdoor formaldehyde concentrations were measured in each area to account for the formaldehyde present in the background ambient air. No results were presented for outside levels since all values were less than the detection limit of the analytical method used. A variety of factors that affect the indoor levels of formaldehyde were also measured at each sampling event, including age of the mobile home, temperature, relative humidity, and occupancy.

The test procedure used was the pararosaniline method. Concentrations of formaldehyde were determined from standard curves prepared daily. QA/QC was good in that either complete analysis (including spectrophotometric evaluation) was performed on site or good sample preservation techniques were employed to ensure the integrity of all transported samples.

Table 20. Summary of Results of Canadian National Testing Survey

Sample[a] categories	Number of houses	Formaldehyde results: Using house average indoor readings — Average[b] (ppm)	Using house average indoor readings — % at or over 0.1 ppm	Using house maximum indoor readings — Average (ppm)	Using house maximum indoor readings — % at or over 0.1 ppm	Average outdoor readings (ppm)
First one hundred (complaint homes)	100	0.139 (0.139)	47%	0.174	57%	0.007
UFFI/ICC Files	651	0.040 (0.041)	5.1%	0.048	8.6%	0.008
UFFI CHIP	1,146	0.054 (0.054)	10.2%	0.067	16.5%	0.009
Control CHIP	378	0.034 (0.035)	2.6%	0.042	4.8%	0.007

[a]See accompanying text for details on sample categories.

[b]Values not in parentheses were calculated assuming "not detected" results are equal to zero. Values in parentheses were calculated assuming "not detected" results are equal to the detection limit (0.01 ppm).

Source: UFFI/ICC (1981).

Table 21. Comparison of Canadian Home Populations by Average Formaldehyde Concentration

Average formaldehyde concentration (ppm)	Control CHIP (non-UFFI) 378 homes		UFFI/ICC Files (UFFI-non complaint) 651 homes		UFFI/CHIP files 1,146 homes	
	Percentage	Cumulative percentage	Percentage	Cumulative percentage	Percentage	Cumulative percentage
<.01	12.7	12.7	11.1	11.1	4.2	4.2
.01-.025	29.4	42.1	26.3	37.4	16.0	20.2
.025-.040	25.7	67.8	24.6	62.0	22.2	42.4
.040-.055	17.7	85.5	14.3	76.3	20.0	62.4
.055-.070	7.9	93.4	9.8	86.1	13.9	76.3
.070-.085	4 0	97.4	4.9	91.0	8.7	85.0
.085-.10	—	—	4.0	95.0	5.2	90.2
.1-.15	2.4	99.8	3.8	98.8	7.6	97.8
.15-.20	0.3	100.1	1.1	99.9	1.4	99.2
>.2	—	—	0.2	100.1	0.9	100.1

Source: UFFI/ICC (1981).

For the 259 observations, formaldehyde levels were adjusted to standard conditions (25°C and 50 percent relative humidity) using Berge's formula. The adjusted levels ranged between 0.02 and 2.9 ppm (see Table 22 for more details), with a mean of 0.62 ppm (std. dev. = 0.58) and a median of 0.38 ppm. A statistical analysis done by Versar correlating the formaldehyde levels with the mobile home ages is presented in Section 7 of this report.

Wisconsin Study

In March of 1980, the Wisconsin Division of Health, Madison, Wisconsin, initiated an indoor air quality study to characterize indoor formaldehyde concentration variations in mobile homes in terms of the effects of home age, temperature, and humidity (Anderson et al. 1983). The project design utilized a stratified random sampling procedure to identify and voluntarily enroll 137 homes in the study. The enrolled homes were sampled once a month for six or more consecutive months followed by a final sample at the one year anniversary.

Originally, 976 data points were collected from 137 mobile homes voluntarily enrolled in the study. Upon review of the data supplied by Wisconsin, 56 points were found missing (53 concentrations and 3 home ages), leaving 920 full observations. Each remaining data point consisted of a formaldehyde concentration value in ppm (adjusted to 25°C and 50 percent humidity via Berge's formula) and the corresponding age of the mobile home monitored. All the mobile homes in the study were categorized as "non-complaint" homes.

Formaldehyde samples were collected in two rooms (usually the kitchen or living room and bedroom) using personal sampling pumps (MSA model G and Bendix BDX 44). Air was drawn through midget impingers containing 15 to 20 mls of one percent sodium bisulfite absorbing reagent. Pumps were run at a flow rate of 0.7 l/minute for approximately one hour. Gas

Table 22. Number of Observations Found in Concentration Intervals by Clayton Environmental Consultants

Concentration interval (ppm)	Number of observations
0.0 - .10	21
.11 - .20	51
.21 - .30	37
.31 - .40	24
.41 - .50	13
.51 - .60	12
.61 - .70	12
.71 - .80	10
.81 - .90	10
.91 - 1.00	20
1.1 - 2.00	38
2.1 - 3.00	11
Total	259

Source: Versar statistical analysis of data supplied by Singh et al. (1982).

appliances were shut off, and smoking was discouraged during the sampling period. Windows were closed approximately one-half hour prior to sampling. Samples were analyzed at the Wisconsin State Laboratory of Hygiene using the NIOSH chromotropic acid procedure (Anderson et al. 1983). QA/QC was reportedly good.

The results of the monitoring study revealed an average concentration of 0.36 ppm (std. dev. = 0.3). The values ranged from 0.02 to 2.26 ppm with a median value of 0.3 ppm; these data are presented in greater detail in Table 23.

The results of the statistical analysis done by Versar correlating the formaldehyde levels with the mobile home age are presented in detail in Section 7.3 of this report.

ORNL/CPSC 40 Tennessee Homes Study

From April to mid-December 1982, Oak Ridge National Laboratory (ORNL) with the U.S. Consumer Product Safety Commission (CPSC) studied indoor air quality in 40 east Tennessee homes. The objective of the study was to increase the data base of formaldehyde monitoring in a variety of American homes and further examine the effect of housing types, inhabitant lifestyles, and environmental factors on indoor pollutant levels.

Homes selected for study were restricted to residential urban and semi-urban areas of Oak Ridge and west Knoxville. Selection was stratified to ensure a good representation of house ages, insulation material used, and heating sources. Hawthorne et al. (1984) did not specify whether any of the homes were "non-complaint," although all homes were enrolled voluntarily. No mobile homes were monitored in the study. Eleven of the homes contained urea-formaldehyde foam insulation (UFFI).

Formaldehyde measurements were made using passive membrane samplers. Twice a month, four samplers at each location monitored formaldehyde levels in three rooms and outside the house. Samplers were exposed to the air for 24-hour periods. No modifications to the residents' life

Table 23. Number of Observations Found in Concentration Intervals by Wisconsin Division of Health

Concentration interval (ppm)	Number of observations
(Missing Values)	53
0.0 - .10	85
.11 - .20	199
.21 - .30	180
.31 - .40	137
.41 - .50	90
.51 - .60	78
.61 - .70	51
.71 - .80	35
.81 - .90	21
.91 - 1.00	7
1.1 - 2.00	37
2.1 - 3.00	3
Total	976

Source: Versar statistical analysis of data supplied by Wisconsin Division of Health (1984).

* The 53 missing values were excluded from the statistical analysis described in Section 7 of this report.

styles were requested during these measurements. The sorbent was subsequently analyzed in the laboratory using the pararosaniline colorimetric method; the detection limit using this procedure is approximately 25 ppb. Calibration measurements were conducted in an exposure chamber using a dynamic formaldehyde generation facility. Quality control measurements were conducted approximately once per month using a refrigerated impinger unit operating for 24 hours in one room of a house concurrently being monitored with the passive units.

From the resulting 5,900 measurements, the overall average formaldehyde concentration equalled 0.062 ppm. A more detailed presentation of these results is found in Table 24. Table 25 presents a comparison of formaldehyde levels observed in houses with and without UFFI. Preliminary analysis of the formaldehyde measurements in the 40-home east Tennessee study led to the following major conclusions*:

1. The average formaldehyde levels exceeded 100 ppb (0.1 ppm) in 25 percent of the homes.

2. Formaldehyde levels were found to be positively related to temperature in homes. Houses with UFFI were frequently found to exhibit a temperature-dependent relationship with measured formaldehyde levels.

3. Formaldehyde levels generally decreased with increasing age of the house. This is consistent with decreased emission from materials due to aging.

4. Elevated levels were found in new houses that did not contain UFFI.

5. Formaldehyde levels were found to fluctuate significantly both diurnally and seasonally for homes of all ages.

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*It should be noted that considerable information concerning temperature, humidity, air exchange rates, combustion sources, and various housing structural characteristics was also gathered during this study. Detailed analyses to determine any correlation between the variables and measured formaldehyde levels have not yet been completed.

Table 24. ORNL/CPSC Mean Formaldehyde Concentrations (ppm) as a Function of Age and Season (Outdoor Means Are Less Than 25 ppb Detection Limit)

Age of house	Season	x^a	s^a	m	n
all	all	0.062	0.077	5903	40
0-5 years	all	0.084	0.091	3210	18
5-15 years	all	0.042	0.042	1211	11
older	all	0.032	0.042	1482	11
0-5 years	spring	0.087	0.093	1210	
	summer	0.111	0.102	1069	
	fall	0.047	0.055	931	
5-15 years	spring	0.043	0.040	626	
	summer	0.049	0.048	326	
	fall	0.034	0.035	259	
older	spring	0.036	0.051	757	
	summer	0.029	0.037	341	
	fall	0.026	0.023	384	
all	spring	0.062	0.076	2593	
	summer	0.083	0.091	1736	
	fall	0.040	0.047	1574	

Note: x = mean concentrations.
s = standard deviation.
m = number of measurements.
n = number of homes.
Includes homes with and without UFFI.
[a] Not detected values were assumed to be equal to 12.5 ppb (e.g., one-half the detection limit).

Source: Hawthorne et al. (1984).

Table 25. ORNL/CPSC Formaldehyde Levels Observed in Houses with and without UFFI

Age of house	Type of house	No. of houses	Mean concentration (ppm)
2 to 5 years	UFFI - prefit	7	0.090
15 to 35 years (UFFI age: 2-4 years)	UFFI - retrofit	4	0.055
2 to 5 years	non-UFFI	5	0.115
all	non-UFFI	29	0.060

Source: Hawthorne et al. (1984).

Minnesota State Health Study

The Minnesota State Health Department reported data from 109 mobile homes sampled over a nine-month period following the department's educational programs instituted to inform physicians and the public about potential formaldehyde exposure symptoms. Mobile homes sampled are considered "complaint" homes in that monitoring was requested by the occupants or family physician. Data included age of the mobile home, measured level of formaldehyde, and symptoms reported on a detailed questionnaire. The average age of the sampled mobile homes was less than 2 years, and the average formaldehyde level was less than 0.61 ppm. The formaldehyde levels were inversely related to the age of the mobile homes.

The only information available on the results of the study was found in a Technology & Economics, Inc. Report (Stone et al. 1981) and is presented in Table 26.

Tennessee Department of Health and Environment Investigations

During the period of March 1982 through September of 1983, the Tennessee Department of Health and Environment sampled 132 mobile homes where physicians had indicated that the homeowners were experiencing symptoms consistent with formaldehyde exposure. Two-hour samples were collected and analyzed in each home using the NIOSH P&CAM 125 method.*

The results of the investigation are summarized in Table 27 (from Hodges 1984) for the 55 sampled homes for which mobile home age information is available. The average formaldehyde concentration measured in another 77 mobile homes for which no age information is available was 0.30 ppm (range: 0.02 to 1.43 ppm). Formaldehyde was detected in all 132 sampled homes.

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*Personal communication between G. Schweer (USEPA/OTS) and R. Foster (Tennessee Department of Health and Environment) on October 24, 1984.

Table 26. Summary of Formaldehyde Monitoring Data from Complaint Homes Collected by the Minnesota State Health Department

Cases reported	109
Duration of sample (months)	9
Percent of Mobile homes	
< 1 year old	37
1-2 years old	24
2-3 years old	13
Percent of Mobile homes with levels Below 1 ppm	83
Average formaldehyde level in all sample (ppm)	0.61
Average formaldehyde level for mobile homes < 2 years old (ppm)	0.83

Source: Stone et al. (1981).

Table 27. Summary of Formaldehyde Concentrations Measured in Complaint Mobile Homes in Tennessee from March 1982 through September 1983

Mobile home Age (Yrs)	Number of homes sampled	Mean conc. (ppm)	Min. conc. (ppm)	Max. conc. (ppm)
<2	9	0.225	0.11	0.459
2	14	0.310	0.053	1.92
2.5	4	0.288	0.043	0.483
3	9	0.383	0.132	0.814
4	3	0.190	0.137	0.283
5	5	0.122	0.018	0.264
6			-	-
7	1	0.091	0.091	0.091
8	3	0.068	0.054	0.090
9	1	0.056	0.056	0.056
10-13	6	0.058	0.033	0.10
All	55	0.233	0.018	1.92

Source: Hodges (1984).

Kentucky Department for Health Services Investigations

During 1979-1980, the Kentucky Department for Health Services received 139 formaldehyde-related complaints from residents of mobile homes of which 103 were investigated and sampled (Conyers 1984). With the exception of one home that was sampled during September of 1979, all samples were collected in 1980. Formaldehyde was detected in all but two of the homes. The average level of formaldehyde detected was 0.43 ppm with a range of 0.01 to 1.99 ppm. Over half of the samples collected were above 0.3 ppm. Samples were collected from mobile homes manufactured in 1969 through 1980. The majority of samples were obtained from homes manufactured during the period 1978 through 1980. Data compiled in Kentucky are further detailed in Table 28.

SAI California Formaldehyde Survey

In an effort to assess the overall formaldehyde exposure problem in California, the California Air Resources Board contracted Science Applications, Incorporated (SAI, 1984) to evaluate formaldehyde emission from all sources, and estimate resulting airborne concentrations and human exposure. As part of the study 73 residences (64 non-mobile homes, 6 "new" non-mobile homes, and 3 mobile homes) were passively sampled for indoor formaldehyde levels -- mean concentrations found were 0.05, 0.08, and 0.11 ppm, respectively.

The Lawrence Berkeley Laboratory Passive Diffusion Sampler was used to monitor all the residences for one week periods (168 hours). Results were reported as time-weighted average concentrations. Other variables evaluated in each home during the monitoring period included:

- Residence type (single or multiple unit)
- Owner/Renter occupied
- Urban/rural
- Geographic location (within California)
- Age of residence
- Type of heating
- Type of insulation
- Age of furniture (i.e., cabinets, carpeting)
- Number of rooms

Table 28. Summary of Formaldehyde Concentrations Measured in Complaint Mobile Homes in Kentucky from September 1979 through December 1980

Year of manufacture	Number of homes sampled	Mean conc. (ppm)	Min. conc. (ppm)	Max conc. (ppm)
1980	8	0.85	0.63	1.53
1979	31	0.73	0.14	1.99
1978	17	0.44	0.01	0.87
1977	7	0.28	0.10	0.72
1976	10	0.25	0.08	0.53
1975	5	0.11	0.04	0.23
1974	8	0.12	0.04	0.31
1973	7	0.11	0.04	0.28
1972	3	0.10	0.01	0.22
1971	2	0.06 (0.04)*	<0.04	0.08
1970	1	0.01	0.01	0.01
1969	4	0.08 (0.07)*	<0.04	0.19
All	103	0.43	0.01	1.99

Source: Conyers (1984).

* Values in parentheses indicate mean concentrations when "not detected" values are assumed to be zero

- Amount of cooking performed
- Window use
- Fireplace use
- Cigarettes smoked

No information was reported on types or amounts of formaldehyde emitting materials (i.e., pressed-wood) present in the monitored residences. In addition, infiltration rates, which can be critical in determining indoor air pollutant concentrations, were not measured.

The statistical evaluation of many of the variables with correlative formaldehyde levels had limited significance. In general, homes with gas cooking and cigarette smoking (12 homes) were found to have significantly higher formaldehyde concentrations (by 0.02 ppm on average) than homes with electric cooking and no smoking (16 homes). The curve of formaldehyde concentration to age of home showed erratic decay (instead of a steady concentration reduction) for both the mobile and non-mobile homes monitored.

The mean formaldehyde concentration for the 64 non-mobile home residences was 0.0498 ppm, with a standard deviation of 0.021 ppm. Concentrations ranged from 0.018 to 0.120 ppm. Concentrations in the 6 "new" non-mobile home residences ranged from 0.046 to 0.153 ppm. The mean and standard deviation were 0.0845 and 0.0375 ppm, respectively. Formaldehyde concentrations in the three mobile homes ranged from 0.068 to 0.144 ppm and had a mean and standard deviation of 0.114 and 0.0404 ppm, respectively.

Naval Housing Study

The U.S. Department of Energy sponsored measurements of indoor air quality and air infiltration in recently constructed government housing.

The study (Parker et. al. 1984) included three units of a multifamily housing complex at the Naval Submarine base in Bangor, Washington, over 5 consecutive days during the heating season of 1983. Three dwelling units of identical size constructed in 1978 were monitored, each in a separate two-story four-unit complex. Two of the units were occupied by smokers. None of the units had combustion appliances.

Formaldehyde was measured indoors and outdoors using an Air Quality Research Inc. PF-1 passive integrated monitor. The monitor is capable of detecting formaldehyde concentration as low as 0.001 ppm over a 7-day exposure period. In addition to monitoring of other conventional indoor air pollutants, indoor and outdoor temperature and windspeed were also recorded. Indoor air exchange was measured about three times during each 24-hour period, using a perfluorocarbon tracer with automatic tracer sampling.

Average formaldehyde concentrations measured indoors at the three homes ranged from 0.005 to 0.124 ppm. The only outdoor formaldehyde value reported in the literature (Parker et.al. 1984) was 0.01 ppm. The daily average air exchange rates ranged from 0.22 to 0.91 air changes per hour (ACH).

Houston Housing Survey

As part of a pilot study to determine the quality of indoor air for a cross section of housing types in southern urban areas, the University of Texas, School of Public Health (Stock and Mendez 1985) conducted a study of formaldehyde concentrations inside 78 homes in the Houston, Texas, area during the summer of 1980. Mobile homes and residences with urea-formaldehyde insulation (UFFI) were not included in the study. No homes characterized by occupant complaints were used.

Air sampling was performed by means of a specially designed multi-pollutant sampling unit which consisted of the following components: a high-flow personal sampling pump with a high capacity battery pack, a six-port stainless steel sampling manifold, and an impinger sampling train for formaldehyde collection. Air sampling and analysis for formaldehyde were performed according for the chromotropic acid procedure.

Indoor concentrations range from below the limit of detection (approximately 0.008 ppm) to 0.29 ppm, with an average value of 0.07 ppm for detectable concentrations (N=75). Only 8 of 13 outdoor measurements resulted in a detectable concentration; the average of these was 0.02 ppm. The difference between the indoor and outdoor means was statistically significant ($p<0.05$). A probability plot presented in the literature (Stock and Mendez 1985) indicates that the indoor formaldehyde concentrations can be reasonable approximated by a log-normal distribution.

Energy efficient condominiums had, as a housing category, the highest mean level (0.18 ppm). Condominiums, apartments, and energy-efficient houses represented the mid-range with mean levels of 0.09, 0.08, and 0.07 ppm, respectively; the mean in conventional houses was 0.04 ppm. Home formaldehyde levels declined with home age.

Wagner 16 California Home Study

As part of a M.S. thesis at the University of California, Berkeley, the author (Wagner 1982) monitored indoor air quality in 16 California homes that fell into a prescribed "worst case" category of building and occupancy characteristics. The worst case criteria included: low infiltration rate, low natural ventilation rates, presence of gas stoves, and new construction.

Monitoring took place between January 13 and February 24, 1982. Formaldehyde sampling was conducted using the LBL sodium bisulfite passive formaldehyde monitor and conventional bubblers. Time-weighted weekly average formaldehyde concentrations in twelve low infiltration homes, measured by passive samplers, ranged from 0.078 to 0.163 ppm. Formaldehyde values from the remaining houses were not reported in the literature. Average infiltration rates for the heating season ranged from 0.19 to 0.50 ACH, in all cases well under the projected design rates of 0.6 and 0.9 ACH estimated for California's new building standards.

4.2 Studies Examining Factors Affecting Air Levels

Dutch Study with Coated Particleboard

J.F. Van der Wal (1982) of the TNO Research Institute for Environmental Hygiene (The Netherlands) measured formaldehyde concentrations in Dutch homes where particleboard was used. In response to inhabitants' complaints, 36 houses were monitored during the period 1977 to 1980. The objective of the study was to investigate the number of homes that violated the 1978 Threshold Limit Value (and legal ceiling value) of 120 ug/m^3 (0.1 ppm) set by the Dutch, and to evaluate the effectiveness in reducing formaldehyde levels of coating the particleboard with a special vinyl-toluene paint.

Analysis was performed using the pararosaniline method. The reproducibility was reported to be $\pm$ 10 percent at 100 ug/m^3, with a detection limit of 5 ug/m^3. Temperature, relative humidity, and ventilation rate were also measured. The following efforts were made in an attempt to standardize the environmental conditions in each home at the time of sampling:

- Room temperatures were manually adjusted to fall as close to 20°C as possible 12 hours before each measurement.

- Ventilation rate was adjusted manually (opening and closing windows) to fall between 0.5 and 1.0 changes per hour.
- Relative humidity was not adjustable, and regulation was not attempted.
- Presence of alternative formaldehyde sources was prevented as much as possible. Smoking, use of natural gas burners, detergents, shampoos, etc., were not permitted.

Table 29 presents the highest formaldehyde concentrations measured in the Dutch houses. Of the 36 houses investigated, only 7 had a formaldehyde concentration throughout (in every room sampled) less than the 120 ug/m^3 (0.1 ppm) ceiling. The highest value was 1.8 mg/m^3 (1.4 ppm). Neither a complete range of measured concentrations nor a mean value was reported in the study.

Table 30 presents results of coating the partcleboard used in inner walls and roof plates of five of the Dutch homes. Formaldehyde concentrations were decreased by a factor of 1.5 to 3.0. Van der Wal concluded that when accounting for all the factors influencing indoor concentrations in this study, diffusion-retarding paint coating on particleboard will not decrease indoor formaldehyde concentrations by more than about 50 percent.

University of Iowa Study

The University of Iowa Study (Schutte et al. 1981), performed for the Formaldehyde Institute, monitored 31 conventional, detached homes not containing urea-formaldehyde foam insulation (UFFI) for formaldehyde concentrations in the indoor air. Samples were evaluated in relation to outdoor formaldehyde concentrations, age of the home, and other environmental factors monitored at each of the sampled homes.

Table 29. Highest Measured Formaldehyde Concentrations in Dutch Houses

Location	Form. conc. ug/m³	Form. conc. ppm	Room where highest conc. was measured	Remarks
Oudenbosch	300	0.241	attic	not inhabited
Haarlem, house 1	820	0.658	bedroom	
Haarlem, house 2	960	0.770	bedroom	
Haarlem, house 3	1800	1.444	hall	
	1100	0.882	bedroom	
Haarlem, house 4	290	0.233	living room	
Drachten	250	0.201	bedroom	not inhabited
Leewarden, house 1	540	0.433	bedroom	
Leewarden, house 2	750	0.602	bedroom	
Leewarden, house 3	250	0.201	bedroom	
Leewarden, house 4	220	0.176	bedroom	
Leewarden, house 5	200	0.160	living room	
Leewarden, house 6	280	0.225	attic	
Leewarden, house 7	390	0.313	attic	
Leewarden, house 8	150	0.120	attic	
Leewarden, house 9	330	0.265	bedroom	
Leewarden, house 10	290	0.233	living room	
Emmen, house 1	150	0.120	attic	
Emmen, house 2	70	0.056	bedroom	
Emmen, house 3	30	0.024	bedroom	
Emmen, house 4	60	0.048	bedroom	
Schoonebeck	40	0.032	bedroom	
Diemen, house 1	290	0.233	attic (bedroom)	
Diemen, house 2	220	0.176	attic (bedroom)	
Lelystad, house 1	250	0.201	study	not inhabited
Lelystad, house 2	320	0.257	bedroom	show house
Waddinxveen	150	0.120	living room	
Monster	230	0.184	bedroom	
Zaandam, house 1	350	0.281	bedroom	
Zaandam, house 2	140	0.112	attic (bedroom)	
Zaandam, house 3	110	0.088	bedroom	
Zaandam, house 4	170	0.136	attic (bedroom)	
Zaandam, house 5	150	0.120	bedroom	
Zaandam, house 6	110	0.088	bedroom	
Zaandam, house 7	150	0.120	bedroom	
Hellendam	90	0.072	attic	

Source: Van der Wal (1982).

Table 30. Formaldehyde Levels in Dutch Houses Before and After Panel Coatings

House	Room	Before coating HCHO conc. ug/m^3	ppm	Temp °C	RH %	ACH h^{-1}	After coating HCHO conc. ug/m^3	ppm	Temp °C	RH %	ACH h^{-1}
Leewarden											
house 2	bedroom 1	750	0.602	19	43	0.5	430	0.345	21	54	1.0
	living room	400	0.321	21	54	0.4	170	0.136	19	60	0.4
house 4	bedroom 1	220	0.176	18	57	0.9	130	0.104	19	60	0.9
	living room	80	0.064	19	58	0.7	70	0.056	20	68	0.5
house 6	attic	280	0.225	18	56	1.6	180	0.144	18	53	0.9
	living room	180	0.144	22	51	0.6	60	0.048	19	55	0.4
house 7	attic	390	0.313	17	54	0.6	230	0.184	19	50	0.5
	living room	130	0.104	23	55	1.0	90	0.072	22	50	0.9
Diemen											
house 1	living room	60	0.048	21	60	1.1	60	0.048	23	66	0.4
	bedroom 1st stock	100	0.080	23	62	0.2	100	0.080	25	72	0.2
	attic bedroom northern side	290	0.233	23	55	0.3	210	0.168	24	66	0.6
	attic bedroom southern side	190	0.152	24	53	1.0	120	0.096	24	62	0.7

Source: Van der Wal (1982).

Note: RH = relative humidity;
ACH = air exchange rate (exchanges per hour)

Twelve 8-hour formaldehyde samples were taken in each dwelling, using the modified NIOSH PCAM #125 method (1 percent bisulfite absorber and dual impingers). Samples were taken at different positions in each house including the kitchen, living room, bedroom, and family room.

The results of the formaldehyde monitoring are presented in Table 31. The average indoor concentration found in the homes was 0.063 ppm (std. dev. = 0.064) with a range of 0.013 to 0.34 ppm. The average outside formaldehyde concentration was 0.002 ppm (std. dev. = 0.0013). In addition, the correlation (from a linear regression) of the natural log [CH_2O] versus age of the home resulted in a correlation significance at the 95 percent confidence level (R = -0.42). This is comparable to the statistical analysis performed by Versar on the Wisconsin and Clayton formaldehyde in mobile home data (see Section 7.3 of this report). Fitted coefficients were not provided by the University of Iowa study, however, so comparison of actual decay curves is not possible without evaluating all the raw data.

In addition to age of the home, several other parameters were tested for correlation with home formaldehyde concentration. The following correlations were significant at the 95 percent confidence interval: number of occupants; hours of home occupation; and inside relative humidity. The linear regression slopes of the above three parameters with formaldehyde level were negative. Although significant correlations were not observed between particleboard and paneling loading rates and formaldehyde levels for all homes combined, significant correlations (at the 95 percent confidence interval) were observed for (1) paneling in those homes which when tested had their windows closed and air conditioning systems on and (2) for particleboard in those homes which when tested had either their windows open or closed and air conditioning

Table 31. Formaldehyde Concentrations Found in Conventional Homes Monitored by the University of Iowa

Home #	Average home CH_2O concentration (ppm)	Outside CH_2O concentration (ppm)
1	0.048	0.001
2	0.037	0.001
3	0.025	—
4	0.013	0.003
5	0.017	0.001
6	0.018	—
7	0.038	—
8	0.084	0.004
9	0.063	0.004
10	0.045	0.002
11	0.018	0.001
12	0.014	0.001
13	0.047	0.002
14	0.040	0.003
15	0.044	0.003
16	0.050	—
17	0.068	0.003
18	0.019	<0.001
19	0.043	0.003
20	0.340	0.002
21	0.061	0.003
22	0.027	0.001
23	0.100	0.006
24	0.058	0.004
25	0.069	0.003
26	0.200	0.004
27	0.043	0.003
28	0.120	0.001
29	0.034	0.002
30	0.120	<0.001
31	0.054	0.003
	$\bar{x}$ = 0.063 s = 0.064 Range = 0.013–0.34	$\bar{x}$ = 0.002 s = 0.001 Range = 0.00041–0.0056

Source: Schutte et al. (1981)

systems off. In general, it was found that alteration of homes from high ventilation (open windows) to low ventilation (closed windows) resulted in an approximate doubling of indoor formaldehyde concentration.

Indiana Studies

A report by Virgil J. Konopinski (1983) of the Indiana State Board of Health summarizes the results of a series of investigations conducted from 1979 through 1983 to determine formaldehyde levels in conventional homes in Indiana. The purpose of the 1983 report was to compare the levels found in homes with urea-formaldehyde foam insulation (UFFI) to the levels found in those homes using other types of insulation.

Airborne formaldehyde was sampled using a midget impinger sampling train and by taking area samples. The impinger sampling was done using a battery powered vacuum pump capable of sampling for at least eight hours duration; actual sample time for this study was two hours. The pump was fitted with a liquid trap and calibrated for an airflow of 1 liter per minute. Air samples were collected in a 1 percent bisulfite absorbing solution. Sample volume varied from 30 to over 100 liters with a sample volume of 45 to 60 liters for most samples. Formaldehyde samples were analyzed following the procedures outlined in NIOSH Method 125, chromotropic acid procedure.

Table 32 summarizes the results of the Indiana Board of Health monitoring study. A total of 239 homes were sampled for formaldehyde, 119 of which contained UFFI and 120 of which contained some other type of insulation. Health problems were reported by the occupants of 103 of the homes (66 UFFI homes and 37 non-UFFI homes). Neither the age of the home nor the age of the UFFI installations was reported. It should be noted that UFFI was not considered the sole source of formaldehyde in those

Table 32. Formaldehyde Levels Found in Indiana Study

	Residences with UFFI	Residences without UFFI
Number of samples	119	120
Mean concentration of formaldehyde-ppm[a]	0.05	0.09
Maximum concentration of formaldehyde - ppm	0.18	1.35
Minimum concentration of formaldehyde - ppm	Not detectable	Not detectable
Number of nondetectable situations	18	35

Source: Konopinski (1983).

[a]"Not detected" values were assigned a zero concentration for calculation of mean concentrations

homes containing it. The outdoor concentration of formaldehyde was also measured in 47 situations; the mean concentration was 0.005 ppm. There were 28 instances of non-detectable formaldehyde for outdoor measurements.

Godish (1983) reported the results of monitoring for formaldehyde in 28 residences containing UFFI, but no particleboard flooring, and 29 residences that contained neither UFFI nor particleboard flooring, cabinetry or paneling. Ninety minute air samples were collected and analyzed using the modified NIOSH bubbler/chromotropic acid procedure. Formaldehyde levels in the UFFI residence ranged from 0.02 to 0.13 ppm with mean and median values of 0.07 ppm. Levels in conventional residences containing miscellaneous low-level sources (e.g., carpeting, upholstery, furniture) but no particleboard or UFFI had a range of 0.03 to 0.07 ppm with mean and median values of 0.05 ppm.

Meyer and Hermanns Studies Showing Effect of Temperature In and Around Mobile Homes

Meyer and Hermanns (1984b) reported the result of field studies on diurnal fluctuations in formaldehyde indoor air concentrations in a mobile home in Florida during the summer. They found substantial variations, and related those variations to changes in indoor wall temperatures as a function of solar radiation or simply ambient outdoor temperature. They describe peaks in indoor air levels corresponding to times of the day when the sun strikes the mobile home; levels declined when the temperature dropped. There was an approximately one-hour time lag between the temperature peaks and concentration peaks. Figure 18 illustrates the calculated time-weighted levels as a function of time of day. It is not clear from their report whether the indoor air temperature was allowed to vary with the ambient temperature. Figure 19, reproduced from Meyer and Hermanns (1984a), illustrates temporal variability in data reported by George Myers for an unoccupied mobile home.

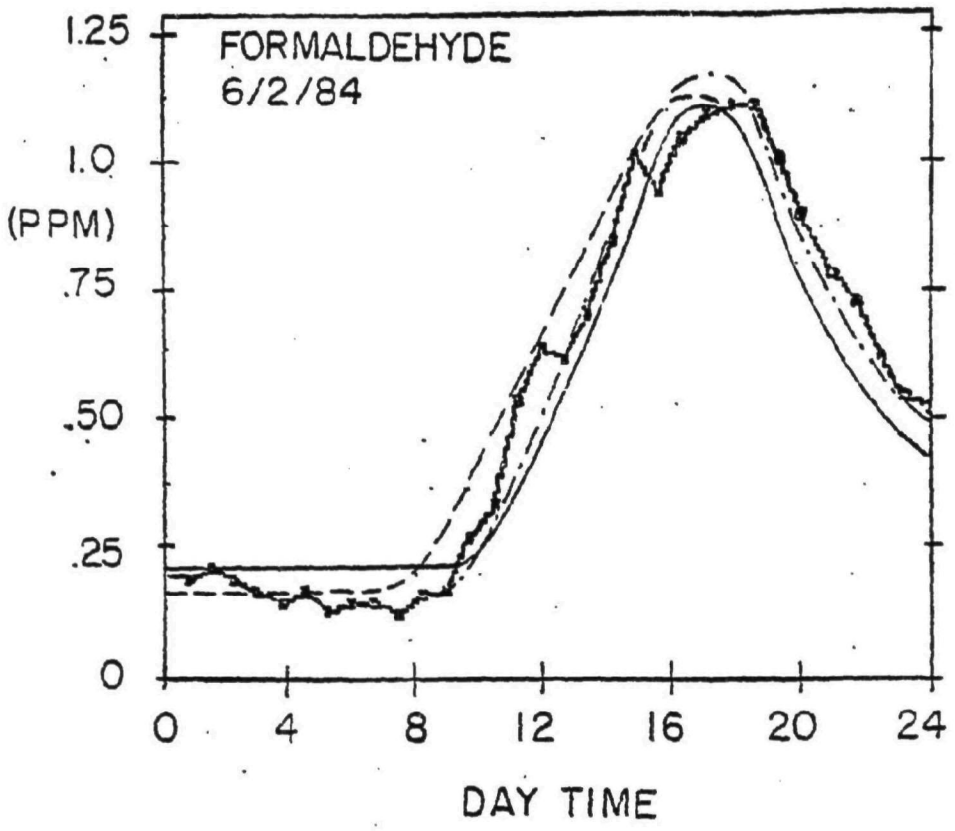

Figure 18. Calculated Time-Weighted Average Formaldehyde Levels in a Mobile Home

Source: Meyer and Hermanns (1984b).

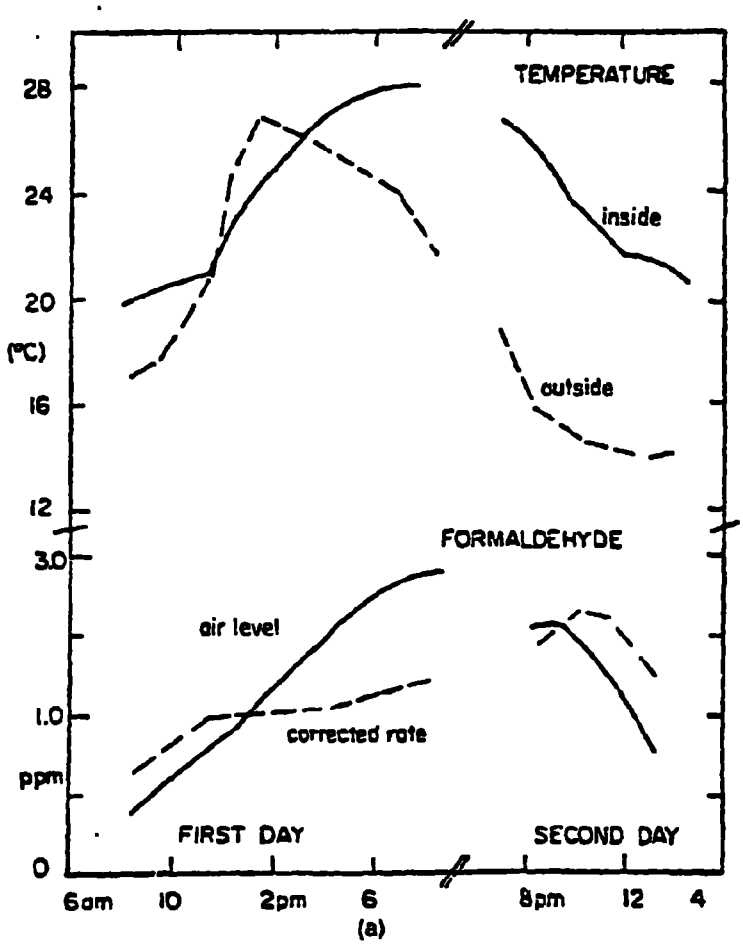

Figure 19. Formaldehyde Levels in a New, Unoccupied Mobile Home as a Function of Time of Day and Temperature

Source: Meyer and Hermanns (1984a).

A similar study was recently conducted in Texas during the summer months using two mobile homes (University of Texas 1984). The indoor temperature of one mobile home was allowed to vary with ambient temperature while the temperature in the other home was maintained at about 22°C with air conditioning. The results reported for the first home are very similar to the results of Meyer and Hermanns (1984b). However, the controlled temperature home had much lower variation in formaldehyde concentration throughout the day, presumably due to an increased air exchange rate caused by the air conditioning and the indoor/outdoor temperature differential.

Fleming and Associates Study

The objective of this study (Traynor and Nitschke 1984) was to monitor residences for nitrogen dioxide, carbon monoxide, formaldehyde, respirable suspended particles, and air exchange rates where suspected combustion-related indoor pollution sources could be readily identified. These sources and associated formaldehyde levels are summarized in Table 33. The average formaldehyde level observed in all the test homes was 40 ppb; a high value of 151 ppb was found in one of the tested residences categorized as containing new furnishings and new paneling as a suspected pollution source.

Formaldehyde was monitored in thirty New York state homes for forty-one one-week periods. The sampling was performed primarily during the winter months when the usage of some of the suspected sources was greatest. Formaldehyde was monitored with a passive diffusion sampler developed at Lawrence Berkeley Laboratory. Air exchange rates were monitored using passive perfluorocarbon emitters and collectors.

This study was funded by a private power utility in New York specifically to investigate the role of conbustion-related appliances in indoor pollution. The investigators, contacted by phone, acknowledged that little more was to be done on the project other than submitting a final report to the utility. A copy of the draft report will be made available to the EPA in early 1985.

Table 33. Summary of One-Week Average Indoor Formaldehyde Data Observed in the Fleming and Associates Study

Source/house code	Air exchange rate[a] (h^{-1})	House volume (m^3)	HCHO[a] (ppm)
No source			
14	0.11	350	0.077
16	0.24[b]	508	0.026
24	0.40[b]	329	0.034
50-1	0.15	644	0.007
New furnishings			
23	0.25	429	0.061
45	0.13	483	0.015
61	0.26[b]	480	0.023
Smokers (S)			
02-1	0.17	473	0.060
02-2	0.16	473	0.056
36	0.37	455	0.040
50-5	0.12	644	0.032
Kerosene-fired space heater (KH)			
20	0.30[b]	468	0.031
32-1	0.19	701	0.032
50-2	0.13	644	0.025
Wood-burning stove (WS)			
25	0.10	733	0.031
44	0.10[b]	606	0.036
51-1	0.12	443	0.012
Coal-burning stove (CS)			
31	0.11	1020	0.028
Fireplace w/wood (FW)			
47	0.15	433	0.019
50-3	0.16	644	0.018

Table 33. (continued)

Source/house code	Air exchange rate[a] (h^{-1})	House volume (m^3)	HCHO[a] (ppm)
Fireplace w/coal (FC)			
50-4	0.11	644	0.019
Gas-fired range (GR)			
05-2	0.28	472	0.056
50-6	0.13[d]	644	0.036
Gas-fired furnace (GF)			
01	0.30	315	0.026
21	0.32	379	0.048
43	0.32[b]	652	0.017
Oil-fired furnace (OF)			
17	0.35	682	0.023
38	0.06	798	0.027
56	0.32[b]	588	0.022
Combination of sources			
03 (S, WS)	0.27	289	0.024
05-1 (WS, GR)	0.33	472	0.062
10 (OF, SW)	0.07[b]	690	0.064
18-1 (S, KH, GR)	0.57	441	0.039
18-2 (S, KH, GR)	0.57[c]	441	0.032
22 (CS, WS)	0.17[b]	697	0.020
32-2 (WS, KH)	0.33	701	0.046
32-3 (CS, KH)	0.14	701	0.053
33 (WS, KH)	0.24	579	0.022
51-2 (WS, GR)	0.13	443	0.013
55 (S, AG)[d]	0.09	270	0.047
60 (S, GR, GF)	0.11[b]	468	0.059

[a]Reported standard deviations, based on multiple measurements at different indoor locations, were not included in this table.

[b]Based on average ratio of the measured air exchange rate to the air exchange rate at 50 pascals (0.049 ± 0.029).

[c]Average air exchange rate of house measurements made during other time periods.

[d]Attached garage.

Source: Traynor and Nitschke (1984).

4.3 Ongoing Monitoring Studies

California Mobile Home Survey*

The California Department of Health Services is currently analyzing the results of a large formaldehyde monitoring survey of 700 mobile homes conducted during August and September of 1984. This is apparently the largest coordinated survey of mobile home formaldehyde exposures ever undertaken in the United States. The survey was designed to be as "random" as possible and to be stratified by age of mobile home. Approximately 60 percent of the sampled homes were less than three years of age. Thus, the results should be useful in determining whether current efforts (i.e., within the last several years) by industry to reduce formaldehyde emissions from pressed wood products have been successful in actually reducing in-home formaldehyde exposures. Monitoring was performed using the five- to-seven-day exposure Air Quality Research passive dosimeters.

The preliminary results of the survey will not be available until the first quarter of 1985. The California Department of Health Services hopes to repeat the survey of the same homes in February of 1985 so as to obtain winter indoor air levels that can be compared to the summer indoor air levels obtained in the first survey. The results of this second survey will not be available until the third quarter of 1985.

Bonneville Power Administration Surveys**

The Bonneville Power Administration (BPA) in the State of Oregon is initiating two large-scale surveys of the levels of formaldehyde and

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*Personal communication between G. Schweer (USEPA/OTS) and Dr. K. Sexton (California Department of Health Service) on 10/17/84.

**Personal communication between G. Schweer (USEPA/OTS) and R. Rothman (Bonneville Power Administration) on 7/24/84 and 11/19/84

several other pollutants in conventional housing. The goals of the surveys are to determine the effects of energy conservation techniques on indoor pollutant levels both in new and existing electrically heated housing.

Approximately 150 existing homes (typically 5 years or older) and as many as 600 new homes will be screened for formaldehyde levels using the Air Quality Research/Lawrence Berkeley Laboratory passive dosimeter (which measure five to seven days' exposure). Approximately 40 existing homes and 100 new homes will be selected for more in-depth testing of the effects of various energy conservation retrofit techniques and ventilation controls including air-to-air heat exchanges. Preliminary results are not expected before the spring of 1985. The studies will probably continue through the winter of 1985/1986.

DiNardi/Rush-Hampton House Study

The DiNardi/Rush-Hampton house is a 3600 square foot contemporary, passive solar house located in Amherst, Massachusetts. This house is instrumented for the continuous analysis of hydrocarbons, formaldehyde, carbon monoxide, infiltration, ambient meteorological conditions, insolation, energy consumption, and indoor thermal comfort parameters.

The overall project has many objectives, all focusing on the effect of different factors on indoor air contaminant levels. The factors being considered range from broad-based spatial, temporal, and seasonal variations to the very specific infiltration rate, temperature, and inhabitant living activities. For formaldehyde specifically, the study hopes to compare several different analytical techniques used by their laboratories (i.e., the NIOSH chromotropic acid method, the DNPH2 method, and an automated pararosaniline analyzer), as well as to evaluate the effectiveness of several indoor air treatment regimes on low-level formaldehyde concentrations (i.e., air-to-air heat exchangers and air ventilator/washers).

Air sampling, including tha. for formaldehyde, is accomplished with a sample inlet connected to a 5-point all Teflon sequential sampling system which is controlled automatically by the microprocessor in the data acquisition system. The sampling lines are 1/4 inch Teflon tubing extending from the 5-point sampling system in the laboratory to sampling ports located in various rooms throughout the house. The locations sampled with this system are zero air, permeation calibration source, master bedroom, kitchen and ambient (DiNardi et al. 1984).

According to the researchers*, the data on formaldehyde levels in the DiNardi/Rush-Hampton house collected during the last heating season (last winter) are still being evaluated. Preliminary results, however, show formaldehyde levels (hourly averages) in the range of 30 to 80 ppb (0.03 to .08 ppm). The data will be formally presented and available for distribution in early 1985.

Future plans include one more study, from January to March 1985, evaluating indoor formaldehyde levels and comparing analytical techniques.

Texas Indoor Air Quality Study

The Texas Indoor Air Quality Study, being performed by the University of Texas, School of Public Health (1983), is an ongoing, in-depth indoor air monitoring study involving a total of 164 "non-complaint" mobile homes in four Texas counties. In addition to providing one of the largest data bases on formaldehyde levels in mobile homes, this study will ultimately provide information on many related issues, such as: effects of air exchange; comparison of long- and short-term formaldehyde levels; a study of air filter intervention; a cooking fuel emission study (see Section 3.0 for a summary of preliminary results on this); a comparison of four types of formaldehyde monitoring methods; an architectural study of mobile home designs and furnishings; and a study

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*Personal communication between S.R. DiNardi, University of Massachusetts, and T. Chambers, Versar Inc. November 29, 1984.

of the effects of temporal and environmental factors. As stated in the report, "any conclusions drawn from the data produced by this study at this time may be subject to revision upon further analysis .... The influence of specific architectural characteristics of the homes on the observed formaldehyde levels has yet to be determined" (University of Texas 1983).

The sampling for formaldehyde (as well as respirable suspended particulates, aeroallergens, and other volatile organics) was performed from October 1982 through August 1983. On a typical sampling day, monitoring equipment would be assembled inside the mobile homes by about 11 a.m.; the monitoring period would begin by 12 p.m. and finish at about 7 p.m. Mean daily levels of formaldehyde were measured with the CEA-TGM-555 continuous monitor and are presented in Table 34 by age of mobile home within each county. The overall mean for sample sets equalled 0.12 ppm. All mobile home age groups in El Paso have approximately the same mean formaldehyde concentration during the first sampling period of 0.05 to 0.07 ppm, as well as the lowest levels measured in all counties. This is most probably explained by the predominant use of evaporative air coolers (in use in 98 percent of the mobile homes) during April; these significantly increase the air exchange rate. Although 49 percent of the mobile homes in Midland also use evaporative coolers, sampling was performed during March, when they would not be in use.

To determine the variability of formaldehyde concentrations over short time periods and the factors potentially affecting the levels, sequential measurements were taken daily over two one-week periods. One-Week Study I was done during June 1983, and One-Week Study II was performed during September 1983. During both studies, a two-hour dual 20 ml (one percent bisulfite) impinger sample was collected at 150 ml/min., at approximately 8 a.m. and 4 p.m., and dry and wet bulb temperatures were recorded for four consecutive days. On the third day of each study, samples were collected for two hours every four hours for 24 hours,

Table 34. Indoor Mean Formaldehyde Concentrations Measured in 164 Mobile Homes by the Texas Indoor Air Quality Study (Preliminary Results).

Mobile home location/age	First sampling survey				Repeat sample			
	Mean (ppm)	±sd	N	Sampling dates	Mean (ppm)	±sd	N	Sampling dates
Harris Co								
< 1 year	.21	.11	9	October				August 1983
1-2 years	.20	.12	12	1982 -	.17	.05	3	
2-3 years	.20	.12	3	May 1983				
3-4 years	.14	.05	5					
> 4 years	.14	.10	9		.04	.01	3	
Group mean	.18	.11	38					
Tarrant Co.								
< 1 year	.35	.21	10	February 1983				
1-2 years	.19	.11	13					
2-3 years	.23	.13	11					
3-4 years	.21	.06	5					
> 4 years	.22	.07	3					
Group mean	.24	.15	42					
Midland Co.								
< 1 year	.13	.07	19	March 1983				
1-2 years	.09	.04	18					
2-3 years	.09	.04	3					
3-4 years	.04		1					
> 4 years								
Group mean	.11	.06	41					
El Paso Co.								
< 1 year	.07	.05	10	April 1983	.04	.02	4	July 1983
1-2 years	.05	.05	12		.12	.14	2	
2-3 years	.06	.05	12		.25	.15	2	
3-4 years	.06	.04	4					
> 4 years					.03		1	
Group mean	.06	.05	38		.10	.12	9	

Table 34. (Continued)

Mobile home location/age	First sampling survey				Repeat sample			
	Mean (ppm)	±sd	N	Sampling dates	Mean (ppm)	±sd	N	Sampling dates
All Counties Except El Paso								
< 1 year	0.21	—	38					
1-2 years	0.15	—	43					
2-3 years	0.20	—	17					
3-4 years	0.16	—	11					
>4 years	0.16	—	12					
Group mean	0.18	—	121					

Source: University of Texas (1983).

N = number of samples

sd = standard deviation

starting with the 8 a.m. sample. During One-Week Study I, samples were collected simultaneously in the living room, main bedroom, and second bedroom. Samples were collected in the living room area only during Study II.

Over the three-month period between the One-Week Studies, the formaldehyde concentration decreased by approximately 0.1 ppm. Table 35 shows the comparison of the total one-week formaldehyde concentrations and temperature measurements for the two studies. The indoor temperatures in mobile home No. 1 and 2, were similar for both studies. However, from One-Week Study I to II in mobile home No. 1, the formaldehyde level decreased from 1.29 ppm to 1.12 ppm, and in mobile home No. 2, from 0.36 ppm to 0.24 ppm.

University of Wisconsin Survey

The University of Wisconsin, under a grant from the Wisconsin Power and Light Company, is investigating the influence of a residential weatherization program on indoor air quality (Quackenboss et al. 1984). Fifty homes, belonging primarily to low-income or elderly individuals, are being weatherized at no cost to the homeowners.

Prior to the initiation of home weatherization activities, each home was sampled three times during the 1982 to 1983 heating season to determine the levels of indoor air pollutants and air infiltration rates. Air Quality Research passive dosimeters were employed for monitoring formaldehyde concentrations over week long periods. The available published results of the pre-weatherization sampling indicate an overall average formaldehyde concentration of 0.031 ppm (standard deviation of 0.016 ppm) in the 50 homes and a median concentration of 0.028 ppm (Quackenboss et al. 1984). (Additional unpublished information on the individual home formaldehyde levels has been requested from the researchers*).

*Personal communication between Dr. James Quackenboss, University of Wisconsin, and G. Schweer, USEPA-OTS, on 10/1/84 and 12/27/84.

Table 35. Mean Formaldehyde Concentration and Temperature Measurements for Texas Indoor Air One Week Study I and II

	One Week Study I		One Week Study II	
	Mean formaldehyde concentration (ppm)±sd	Mean temperature (°C)±sd	Mean formaldehyde concentration (ppm)±sd	Mean temperature (°C)±sd
Mobile home No. 1	1.29 ± 52	31.9 ± 5.2	1.2 ± .33	32.7 + 4.0
Mobile home No. 2	0.36 ± .07	22.9 ± 1.6	0.24 ± .03	22.4 ± 0.8

NOTE: Indoor air temperatures in mobile home No. 1 were allowed to fluctuate with ambient outdoor temperatures. Indoor air temperatures in mobile home No. 2 were controlled with air conditioning. Thus, due to the higher indoor/outdoor temperature differential in mobile home No. II and the use of the air conditioner, air exchange rates were probably higher in this home. This may account for the lower levels in Home No. 2.

sd = standard deviation

In addition to formaldehyde and air infiltration measurements, information was also gathered on the building materials, physical layout, and furnishings of each home as well on occupant activities that may influence formaldehyde levels (e.g., smoking and use of combustion appliances). This information has not yet been published*.

4.4 European Studies

Switzerland

In a study done by the Swiss Federal Institute of Technology, Department of Hygiene and Applied Physiology, Zurich, Switzerland (Kuhn and Wanner 1984), the formaldehyde content in room air was measured in 8 one-family houses and 38 multiple dwellings.

Formaldehyde was measured with two consecutive gas-washing bottles (midget-impinger) containing an aqueous solution of methyl-benzthiazolon-hydrazon (MBTH), called a "FOMA." The color intensity of the reaction mixture was subsequently evaluated spectrophotometrically.

In the spring (before occupancy), the residential concentrations ranged from 0.2 to 0.7 ppm; a year later, measured concentrations were reduced by about one-half. Table 36 summarizes mean concentrations measured during four seasonal periods.

Holland

In a study performed by the Product Analysis Agency, Haarlem District, the Netherlands, formaldehyde concentrations were measured in 49 houses and 3 homes for the elderly in which particleboard was specifically not used as a building material and in which no urea-formaldehyde foam insulation was used. The formaldehyde concentration, the ventilation flow, the temperature, and the relative humidity were measured in the living rooms, kitchens, and the bedrooms (as well as the approximate age of each building). The analysis was

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*Personal communication between Dr. James Quackenboss, University of Wisconsin) and G. Schweer USEPA/OTS on 10/1/84 and 12/27/84.

Table 36. Formaldehyde Measurements in Swiss Houses over Four Seasonal Periods (ppm)

	Spring I		Summer		Winter		Spring II	
	$\bar{x}$	sd	$\bar{x}$	sd	$\bar{x}$	sd	$\bar{x}$	sd
One-family houses	0.29	±0.12	0.33	±0.04	0.15	±0.04	0.14	±0.03
Multiple family dwellings	0.37	±0.18	0.40	±0.19	0.24	±0.09	0.18	±0.05

Source: Kuhn and Wanner (1984).

$\bar{X}$ = Mean concentration
sd = Standard deviation

intended to provide an insight into the usual concentrations of formaldehyde in homes in which the residents have made no complaints and in which any formaldehyde emanates from self-introduced sources, such as smoking, floor coverings, curtains, gas-powered appliances, open fireplaces, cleaning products, and particleboard-containing furniture.

The investigative monitoring was performed between April 1981 and April 1982. Concentration measurements were taken in two-day periods. Preparation involved standardizing the indoor environment to approximately 18° to 22°C, 0.5 to 1.0 air changes per hour. Five hours before measurement, the areas to be measured were ventilated thoroughly and then the windows and doors were kept closed; no smoking was permitted. The chromotropic acid method and the fluorescence method with acetyl-acetone (Hatzsch reagent) were both used in the analysis. An average of two values per room per sampling event was reported.

The data presented in the study documentation (Cornet 1983) are too voluminous to reproduce in this report. In the documentation, sets of tables are presented for each of the three rooms: living, kitchen, and bedroom. Data include location and age of home, number of residents, smoking behavior or residents, home renovations, outdoor wind speed, wind direction, outdoor temperature, and weather conditions. Also reported are indoor temperature, relative humidity, ventilation rate, materials used for walls, floors, and ceilings, surface and finishing of sheet material, type of heating, usual day and night temperatures, wall cavity material, and secondary formaldehyde sources (such as boilers). Table 37 summarizes the data by presenting average, median, 10th and 90th percentiles, and highest and lowest formaldehyde values found in the three rooms. The average concentration of formaldehyde observed by the study was 0.054 ppm.

Table 37. Average, Median, 10th and 90th Percentiles and Highest and Lowest Values Found in Haarlem District Study (The Netherlands)

Measurement	Average	Median	10th Percentile	90th Percentile	Lowest value	Highest value
Chromotropic acid method						
Formaldehyde concentration standard measurement, living room, in ug/m^3	61	50	32	93	20	152
in ppm	0.049	0.040	0.026	0.075	0.016	0.122
Formaldehyde concentration standard measurement kitchen in ug/m^3	60	54	31	108	3	149
in ppm	0.048	0.043	0.025	0.087	0.002	0.119
Formaldehyde concentration, standard measurement, bedroom in ug/m^3	68	48	24	155	15	288
in ppm	0.055	0.038	0.019	0.124	0.012	0.231
Fluorescence method						
Formaldehyde concentration, standard measurement, living room in ug/m^3	66	63	33	108	17	146
in ppm	0.053	0.051	0.026	0.087	0.014	0.117
Formaldehyde concentration, standard measurement, kitchen in ug/m^3	69	64	26	121	7	203
in ppm	0.055	0.051	0.021	0.097	0.006	0.163
Formaldehyde concentration, standard measurement, bedroom in ug/m^3	77	59	26	161	9	280
in ppm	0.062	0.047	0.021	0.129	0.007	0.225

Source: Cornet (1983).

Germany

Formaldehyde indoor air concentrations are also reported for German homes in a study by Schulze (1975). The results are presented in Table 38. No other information on the study was available.

Sweden

Indoor air formaldehyde concentrations were presented by Sundin (1978) from monitoring performed from September 1975 to October 1977 in 319 Swedish homes. Formaldehyde levels found in the homes tested were attributed to particleboard use and a new type of ceiling panel utilizing an improper glue application technique. Approximately 75 percent of the homes contained the ceiling panels. Also, more than 90 percent of the tests could be categorized as coming from complaint homes.

All analyses were made with the chromotropic acid method, which is the official test method in Sweden for quantitative determination of formaldehyde in the air. No other testing conditions or procedures were available from the related literature. Results ranged from 0.1 to 2 ppm; the average being 0.58 ppm. The results are further summarized in Table 39.

Denmark

Andersen et al. (1975) sampled indoor formaldehyde concentrations in 25 rooms of 23 dwellings (19 houses and 4 flats) from February to September 1973 in suburban areas of Jutland, Denmark. The objective was to evaluate indoor air concentrations in homes that exclusively used chipboard (or particleboard) in walls, floors, and ceilings (with U:F molecular ratios of approximately 1:1.4). Other environmental factors considered in this study included age of house, temperature, humidity, and air changes. The average concentration was 0.50 ppm with a range of 0.06 to 1.79 ppm. A complete summary of results is presented in Table 40.

Table 38. Formaldehyde Concentrations in German Homes (ppm)

	Kitchen	Living room	Average
First new building	0.129	0.081	0.105
Second new building	0.068	0.060	0.064
Old building	—	0.195	0.195

Range = 0.06 - 0.20
Overall average = 0.12

Source: Schulze (1975).

Table 39. Frequency Distribution of Formaldehyde Concentrations of Swedish Homes

Concentration interval (ppm)	Number of houses	Percent	Cumulative percent
Less than 0.30	72	22.6	22.6
0.30-0.39	60	18.8	41.4
0.40-0.69	100	31.3	72.7
0.70-0.99	49	15.4	88.1
More than 0.99	38	11.9	100.0

Average = 0.58 ppm

Total number of houses = 319

Source: Sundin (1978).

Table 40. Formaldehyde Concentration in Danish Homes

Room no.	House age (months)	Room volume (m^3)	P/V*	Temperature (°C)	Humidity (g/kg air)	Air changes (per hour)	HCHO concentration (ppm)
1	0.3	20	1.6	17.1	6.0	0.5	0.54
2	0.3	20	1.6	21.4	4.7	0.5	0.64
3	0.3	20	1.6	8.6	4.8	0.4	0.36
4	0.3	97	1.1	20.1	4.9	0.9	0.74
5	0.3	97	1.1	36.8	9.5	0.8	1.87
6	5.0	14	1.7	26.1	5.6	1.2	0.28
7	5.0	12	1.1	25.4	5.0	2.3	0.28
8	5.0	14	1.7	22.5	5.9	0.9	0.50
9	5.0	14	1.7	23.3	6.7	0.3	0.57
10	5.0	14	1.7	23.0	8.0	1.2	0.59
11	5.0	14	1.7	23.8	6.5	0.6	0.48
12	29	14	0.4	21.4	4.8	1.3	0.07
13	29	14	0.4	26.8	4.8	0.9	0.17
14	29	14	0.4	31.1	5.4	1.1	0.22
15	29	14	0.4	24.4	4.2	4.6	0.08
16	2	16	1.3	26.0	7.3	0.4	0.56
17	0.2	16	1.3	25.9	10.2	0.1	1.08
18	0.2	16	1.3	19.6	7.7	—	0.56
19	36	22	1.8	21.6	6.6	0.3	0.41
20	42	18	1.1	23.3	9.7	0.2	0.88
21	42	18	1.8	25.8	10.7	—	0.98
22	56	21	0.3	22.3	9.4	0.3	0.35
23	54	15	0.3	21.6	7.1	0.6	0.30
24	0.2	21	1.0	22.1	8.7	0.4	0.62
25	0.2	21	1.0	19.4	8.5	1.4	0.17

* Surface area of particleboard per net volume of room.

Source: Andersen et al. (1975)

According to the study documentation, houses sampled were selected at random. The only criterion was to include houses with different contents of particleboard. Particleboard was used as a construction material in 17 rooms and for fixtures only in eight rooms.

Samples were taken by drawing 50 liters of room air through two washing bottles. The laboratory analysis was carried out with the chromotropic acid test method with a reproducibility of $\pm$ 5 percent at 0.8 ppm and a detection limit of 0.08 ppm.

UK Study

Over a two and a half year period, the Building Research Station, England (Everett 1983) made over 2,000 measurements of formaldehyde levels in some 120 homes and 58 other buildings. The overall objective of the study was to compare measured formaldehyde levels in houses with and without urea formaldehyde foam insulation (UFFI), and to compare levels in houses before and after installation of UFFI. The buildings in the main survey and in the more detailed monitoring exercises were selected because of important design features, and included buildings where occupants had reported some discomfort as well as those where no complaint had been made.

The study provides results in the following areas:

- Outdoor formaldehyde levels
- Indoor formaldehyde levels, for
  - Buildings with uninsulated walls or insulants other than UFFI
  - Buildings with UFFI
  - Houses monitored before and after installation of UFFI
  - Houses of conventional (all masonry) construction
  - Houses of prefabricated concrete construction
- Formaldehyde levels in wall cavities

Unfortunately, sampling and analysis procedures used in the study were not described in the available documentation. Only a summary of the sampling results and a comparison to the results of the Canadian UFFI/ICC

study were provided in Everett (1983). These are presented in Table 41. The average outdoor concentration found across 60 sites, regarded by the author as typical for normal urban environments in the UK, was 0.006 ppm (std. dev. of 0.004 ppm).

4.5 Summary of Monitoring Data

Table 42 briefly summarizes the formaldehyde monitoring in residences reviewed within this section. It is divided between conventional and mobile homes for ease of comparison.

The studies performed by the Lawrence Berkeley Laboratory, the Consumer Product Safety Commission, the government of Canada, and researchers in Iowa and Indiana are the most recent studies of conventional homes and are not generally based on homeowner complaints. The age and construction of the home appear to be major determinants of the formaldehyde concentration. Newer, energy-efficient homes tend toward higher levels, likely due to the low air exchange rates of energy-efficient housing. Comparison of these data with data collected prior to 1980 indicates that there has been little change in conventional home formaldehyde levels since 1978.

Initial levels in new mobile homes are less well-defined, but appear to have declined in recent years. The Clayton study and the Wisconsin study, conducted prior to 1982, sought to define mobile home levels by age. The mean of an aggregated data set of these two studies (with nearly 1,200 observations - see Section 7.3) is 0.43 ppm, corresponding to a home age of less than one year. The more recent Texas study of formaldehyde levels in mobile homes of various ages found that levels in homes less than one year old averaged 0.21 ppm, and that the average levels were essentially the same for the 1-to-2, 2-to-3, and 3-to-4-year-old age groupings.

Table 41. Summary of U.S. Study and Comparison with Canadian UFFI/ICC Data

		Arithmetic mean, ppm	Standard deviation, ppm	Standard error
Candian data				
I.	Control (no UFFI)	0.034	0.029	0.0015
II.	100 problem houses	0.139	0.281	0.0281
III.	UFFI houses (UFFI/ICC files)	0.040	0.036	0.0014
IV.	UFFI houses (CHIP files)	0.054	0.044	0.0013
UK data				
	50 control buildings (no UF foam)	0.047	0.042	0.0020
	Foam insulated buildings	0.093	0.099	0.0026

Source: Everett (1983)

Table 42. Summary of Residential Formaldehyde Monitoring

Study/date(s)	Number of homes	Number of samples	Mean (ppm) or range of means	Range (ppm)	Comments
CONVENTIONAL HOMES					
Fleming & Associates New York Study (Traynor & Nitschke 1984)	30	--	0.040	0.007-0.151	Non-complaint homes.
Univ. Washington (1982-1983) (Breysse 1984)	59	113	--	<0.1 to > 1.0	Primarily complaint homes. Only 4 of 113 measurements >0.5 ppm.
LBL (1979-present) (Girman et al. 1983)	24 (EE) 16 (W)	--	--	<0.005 to 0.214 <0.005 to 0.079	Includes energy-efficient (EE) and older, weatherized (W) non-complaint homes around the U.S.
Geomet (1978) (Moschandreas et al. 1978)	17	~714	0.02 to 0.16 0.05 (overall)	<0.01 to 0.50	Includes conventional, "experimental", and apartment homes around U.S. Non-complaint homes. Assuming 60% of total aldehydes is formaldehyde.
Canadian UFFI/ICC (1981) (UFFI/ICC 1981)	378	--	0.034	<0.01 to ≤ 0.20	Study of UFFI and non-UFFI homes; mean is for non-UFFI homes. (Mean w/UFFI = 0.054 ppm for 1,897 homes).
ORNL/CPSC 40 Tennessee home Study (1982) (Hawthorne et al. 1984)	29	--	0.060	<0.025 to > 0.25	Study of UFFI and non-UFFI homes; mean is for non-UFFI homes. (Mean w/UFFI = 0.077 ppm for 11 homes).
Private Washington labs (1983) (Breysse 1984)	25 specified conventional	76	--	<0.05 to >0.5	None exceeded 1.0 ppm. 45 of 76 between 0.05 and 0.09 ppm.
UK study (~1980-1982) (Everett 1983)	50	--	0.047	--	Study was of UFFI and non-UFFI homes; mean is for non-UFFI homes. (Mean w/UFFI = 0.093 ppm).
Dutch study (1977-1980) (Van der Wal 1982)	36	--	--	0.032 to 1.444 (range of maximums)	Prior to control implementation. Largely complaint homes.
	5	--	--	0.048 to 0.602	After panel coating.

Table 42 (Continued)

Study/date(s)	Number of homes	Number of samples	Mean (ppm) or range of means	Range (ppm)	Comments
Iowa study (1980) (Schutte et al. 1982)	31	312	0.063	0.013 to 0.34	Non-complaint homes.
SAI California survey (1984)	6	—	0.084	0.046 to 0.153	New, non-complaint homes.
(SAI 1984)	64	—	0.050	0.018 to 0.120	Older, non-complaint homes.
Indiana Board of Health study (1979-1983) (Konopinski 1983)	120	—	0.09	ND to 1.35	Study of UFFI and non-UFFI homes; mean is for non-UFFI homes; includes some complaint homes. (Mean w/UFFI = 0.05 ppm for 119 homes).
Godish (1983)	29	—	0.05	0.03 to 0.07	Study of UFFI and non-UFFI homes; mean is for non-UFFI homes containing no particleboard flooring, cabinetry or paneling. (Mean w/UFFI = 0.07 ppm for 28 homes).
Cohn (1981)	103	—	0.027	—	
Swiss homes (1983) (Kuhn and Wanner 1984)	46	--	—	~0.1 to 0.7	Highest level prior to occupancy.
Netherlands study (1981-1982) (Cornet 1983 - Holland study)	52	—	0.048 to 0.055	—	Homes without particleboard, as measured by the chromotropic acid method.
German homes (1975) (Schulze 1975)	3	—	0.12	0.06 to 0.20	Few details available.
Swedish homes (1975-1977) (Sundin 1978)	319	—	0.58	0.1 to 2.0	Few details available.
Danish homes (1973) (Andersen et al. 1975)	23	--	1.44	0.07 to 1.87	Homes known to have particleboard construction materials.
MOBILE HOMES					
Geomet (1978) (Moschandreas et al. 1978)	2	84	0.21	0.07 to 0.46	Assuming 60% of total aldehydes is formaldehyde. Non-complaint homes.

Table 42. (Continued)

Study/date(s)	Number of homes	Number of samples	Mean (ppm) or range of means	Range (ppm)	Comments
Univ. Washington (1982-1983) (Breysse 1984)	430	822	—	<0.1 to >1.0	37 of 822 measurements >1.0 ppm. Complaint homes.
NHI (1984) (Conners 1984)	1	15	0.34	0.24 to 0.46	3-month old home built specifically for test.
Clayton (1980-1981) (Singh et al. 1982a)	259	--	0.62 (adjusted)	0.02 to 2.9 (adjusted)	Non-complaint, occupied and nonoccupied. Concentration by home age evaluated.
Wisconsin (1980) (Anderson et al. 1983)	137	920	0.38	0.02 to 2.26	Non-complaint, occupied homes. Concentration by home age evaluated.
Minnesota (1980-1981) (Stone et al. 1981)	109	--	0.61	--	Average home age <2 yrs. Complaint homes.
Tennessee (1982-1983) (Hodges 1984)	77	—	0.30	0.02 to 1.43	Complaint homes; no age data.
	55	—	0.23	0.02 to 1.92	Complaint homes, see Table 27 for data by home age.
Kentucky (1979-1980) (Conyers 1984)	103	—	0.43	0.01 to 1.99	Complaint homes, see Table 28 for data by home age.
Texas study (1982-1983) (Univ. Texas 1983)	121	—	0.18	0.04 to 0.35	Non-complaint homes. Excludes results from one county (El Paso) where evaporative coolers were in use.
SAI California survey (1984) (SAI 1984)	3	—	0.114	0.068 to 0.144	Passive LBL sampler; one week; non-complaint.

- Insufficient data in reviewed literature to report value.

ND = Not Detectable, or Below Detection Limit

5. SHORT- AND LONG-TERM EFFECTIVENESS OF FORMALDEHYDE CONTROL OPTIONS

This section discusses many of the control options that are promising for reduction of formaldehyde exposure in residential settings. Some options that have been evaluated or described by investigators are not discussed; among these are air cleaners (discussed by ADL Inc. 1981) and some esoteric chemical treatments and resins. This section describes the potential options, discusses the basis for the formaldehyde reduction effect, and presents available data on effectiveness.

In general, there are limited data demonstrating the effectiveness of the formaldehyde emission control options described in this report. Most of the available data concern only short-term effectiveness in reducing emissions of the residual free formaldehyde from boards. Virtually no information is available concerning the effectiveness of any technique in reducing formaldehyde emissions over months or years of product life. For those techniques investigated (see Table 43 for a summary of available information), the usefulness of the results is further limited by the absence of correlation between independent testing methods and conditions. The available data on control options effectiveness summarized in this section reflects only those data that were measured, not estimated. Any modeled or otherwise estimated values for control effectiveness were omitted from this summary.

5.1 Changes in UF Resin Formulation

Two major classes of control options fall under this category: variation of the ratio of formaldehyde to urea in UF resins, and adding chemicals to the resin/wood system to act as scavengers of excess formaldehyde, preventing its release.

Table 43. Summary of Data on Formaldehyde Emission Control Options*

Control option	Board type and thickness	Test type	Test results	Reference
Changes in UF Resin Formulation				
Reduction in FU Ratio				
U:F ratio in resin				
1:1.20	Particleboard	Perforator	0.025-0.035% released	Pizzi (1983)
1:1.30			0.04-0.05% released	
1:1.40			0.08-0.1% released	
1:1.65	MDF	Desiccator Perforator	1.6-8.4 ug/ml 80 mg/100g board	Meyer et al. (1983)
1:1.26		Desiccator Perforator	1.4 ug/ml 34 mg/100g board	
1:1.20		Desiccator Perforator	0.72 ug/ml 23 mg/100g board	
1:1.05		Desiccator Perforator	0.36 ug/ml 9.3 mg/100g board	
Formulation of Scavengers Into the Resin/Wood System				
Sodium sulfite scavengers	Plywood	Desiccator	0.00 ppm (100% improvement) 0.25 ppm (98% improvement) 0.34 ppm (97% improvement) 0.26 ppm (98% improvement)	Meyer (1979)
Urea scavengers (unspecified)	Plywood	Desiccator	0.07 ppm (99% improvement)	Meyer (1979)
Kenosize FR4514 urea scavenger	22 mm particleboard	Perforator WK1 (modifed) Swedish chamber after 5 months	12 mg/100g 91 mg/m^2 0.29 ppm 0.45 ppm	Johansson (1982)

Table 43. Summary of Data on Formaldehyde Emission Control Options* (continued)

Control option	Board type and thickness	Test type	Test results	Reference
Post-Cure Board Treatments				
<u>Ammonia Fumigation</u>				
Verkor FD-EX	Particleboard	Perforator	5.5 mg/100 gr. board (97% improvement) 4.4 mg/100 gr. board (86% improvement).	Simon (1980)
	Plywood	Desiccator	0.15 ppm (99% improvement) 0.37 ppm (97% improvement) 0.01 ppm (100% improvement) 0.64 ppm (95% improvement)	
RYAB	Particleboard			Johansson (1982)
	10 mm	Swedish chamber	0.14-0.39 ppm	
	19 mm		<0.1 ppm	
	22 mm		0.71 ppm	
	10 mm	Perforator	11 mg/100 g.	
	19 mm		13 mg/100 g.	
	22 mm		19 mg/100 g.	
	10 mm	WKI (modified)	199 mg/m^2	
	19 mm		110 mg/m^2	
	22 mm		166 mg/m^2	

Table 43. Summary of Data on Formaldehyde Emission Control Options* (continued)

Control option	Board type and thickness	Test type	Test results	Reference
Swedspan	Particleboard			Johansson (1982)
	10 mm	Swedish chamber	<0.1-0.32 ppm	
	19mm		0.25 ppm	
	22 mm		0.22 ppm	
	10 mm	Perforator	15 mg/100 g.	
	19 mm		14 mg/100 g.	
	22 mm		14 mg/100 g.	
	10 mm	WKI (modified)	166 mg/100 g.	
	19 mm		79 mg/100 g.	
	22 mm		113 mg/100 g.	
	Particleboard			Swedspan (undated)
	10 mm	Perforator	9 mg/100 g.	
	28 mm		12 mg/100 g.	
	36 mm		8 mg/100 g.	
	8 mo. after production			
	10 mm	Perforator	5 mg/100 g.	
	28 mm		13 mg/100 g.	
	36 mm		6 mg/100 g.	
Weyerhauser in-home fumigation	N/A	Indoor air levels	0.07-0.26 ppm (up to 85% reduction)	Jewell (1982)

Table 43. Summary of Data on Formaldehyde Emission Control Options* (continued)

Control option	Board type and thickness	Test type	Test results	Reference
Non-scavenger emission barriers				
Melamine-containing surface coating	Particleboard	Perforator	0.02 ppm (98% improvement) 0.10 ppm (92% improvement)	Meyer (1979)
Falima-F (coating)	Particleboard	Dynamic Chamber (emission rate)	0.03 ppm (97% improvement) 25.4-70.8 ug/m^2/hr	Molhave (1983)
Valspar 50100	Plywood	Dynamic Chamber	0.3 ppm (90% improvement)	Myers (1982b)
Nitrocellulose surface coating	(Unspecified)	Desiccator	0.09 ppm (92% improvement) 0.02 ppm (98% improvement)	ICF (1984)
Polyurethane surface coating	Plywood	JIS Desiccator	0.5 ppm (96% improvement)	Meyer (1979)
Wall paper	(Unspecified)	(Unspecified)	33% improvement	Meyer (1979)
Nacore overlay	(Unspecified)	(Unspecified)	84% improvement	
Varnish	(Unspecified)	(Unspecified)	92-98% improvement	
Overlay paper	(Unspecified)	(Unspecified)	93% improvement	
Decorative vinyl overlay	Plywood paneling	Large chamber 2-hr desiccator	0.04 - 0.75 ppm 0.53 - 3.04 ug/ml	Groah (1984)

Table 43. · Summary of Data on Formaldehyde Emission Control Options* (continued)

Control option	Board type and thickness	Test type	Test results	Reference
Substitute Resins				
Phenol-formaldehyde resin adhesives	Pine plywood	2-hr desiccator Dynamic chamber	0.08-0.34 g/ml 0.011-0.04 ppm	APA (1984)
	Fir plywood	2-hr desiccator Dynamic chamber	0.18 g/ml 0.017-0.05 ppm	
	Oriented strand board	2-hr desiccator Dynamic chamber	0.02-0.14 g/ml 0.03-0.07 ppm	
	Particleboard	2-hr desiccator Dynamic chamber	0.15-0.51 g/ml 0.01-0.08 ppm	
	Waferboard	2-hr desiccator Dynamic chamber	0.03-0.18 g/ml 0.01-0.08 ppm	
	Particleboard		3.85 x 10^{-6} mg/ml	Meyer (1981)
	Fir plywood		2.50 x 10^{-6} mg/ml	
	Fir/hemlock plywood		1.30 x 10^{-6} mg/ml	
	Waterboard		1.45 x 10^{-6} mg/ml	
	Pine plywood		1.35 x 10^{-6} mg/ml	
Other Controls				
Board aging				
30 days	Hardwood plywood	Dynamic chamber	0.31 ppm (90% improvement)	Myers (1982b)
60 days			<0.01 ppm (100% improvement)	
15 days		24-Hr desiccator	93% improvement	

*Further details on testing methods, conditions, and results can be found in the following text in this section and in the references cited. Many references cited are secondary sources.

5.1.1 Reduction in the F:U Ratio

Resin formulation has changed dramatically over the past decade. In the 1970s, a resin molar ratio of 2.0 parts formaldehyde per part urea was not uncommon; current (i.e., mid-1984) mole ratios commonly reported range from 1.15 to 1.3 for particleboard, 1.2 to 1.5 for hardwood plywood paneling, and in the vicinity of 1.65 for MDF (HPMA 1984, NPA 1984, Podall 1984). This change has occurred, at least in part, in response to the public's and regulatory agencies' concern about formaldehyde exposure. Lessening the amount of formaldehyde in a pressed-wood product, as through this measure, is effective in the short term in reducing formaldehyde release. The long-term stability of these low ratio resins has, however, been questioned; it is possible that even though initial emission rates may be dramatically lower, a similar amount or even more formaldehyde could be released over the life of a low F:U ratio board, via increased hydrolysis, than from a board made with more conventional resin formulations (Swedish Particleboard Association, Dr. Gfeller (Novopan AG), and Dr. Roffael (Wilhelm Klauditz Institute) as reported by Gaudert et al. 1983). Roffael (1978) states that the water solubility of cured resins increases with a decrease in F:U. Meyer (1984) demonstrated a slight increase in emission rate from a low mole ratio particleboard over a three-year test period; experts agree, however, that long-term effectiveness and emissions characteristics are not known for this control (Meyer 1984, Gaudert et al. 1983).

Myers (1984a) points out that mole ratio affects not only the rate and magnitude of formaldehyde release from pressed-wood products but is also a major determinant of the structural properties of the product. His literature review concluded that direct correlation of board properties with UF resin formulation was not possible with existing data, as there was too much variation between investigators' test methods and materials. He did present the following criteria as limitations on the F:U ratio in particleboard:

F:U must be less than 1.1 to 1.2 to meet the West German formaldehyde emission standard for E-1 particleboard (i.e., less than 0.1 ppm in a specified chamber test).

F:U must be less than 1.2 to 1.3 to meet the HUD emission standard for particleboard in mobile homes (i.e., 0.3 ppm in a chamber test).

F:U must be more than 1.2 to provide sufficient bending strength.

F:U must be more than 1.1 to 1.2 for internal strength of bonds.

F:U must be more than 1.2 to 1.3 for control of thickness swell.

These conflicting standards illustrate the difficulty with using low mole ratio resins to meet formaldehyde emission standards.

The mole ratio of the UF resin is correlated with the free formaldehyde content of pressed-wood products, which is a determinant of the emission characteristics of the product at least during the early part of the product life. Myers (1984a) collected data from numerous published studies and produced a correlation with a wide range of values around the curve. This is reflective of the other parameters affecting free formaldehyde content (press time and temperature, amount of catalyst, and other manufacturing variables).

Myers also states that F:U ratio is a determinant of the hydrolytic stability of resin bonds, thus affecting another potential mechanism of formaldehyde release (Myers 1982a). He measured the hydrolysis rates of two resins of different mole ratios, keeping other parameters constant. He found that resin variation strongly affected hydrolysis rate and consequent formaldehyde release when the cure was more complete (higher press time and temperature), and that at less-complete cure conditions resin mole ratio did not affect hydrolysis. The resin with a mole ratio of 2.0 exhibited more hydrolysis over the test period than did the resin of mole ratio 1.4. The usefulness of these results is somewhat

diminished by the fact that this test was performed on resin only; no wood was incorporated, so that the object was not a pressed-wood product. In addition, the low mole ratio resin used (1.4) is at the upper end of the range of low mole ratio resins used commercially during 1984. Furthermore, Meyer et al. (1983) have reported results somewhat contradictory to this theory on the effect of molar ratio.

The constraints listed previously limit the lowering of mole ratio much below 1.2 for particleboard resins. The NPA states that current mole ratios in particleboard range from 1.15 to 1.25 (NPA 1984), though the USDA reports that resins with an F:U ratio of 1.05 have been tested that do not cause excessive sacrifice in product properties (USDA 1984).

The data that have been reported for the use of low mole ratio resins in MDF indicate that low mole ratios do diminish emissions; no data on board properties are, however, available. It has been stated (NPA 1984) that MDF is more sensitive to low mole ratio UF resin than is particleboard, and that successful formulations require a ratio of 1.2 to 1.4. HPMA (1984) states that mole ratio resins as low as 1.2 to 1.4 are being used in the manufacture of hardwood plywood paneling.

The results of emission characterization surveys conducted by NPA indicate that initial emissions (i.e., dessicator results for fresh boards) from mobile home decking, particleboard underlayment, and industrial particleboard decreased, or the average, by 67 percent, 56 percent, and 60 percent, respectively between 1980 and 1982 (NPA 1984). Reduction in F:U resin mole rations was cited as being responsible for most of the observed decrease in emissions. NPA is currently conducting its 1984 emission survey (see Section 2.5).

The improvement in formaldehyde emission is accomplished by varying the amount of excess formaldehyde. The results of two such studies are presented in Tables 44 and 45. Meyer et al. (1983) tested seven adhesives by manufacturing MDF and measuring resultant formaldehyde

Table 44. Comparison of Formaldehyde Emission from Particleboard Prepared with UF Resins of Different Molar Ratios

U/F molar ratio	Percent CH_2O released (perforator method) (mg/100g)
1:1.4 - 1.5	80 - 100
1:1.3 - 1.35	40 - 50
1:1.2 - 1.25	25 - 35

Source: Pizzi (1983).

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Table 45. Comparison of Formaldehyde Emission from MDF Prepared with UF Resins of Different Molar Ratios

Resin adhesive[a] F:U	2-Hr desiccator value[b] (micrograms/ml)				Perforator (mg/100g)
	3 Days	6 Weeks	5 Mos.	10 Mos.[f]	6 Weeks
(1:1.85)[c]	—	8.4	—	4.4	—
(1:1.65)[d]	4.8	2.3	2.0	1.9	68
(1:1:65)[e]	5.6	3.0	2.3	2.0	80
(1:1.65)[e]	2.6	1.6	0.86	0.70	—
(1:1.26)[e]	2.5	1.4	0.85	0.71	34
(1:1.20)[e]	1.4	0.72	0.62	0.59	23
(1:1.05)[e]	0.54	0.36	0.38	0.40	9.3

a Boards were manufactured with dimensions 0.9 m x 0.9 m x 16 mm. Kenosize wax dispersion (contains a scavenger) was added to each resin at 1 wt %.

b Board sample edges were not sealed. Average values for at least three boards for each resin type.

c Commercial domestic resins.

d Lab-made resin

e Commercial European resins

f Samples cut one day before testing from center of boards.

Source: Meyer and Hermanns (1984a); Meyer et al. (1983); Meyer et al. (1984).

emissions. Two of the resins were commercial UF preparations with a mole ratio of 1.65. One resin was a UF-urea scavenger mixture, also with a mole ratio of 1.65. The four remaining resins were manufactured in Sweden, and had mole ratios of 1.65, 1.26, 1.20, and 1.05. As seen in Table 45, emissions declined with declining F:U ratio. The structural properties of these boards were not reported.

Data are also available on the combined effect of varying F:U molar ratios in resin adhesives with other formaldehyde emission control options discussed in this report. These data are presented in Tables 46 and 47.

5.1.2 Formulation of Scavengers into the UF Resin/Wood System

Reactive chemicals, which will react with excess or free formaldehyde present in pressed-wood products, can be added to the resin formulation or the wood or one of the fillers (like wax) prior to cure. These chemicals are often sulfurous or nitrogenous compounds that form stable complexes with formaldehyde in the resin. Other carconaceous compounds, such as resorcinol derivatives (Dietrick and Terbilcox 1983) may also be effective.

Champion International, Union Camp, the Potlatch Corp., the NPA, and other industry representatives provided the EPA with comments regarding this proposed control option, discussing both the effectiveness and costs (economic and in terms of reduced properties). These comments and the review report of Myers (1984d) state that scavengers can be effective but add cost and can be deleterious to the finished product. The most popular scavenger is urea, added as an aqueous solution or as a dry compound. The effect of this action is the same as a variation in the formaldehyde:urea ratio, in terms of lessening emissions as well as reducing strength (Champion 1984, Potlatch 1984). An overaddition of scavenger like urea can prohibit the proper cure of the resin during

Table 46. Combined Effect of Aging and Varying Molar Ratios in Adhesives on Formaldehyde Emissions from Particleboard

Test (Units)	Mole ratio F:U	Aging condition (temp./RH)	Aging time	Test values
WKI (mg/100 g)	1.27	20°C/65%	1 day	83
			6 weeks	49
	1.55		1 day	127
			6 weeks	78
	1.40		7 weeks	80
			15 months	48
	1.60		7 weeks	139
			15 months	60
	1.80		7 weeks	175
			15 months	72
Perforator (mg/100 g)	1.6	Probably 20°C/65%	0	100
			8 days	73
	1.8		0	125
			8 days	85
2-Hour desiccator (ug/mL)	1.0	Probably ambient	1 day	0.8
			15 days	0.4
	1.2		1 day	1.4
			15 days	0.8
	1.3		1 day	3.0
			15 days	1.7
	1.6		1 day	8.0
			15 days	4.3

Source: Myers (1984a).

Table 47. Combined Effect of Press Temperature/Time and Varying Molar Ratios in Adhesives on Formaldehyde Emissions from Particleboard

Mole ratio F:U	Varying press temperature: Press temperature/time (°C/min.)	Varying press temperature: Perforator value (mg/100g)	Varying Press Time: Press temperature/time (°C/min.)	Varying Press Time: Perforator value (mg/100g)
1.40	140/8	48	180/5	41
	180/8	37	180/8	40
1.60	140/8	85	180/5	80
	180/8	65	180/8	70
1.80	140/8	157	180/5	150
	180/8	110	180/8	110
1.27	180/3.2	27	220/2.1	26
	220/3.2	18	220/3.2	18
1.50	180/3.2	59	220/2.1	57
	220/3.2	40	220/3.2	40
1.25			170/2.5	28
			170/4.2	17
1.37			170/2.5	47
			170/4.2	30
1.53			170/2.5	84
			170/4.2	55

Source: Myers (1984a).

press; any formaldehyde scavenger can interfere to some extent with proper curing by removing essential formaldehyde. The Hardwood Plywood Manufacturers Association states that scavengers are less effective when used with low molar ratio resins than with more conventional resin formulations (HPMA 1984), evidently because there is less free formaldehyde for the scavenger to react with when the resin has a low F:U ratio.

The National Particleboard Association (NPA) discusses the use of ammonium compounds, and adds that inclusion of the scavenger as a component in the wax sizing is probably the most effective method of use in particleboard manufacture; Myers (1984d) also concluded that this may be the most effective scavenger technique. NPA lists the following as useful scavengers: urea, protein, lignosulfonates, and ammonium carbonate. The usefulness of a urea in wax formulation is confirmed by reports that European manufacturers (Casco and BASF) have produced boards that meet emission standards by using this control (Gaudert et al. 1983, Myers 1984d).

Johansson (1982) evaluated the effectiveness of Kenosize FR 4514, a urea scavenger that is formulated into the wax added to the resin/wood system. Use of the Kenosize reduced emissions, as measured by the perforator method, from 26 mg/100g (control board) to 12 mg/100g (treated board). Even though resin weight, as a percent of the board, must be increased somewhat with the use of Kenosize (Shields and Serveau 1983), this control was found to be the most effective of the four reviewed by Johansson (two ammonia treatments, a low molar ratio resin, and the scavenger). Long-term effectiveness of this control was measured by chamber tests. Johansson reported that measurements five months after treatment showed increased emissions (0.45 ppm) over emissions immediately after treatment (0.29 ppm).

Myers (1984d) summarized the rather limited information available on the effectiveness of scavenger additions to wood furnish or veneer. He divided the various treatment techniques reported in the literature into

four basic approaches: (1) addition of lignocellulosic materials impregnated with scavengers (e.g., melamine and urea) to the furnish before resin addition; (2) spraying furnish or veneer with simple formaldehyde-reactive chemicals (e.g., ammonium carbonate, ammonium lignosulfonate) before of after resin addition; (3) spraying furnish with an aqueous wax or polymer dispersion containing urea; and (4) using a urea scavenger in conjunction with a non-UF adhesive in the middle layer of the furnish.

Table 48 summarizes the results of the various studies reported in the literature for these four approaches. The data for Approach 3 compare well with the data of Johansson on Kenosize scavenger discussed above. Although critical evaluation of the results was difficult because of the limited amount of information, Myers concluded that the use of scavengers, in conjunction with resins having F:U mole ratios of about 1.4, can lower the formaldehyde emission of boards by about 50 to 70 percent, although often at some sacrifice in the physical properties of the boards. Thus, other complementary measures may be needed to provide additional reduction in emission while maintaining or even improving physical properties.

This option is obviously potentially useful as a method of achieving short-term reductions in formaldehyde emissions, as can be seen in Tables 48 and 49. Johansson (1982) alludes to the longer-term effectiveness of this control, and her data are promising. Sundin (1985) has measured formaldehyde levels in a home with Kenosize-treated particleboard as its only pressed wood product. The loading rate of particleboard in the home is 1.1 m^2/m^3. The highest level measured in the home was found immediately after construction, and was around 0.15 ppm. The home has been monitored six times in the five years since construction, and only once did the level exceed 0.10 ppm. A sample of the particleboard was removed from the home in 1982 (three years after it was built) and emissions measured via the perforator method. Sundin reports a very low

Table 48. Formaldehyde Emissions from Boards Formulated with Scavengers

Approach	Illustrative board variations[a]	CH_2O value (percent of control)	Data adequacy[b]
1	5% of middle layer furnish with 60% melamine; F/U 1.5	5.0 cc[c] (53)	Low. Few details. Significance of CH_2O test not clear.
	F/U 1.4; 5% fiber; ~2% urea	<10 mg/100 g[d] (<25)	High
	F/U 1.4; 14% fiber; 4% urea	<10 mg/100[d] (<25)	
	F/U 1/4; 2% of urea-treated paper fibers; 0.8% urea	20 mg/100 g[d] (29)	High
2	Ammonium acetate "impregnated veneer"	2.6 ug/ml[e] (--)	Low-medium
	$NaHSO_3$ "impregnated veneer"	0.5 ug/ml (--)	
	Urea-sprayed veneer; 3-ply plywood	~0.5 ug/ml (--)	
	6.5% UF resin; ~ 1.2% lignosulfonate on furnish	13 mg/100 g[g] (50)	Low
	F/U 1/4; 34% $(NH_4)_2CO_3$	20 mg/100[d] (62)	High
	F/U 1.4; 0.67% $(NH_4)_2CO_3$	12 mg/100g (37)	
	F/U 1.2; 0.34% $(NH_4)_2CO_3$	9.5 mg/100 g (58)	
	F/U 1/2; 0.67% $(NH_4)_2CO_3$	7.0 mg/100 g (43)	

Table 48. (continued)

Approach	Illustrative board variations[a]	CH_2O value (percent of control)	Data adequacy[b]
3	~4% of seven different dispersions on furnish; 1 4% urea	10 to 20 mg 100[d] (25 to 50)	High
	F/U 1.4; ~3% of one dispersion system; 1% urea	20 mg/100 g[d] (50)	
	F/U 1.4; ~4% of two dispersions; 0.8% urea	20 mg/1 100 g[d] (29)	
	F/U 1.2; 1/2% Kenosize dispersion in middle layer and 0.67% in surface layer	12 mg/100 g[d] (46)	Medium
	~1% Kenosize dispersion; control perforator ~ 31 mg/100 g	12 mg/100 g[d] (40)	Low
	~1% Kenosize dispersion; control perforator ~15 mg/100g	8 mg/100 g[d] (50)	
4	3% isocyanate and 3% urea in middle layer. F/U 1.4	15 mg/100 g[d] (54)	High
	Middle layer with 2% isocyanate and 2% urea. F/U 1.4	14 mg/100 [d,h]	

[a]Concentrations based on dry furnish unless stated otherwise. UF = urea-formaldehyde, MUF = melamine urea-formaldehyde, F/U = formaldehyde-to-urea mole ratio.
[b]Subjective judgment by Myers (1984d). H = high, M = medium, L = low.
[c]Sample in sealed container 2 hour, 70°C, 50% relative humidity. Formaldehyde and air purged, collected, analyzed.
[d]Perforator test. Analysis of formaldehyde removed by 2 hours in boiling toluene.
[e]Japanese dessicator test1. Measure of formaldehyde transferred from boards through air into dish of water, all within a sealed vessel.
[f]"Bonding strength" after 3 hours of water immersion, 60°C.
[g]Ground sample, 100°C, 3 hours. Evolved formaldehyde trapped in cold water.
[h]Control is board with same isocyanate and no urea.
Source: Myers (1984d).

Table 49. Effect of Several Pre-Press Scavengers on Formaldehyde Emissions from Plywood

Scavenger	CH_2O Emission desiccator method (ppm)	Percent improvement
Control	13.20	-
Urea (CON_2H_4)	0.07	99
Ammonium sulfite ($(NH_4)_2SO_3$)	0.15	99
Sodium sulfite (Na_2SO_3)	0.00	100
Sodium bisulfite ($NaHSO_3$)	0.25	98
Sodium hydrosulfite ($Na_2S_2O_4$)	0.34	97
Sodium metabisulfite ($Na_2S_2O_5$)	0.26	98
Ammonium bicarbonate (NH_4HCO_3)	0.37	97
Ammonium thiosulfate ($(NH_4)_2S_2O_3$)	0.01	100
Ammonium sulfamate ($NH_4OSO_2NH_2$)	0.64	95

Source: Meyer (1979).

perforator value of 8.6 mg/100g for that sample. Sundin (1985) also reports a 22-week study in a manufacturing plant using Kenosize FR 4514 in a 1.2 mole ratio resin. Emissions, measured by the Swedish chamber test, steadily declined from 0.14 mg/m^3 at 23°C/50% RH to 0.08 mg/m^3 over the test period.

One non-industry commenter on EPA's 4(f) rule stated concern over emission of reaction products or the scavenger itself. The reaction product of formaldehyde and a urea scavenger is hexamethylenetetramine; the stability of that compound over a matter of years is questionable. The exact reaction products of formaldehyde and other scavengers varies, but there exists the possibility that they could break down and emit formaldehyde over extended periods of use.

5.2 Post-Cure Board Treatments

5.2.1 Ammonia Fumigation

Several researchers have evaluated the use of ammonia fumigation as a control for formaldehyde emissions from pressed-wood products. There are numerous permutations of the ammonia fumigation process; variations exist in the ammonia concentration, the method of application, and the duration of treatment. Few data exist, however, to quantify the effectiveness of this option in controlling emissions over the long term, regardless of the actual fumigation process used. Very few data on the relative effectiveness of this control on different wood products are available, though existing data do pertain to a wide variety of products. Only medium-density fiberboard appears never to have been tested individually.

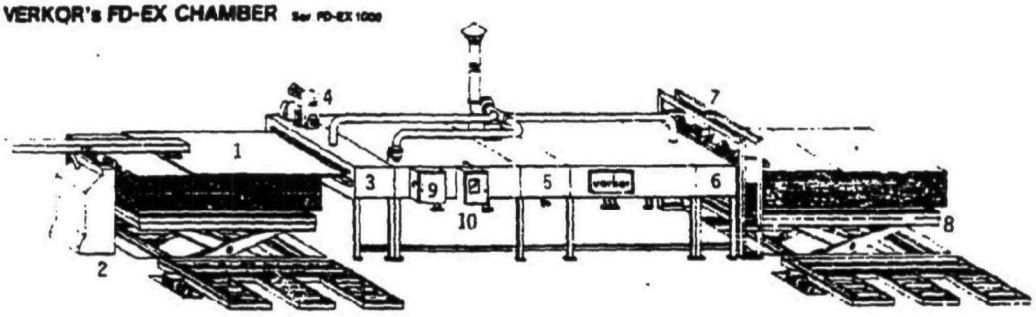
The fumigation process can be performed either on manufactured pressed-wood products or on entire homes containing those products. The National Particleboard Association (NPA 1984) states that, in this country, ammonia fumigation is used primarily as a retroactive treatment for complaint homes, largely in mobile homes. The production line

processes, the Verkor, Swedspan, and RY AB methods, are used primarily in Europe; no domestic manufacturer mentions their use in U.S. plants. Regardless of whether the process is intended for control in homes or in production, the basis of the control option remains the same. Ammonia is introduced to the wood product as a gas, which binds with the free formaldehyde present in the wood, forming hexamethylenetetramine (Smith 1983, Simon 1980).

The Verkor FD-EX method, as described by Simon (1980), is intended for large-scale production applications. Figure 20 illustrates the treatment apparatus. It is designed specifically to control free formaldehyde, and Simon claims a permanent reduction in formaldehyde emission. The fumigation takes place in a series of two carefully-controlled, sealed chambers. In the first, ammonia is introduced as a gas; the concentration of ammonia is determined by the mass of wood to be treated, the formaldehyde:urea ratio of the resin in the wood, and the volume and residence time in the chamber. Residence time ranges from 4.5 minutes for thin particleboard to over 10 minutes for hardwood plywood. The second chamber is used to eliminate free ammonia from the surface layers of the boards by employing controlled ventilation (Simon 1980). Some excess ammonia is left in the boards to allow continued scavenging of formaldehyde, although formic acid is added after ventilation to neutralize some ammonia and reportedly to reduce the chance of future hydrolysis (ICF 1984).

Figure 21 presents the measured effectiveness of the Verkor method. Very high short-term effectiveness levels (in terms of absolute reduction over uncontrolled boards, as a percent) were reported by Simon (1980). He measured the effectiveness of the treatment immediately following ammonia application and up to three years later. That three-year later measurement is apparently the basis for the permanent reduction claim, though comments by the NPA (NPA 1984) state that the reduction may not be

VERKOR's FD-EX CHAMBER



1. Particle boards.
2. Feeder.
3. Inlet chamber.
4. Inside transportation system.
5. VERKOR's FD-EX CHAMBER.
6. Outlet chamber.
7. PERFORATOR GRADE stamping roll.
8. Stacker
9. Control panel.
10. Automatic analyser-controller.

Figure 20. The Verkor FD-EX Chamber

Source: Simon (1980).

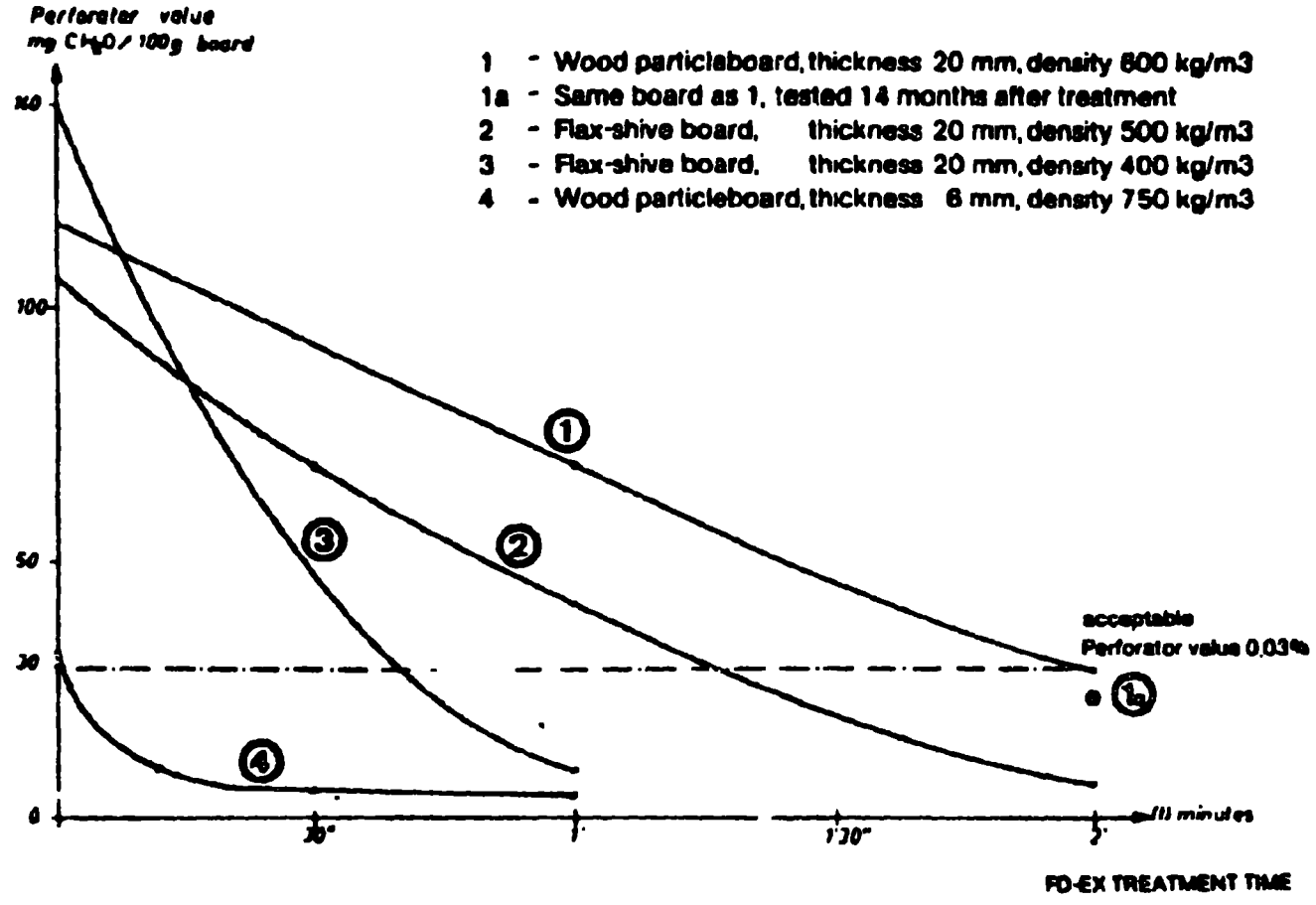


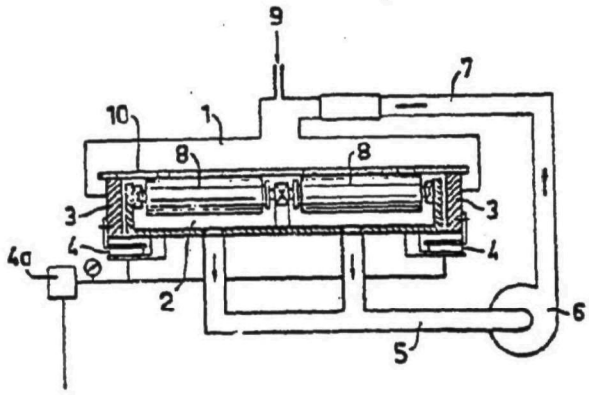
Figure 21. Effectiveness of FD/EX Treatment

Source: Simon (1980).

permanent. Simon's study was extremely limited; only four boards were tested, and retesting was conducted only a few times (seven data points were reported) over the three-year period of the experiment. Results are therefore far from overwhelming.

Little information is available on the RYAB method of fumigating wood products with ammonia, a method known to be used in Finland (Gaudert et al. 1983) and Sweden (ICF 1984). That method employs pressure to introduce ammonia into the boards (see Figure 22); the ammonia therefore enters deeper into the board, scavenging a greater proportion of the formaldehyde present. A pressure differential of 5.8 to 13 psi is employed (ICF 1984). Quantitative efficiency data are available from Johansson (1982). She evaluated the RYAB treatment by comparing emissions, measured by the perforator test, from treated board to emissions from control boards. Table 50 lists these data, along with data for Swedspan treatment efficiency.

The ASSI method mentioned by Smith (1983) is the Swedspan method, a production-stage fumigation method. This method involves spraying of ammonia compounds (carbonates, bicarbonates, sulfates, or acetates) between boards as they are stacked after production (see Figure 23). Smith reports a 60 percent reduction in emissions 44 weeks after treatment, with a reduction of up to 88 percent immediately following spraying. This method may not be expected to be as effective as the Verkor or RYAB methods, mainly because the treatment is more surficial. The data in Table 50 do not bear this out, however. In direct comparison, Swedspan was more effective in controlling emissions. Neither the RYAB nor the Swedspan method was as effective in the long term (5 month chamber tests) as in the short term, as measured by perforator emissions. Additional data on the efficiency of the Swedspan method are presented in Table 51. This manufacturer claims continued effectiveness of up to 89 percent after 5 months and 75 percent after 8



1 = *Upper chamber*
2 = *Low pressure zone*
3 = *Frame*
4 = *Pneumatic rolls*
4a= *Compressor*
5 = 7 = *Injection*
6 = *Vacuum pump*
8 = *Rolls*
9 = *Injection of ammonia*
10 = *Particleboard*

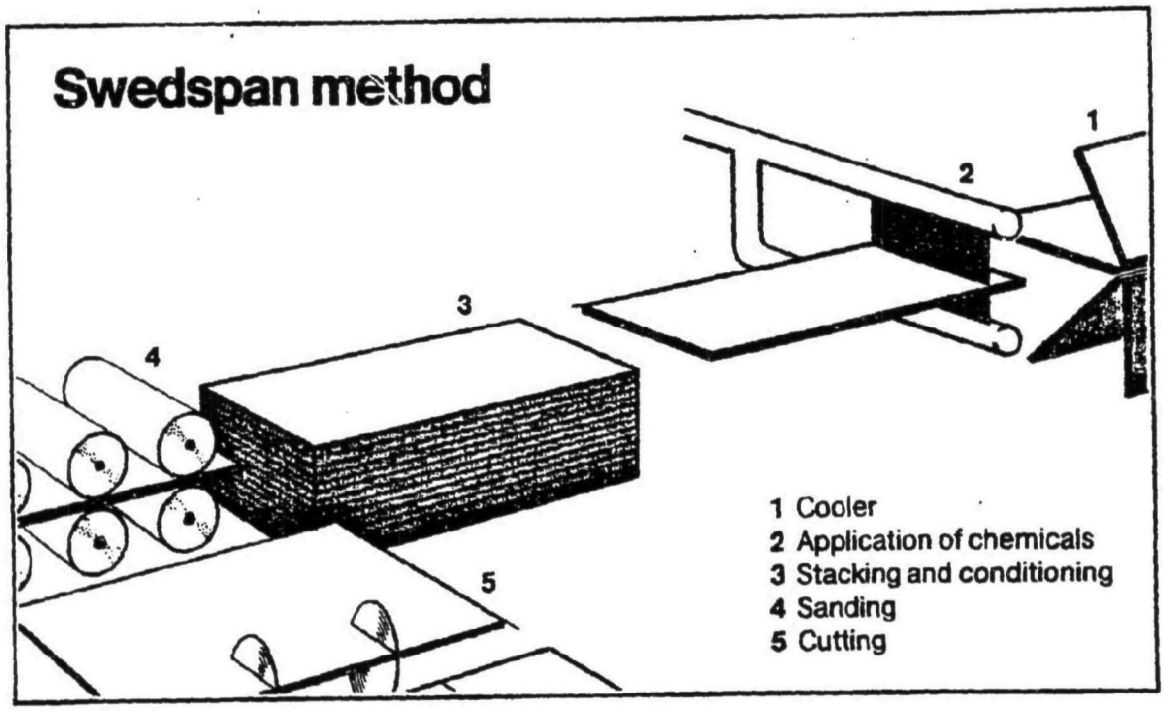
Source: Jewell (1982).

Figure 22. RYAB's Gassing Equipment

Table 50. Effectiveness of RYAB and Swedspan Ammonia Fumigation of Boards

Property	Boards manufactured by RYAB			Boards manufactured by Swedspan		
	RYAB method	Swedspan method	Control	RYAB method	Swedspan method	Control
Chamber emissions, ppm						
immed after manufacture	0.35	0.66	1.40	—	—	—
5 mo. after manufacture	0.48	0.69	0.98	—	—	—
Perforator Value, mg/100 g particleboard						
22 mm board	19	14	26	—	--	—
19 mm board	—	—	--	13	'4	28
10 mm board	10	9	27	11	15	36
MKI (modified) results mg/m^2, 24 hr						
22 mm board	166	113	178	--	—	—
19 mm board	—	--	—	110	79	214

Source: Johansson (1982)



Source: Swedspan (undated).

Figure 23. The Swedspan Ammonia Treatment Model

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Table 51. Effectiveness of Swedspan Method for Formaldehyde Emission Reduction

	Board thickness (type unspecified)		
	10 mm	28 mm	36 mm
One week after manufacture	Perforator values (mg/100g)		
Untreated	25	29	21
Treated	9	12	8
Percent reduction	64	59	62
8 months after manufacture			
Untreated	20	28	20
Treated	5	13	6
Percent Reduction	75	55	70

Source: Swedspan (undated)

months. The best emission reduction is at the surface, but an initial reduction of 50 percent in formaldehyde emission potential from the core of a treated 28 mm board has been measured by Swedspan (undated). No loss of properties is said to occur as a result of treatment. All ammonia treatments are potentially applicable to composition boards and plywood.

Available data on in-home treatment with ammonia are relatively limited. Some work has been done by Weyerhauser, as reported in Jewell (1980a, 1982) and in their comments on the 4(f) notice. In addition, Smith (1983) evaluated the efficiency of ammonia treatment of a four-year-old trailer used as an office building.

The method of in-home ammonia treatment as described by Jewell (1982) and Weyerhauser (1984) involves vacating the residence, then placing pans of 29 percent ammonia throughout the dwelling. The home is sealed, and the indoor temperature is raised to 80 degrees Fahrenheit to increase the rate of vaporization. The fumigation is allowed to continue for at least 12 hours; at that time, the home can be thoroughly ventilated and reoccupied. The Manufactured Housing Institute (MHI 1984) cites an effectiveness of 70 percent for reduction of initial emissions. Weyerhauser (1984) reports an average initial reduction of indoor formaldehyde levels by 75 percent in 12 ammonia-treated mobile homes (treatment performed in 1979 and 1980); however, air sampling performed several weeks after treatment in five of the mobile homes indicated that formaldehyde air levels were increasing (although not to the pre-treatment levels). Table 52 presents these data.

Jewell (1982) has also studied the effectiveness of ammonia treatment of boards (both lauan plywood and 5/8 inch particleboard). The large initial effectiveness of his fumigation treatment is not seen 10 weeks after treatment, when emissions from treated boards are only slightly less than emissions from control boards.

Table 52. Results of Ammonia Fumigations of 12 Mobile Homes[1]

Type of Mobile Home	Location	Before Fum. HCHO (uL/L)	Weeks After Ammonia Treatment HCHO (uL/L)												
			0	5	10	15	20	25	30	35	40	45	50	55	60
Single-W	Alabama	1.0	0.26*	0.26		0.24		0.28*							0.19
Double-W	Wash.	1.1	0.17*	0.23	0.20										
Single-W	Wash.	0.71*	0.07		0.17		0.19*				0.30*				
Single-W	Wash.	0.89	0.13*	0.27*							0.42*				
Single-W	Kentucky	0.65	0.23*												
Single-W	Florida	0.41	0.11												
Single-W	Wash.	0.93	0.16*	0.37*		0.36*						0.57*			
Single-W Office	Illinois	0.56	0.31*				0.25*								
Double-W	Oregon	0.88	0.20*												
Single-W	Oregon	1.0	0.13												
Double-W	Florida	0.51	0.18*												
Double-W	Florida	0.41	0.12												

[1] Formaldehyde measurements made using a modified NIOSH chromotropic acid method.

* Data temperature corrected using the mathematical model of Berge, et.al. (1980).

W = Wide

The study by Smith actually simulated in-home fumigation rather than being a full-scale field study. He identified the highest formaldehyde emitter in the mobile homes that had been converted into offices, then removed that source (paneling) and fumigated the panels with ammonia. He was able to correlate emission reduction with both ammonia concentration and duration of exposure to the ammonia. This study obtained formaldehyde emission data for a period of 48 days following treatment and observed no reduction in effectiveness. Smith's 1983 thesis does, however, raise the question of the long-term effectiveness of the treatment, citing the potential for uncontrolled resin hydrolysis to evolve formaldehyde.

The effectiveness of this control option, both in the short and long term, is a function of the chemistry of the ammonia-formaldehyde reaction and the characteristics of the remaining formaldehyde in the resin. Neither of those parameters is particularly well-characterized, necessitating speculation regarding their importance.

As stated previously, the reaction between ammonia and formaldehyde results in the formation of hexamethylenetetramine, which is said to be a stable adduct (Smith 1983, Weyerhauser 1984). It is conceivable that there could be degradation of that complex over periods of time not yet measured by investigators. In addition, the ammonia treatment may be effective only for free formaldehyde present in the pressed-wood product at the time of treatment and not for any formaldehyde liberated by resin hydrolysis or other mechanisms (unless an excess of ammonia could be maintained to act as a continuous scavenger).

The information presented in this discussion does not provide unequivocal evidence of the long-term effectiveness of this control option. The treatment is, however, apparently effective for months or possibly years; Weyerhauser suggests repeated use to ensure reduced exposure to mobile home residents (Weyerhauser 1984).

An important consideration in the evaluation of control option feasibility is the effect of a treatment on the physical characteristics of the pressed-wood product. Little research has been reported in this area, but one investigator has shown that ammonia fumigation does not adversely affect particleboard shear strength (Myers 1982b). No data were found regarding the effect of ammonia fumigation on aesthetic properties of pressed-wood products (primarily color changes).

5.2.2 Post-Cure Board Treatments with Other Scavengers

A variety of post-cure scavengers can be applied as coatings (paints, varnishes, etc.) or as aqueous solutions. The chemicals most often tested are sulfur and nitrogenous compounds, which act similarly to ammonia in that they react chemically with free formaldehyde to produce a more stable complex.

Geomet (1980) summarized literature available at that time regarding the effectiveness of various scavengers applied to pressed-wood products as coatings. Sundin (1978) tested a urea-based paint and found that it reduced formaldehyde emissions by nearly 75 percent. Those results must be termed short-term effectiveness; no long-term emission testing was undertaken.

Smith's (1983) literature review discusses the work done by Japanese scientists on formaldehyde emission reduction by use of surface scavengers. It is implied that these applications are not in paints but rather are directly-applied solutions of scavenger. Ammonium salts, guanidine derivatives, amides, and urea have been tested and all found effective in the short term to some extent. Smith concludes that this control option has as yet undeveloped potential.

Barghoorn (1979, in Meyer 1979) presented results showing a reduction in formaldehyde emissions from particleboard treated with various surface coatings containing formaldehyde scavenging chemicals. The procedure involved a chamber test that compared the value of an

untreated board (1.20 ppm) to the values of coated boards. Boards were tested with and without edges sealed, at a loading of 0.6 m^2/m^3, one air change per hour, and sample dimensions equal to 1 x 2 m. Results showed that the edge finish alone reduced emissions by a factor of ten. One coating tested was a melamine-containing coating which, with sealed edges, reduced test chamber concentrations to 0.02 ppm (a 98 percent reduction); with exposed edges, it reduced formaldehyde concentrations to 0.10 ppm (a 92 percent reduction). Falima-F, a commercially available scavenger- containing coating of undisclosed composition reduced dynamic test chamber concentrations from 1.20 (untreated) to 0.03 ppm (a 97 percent reduction). No long-term effectiveness data were available.

Molhave et al. (1983) examined the emission rates of formaldehyde from particleboards treated with Falima-F surface coating. Under standard conditions (23°C, 45 percent relative humidity), with an air exchange rate of 0.25 and a loading of 2.2 m^2/m^3, emission rates varied 38 percent (standard deviation) in the range 25.4 to 70.8 $ug/m^2/hr$. Emission rates for control boards (i.e., not coated) were not provided for comparison.

Another coating tested was Valspar 50100, a urea-containing wood product surface coating. Myers (1982b) compared dynamic test results between untreated commercially unfinished lauan plywood (5.6 mm thick), deliberately selected because of its high formaldehyde emission, with Valspar treated boards. Samples were cut to 50 x 125 mm sizes and had their edges sealed. Results at 35°C and 60 percent relative humidity showed a ten-fold (90 percent) reduction in formaldehyde emissions from 3.00 ppm to 0.3 ppm. Scavengers are also sprayed over newly pressed boards, usually in an aqueous solution to facilitate their absorption into the board.

Myers (1982b) found that the Valspar varnish acted both as a barrier to reduce water and formaldehyde transmission across the board surface and as a scavenger. The control was about 90 percent effective in

reducing emissions. He cites continued reduction after 30 days as evidence of the long-term effectiveness of the treatment. Little is known about the effects of surface scavengers on board properties, though little effect is expected. Scavengers are used to react with free formaldehyde; when the scavenging chemical is used in a post-cure treatment, as in a varnish, the effects on resin bonds should not be significant.

The Dutch have also tested the effectiveness of a vinyl-toluene paint by coating the inner walls and roof plates (made from particleboard) of experimental houses in Haarlem District, the Netherlands (Van der Wal 1982). Although little is stated about the conditions of the boards and the testing itself, detailed results were provided. These were summarized in Table 26 this report. Overall, the painting reduced formaldehyde concentrations by a factor of 1.5 to 3.0 (in the short term).

Use of scavengers with boards of low F:U ratio will not be as effective as with boards of higher F:U, simply because there would be less free formaldehyde for the scavenger to remove (as indicated by comments by the HPMA). The long-term effectiveness of this treatment is essentially unknown. Union Camp (1984) specifically questions the effectiveness of this method.

5.2.3 Non-Scavenger Emission Barriers

The primary function of non-scavenging surface coatings in the treatment of formaldehyde emitting boards is to prevent the absorption of water by the boards thereby mitigating subsequent resin hydrolysis and formaldehyde off-gas. Since none of the studies cited in this subsection referred to their results as being long term, it is assumed that all of the effectiveness indications are reports of short term emission tests. There do not appear to be any data on the long-term effectiveness of these coatings.

ICF (1984) reports that the use of nitrocellulose coatings decreases measured formaldehyde emissions (desiccator method) from 1.20 ppm to 0.09 ppm (92.5 percent) and 0.02 ppm (98 percent) for boards with exposed and sealed edges, respectively. Meyer (1979) reports studies that indicate 96 percent emission reductions (13.2 to 0.5 ppm JIS desiccator method) for plywood treated with polyurethane applied at a rate of 100 grams per square meter.

Other surface coatings tested for effectiveness on particleboards in chamber tests (1 air change/hr; $0.6m^2/m^3$), also reported in Meyer (1979), include: wallpaper with sealed edge, 33 percent reduction (untreated board caused 1.20 ppm); macore overlay with coated edge, 84 percent reduction; varnished surface with coated edge, 98 percent reduction; varnished surface with uncoated edge, 92 percent reduction; and overlay paper with sealed edge, 93 percent reduction.

Kazakevics (1984) evaluated the effect of emission barriers (paints and wallpaper) on formaldehyde release from particleboard. The barriers he evaluated successfully reduced emission rates below his method detection limit (0.01 $mg/m^2/hr$).

The effect of a decorative vinyl overlay on formaldehyde emissions was studied by Groah et al. (1984). The study examined large chamber and 2-hour desiccator tests on 4.0 mm thick plywood paneling with a 2 mil (0.002 inch) vinyl film adhered to one panel face. Average concentrations found in the large chamber tests were 0.75 ppm when both vinyl faces and unfinished backs were exposed, and 0.04 to 0.08 ppm when only vinyl faces were exposed. Average desiccator test values were, for exposed faces and backs, 2.06 ug/ml; for vinyl faces only, 0.13 ug/ml; and for unfinished back only, 3.04 ug/ml.

Hardwood plywood wall paneling is almost always produced with a factory applied finish; the user does not have to paint or decorate the

panel after installation. With the reduction in emissions from UF adhesive systems, it has become apparent that the impact of the coating system has become more important. Over the past 5 to 10 years, the flat wood paneling industry has been changing from solvent to water-base coatings to reduce volatile organic emissions (VOC) so as to meet ambient air quality guidelines for atmospheric releases from manufacturing plants. Some water-based systems now contain formaldehyde in order to minimize VOC emissions, to achieve a decorative finish with appropriate application characteristics, and to provide for a product with suitable durability and with aesthetic and surface properties acceptable to the consumer. Therefore, some coatings may even enhance the formaldehyde emissions. It is not known at this time whether formaldehyde emitted from the top coat of water-based finishes has significant impact on long term formaldehyde concentrations in living spaces (HPMA 1984).

The Council of Forest Industries of West Germany (1981) lists over 14 approved coatings or finishes for reduction of formaldehyde emissions from untreated particleboard of emission categories E2 and E3 (see Section 6.3 for definition of these codes). Those finishes and their application rates include:

- melamine resin inpregnated paper
- laquer coating on film underlay ($\geq$250 g/m^2)
- polyester (styrol) varnish ($\geq$250 g/m^2)
- 2-component polyurethane varnish ($\geq$300 g/m^2)
- oil-based alkyd resin paint ($\geq$230 g/m^2)
- veneers (walnut, makore, oak, pine) plus nitrocellulose ($\geq$34 g/m^2), polyurethane ($\geq$30 g/m^2), or polyester ($\geq$35 g/m^2)
- Falima - F coating (200 g/m^2)
- Falima - 271 coating (200 g/m^2)
- 0.5 mm plastic laminate
- Rigid PVC film (100 - 180 mm)
- PVC film, 18% plasticizer (0.08 - 0.1 mm)
- Laminated plastic on unsaturated polyester (0.5 mm)
- PVC film, 16% plasticizer (0.18 mm)

Use of any of these coatings or finishes will produce boards conforming to German E1 standards, discussed further in Section 6.

5.3 Substitute Resins

Much work has been done on substituting other resins for the urea-formaldehyde now used in pressed-wood products. Potential substitute resins include tanning-formaldehyde (Forss and Fuhrmann 1980, Coppens et al. 1980); melamine formaldehyde, resorcinol formaldehyde, and polyvinyl acetate adhesives (NPA and HPMA 1984); spent sulfite liquor adhesive (Shen 1983) or lignin-UF-PF combinations (Forintek 1983); and urethane or polyester binders (White 1979). There are, however, technological or cost restraints severely restricting the use of any of the above-listed adhesives. This report will discuss only the most promising substitutes for urea formaldehyde in pressed-wood products -- phenol formaldehyde resin and isocyanate resin.

5.3.1 Phenol Formaldehyde Resin as a Substitute for Urea Formaldehyde Resin

Of all the substitute resin mixtures known, phenol-formaldehyde resins have been studied most extensively. From collective testing, the pressed-wood product industry is convinced that phenolic panel products (pressed-wood products made with phenol-formaldehyde resin adhesive) (1) emit very little formaldehyde in the long or short term and (2) insignificantly affect the formaldehyde levels found indoors and outdoors. Their large test chamber studies have shown that even freshly manufactured phenolic panel products produce formaldehyde levels at less than 0.1 ppm (American Plywood Association 1984). Monitoring conducted in three mobile homes containing only phenolic panel products showed formaldehyde levels to be less than 0.1 ppm (average levels ranged from 0.02 to 0.07 ppm) (Singh et al. 1982a).

Phenol is more reactive with formaldehyde than is urea; the resultant resin is therefore more durable and emits considerably less formaldehyde according to the American Plywood Association (APA 1984). The formaldehyde that is released is the small amount of free formaldehyde present after manufacture, and no release via resin hydrolysis is expected (APA 1984). The substitution of PF resin thus represents a viable short- and long-term emission reduction option. This control option could include substitution of PF resin in products now formulated with UF resin or actual product substitution (i.e., use of PF-bonded softwood plywood in place of hardwood plywood made with UF). The latter, product substitution, is discussed separately in Section 5.4 of this report. Phenol-formaldehyde is currently used by a small number of particleboard manufacturers, accounting for about 6 percent of 1983 U.S. particleboard production capacity (ICF 1984) and is the sole adhesive used by makers of softwood plywood (APA 1984). Waferboard, hardboard, and oriented-strand board are also based on phenolic resins and are consequently low formaldehyde emitters.

Phenol formaldehyde is a suitable resin for substitution in essentially all pressed-wood products that now use UF. The NPA cites some difficulties with PF (low tack, loss of dimensional stability), and the Manufactured Housing Institute adds that PF particleboard is, by necessity, of costly tongue-and-groove construction (MHI 1984). An additional consideration is that PF resins cause certain light-colored woods to be darker than they would be if UF resin were used, thus limiting its applicability in some furniture and fixture construction (Champion 1984). The major drawback cited by industry is the higher cost and limited availability of PF resin, which stems from the general economic instability of all petroleum-based products (including phenol).

It appears from all relevant data that PF could be substituted in pressed-wood products for UF as follows:

Particleboard - Phenol-formaldehyde can be substituted for urea formaldehyde in all types of particleboard except for panels thicker than 1 3/4 inches, according to Champion (1984). The limiting factor on thicker particleboard was not stated, but relatively little particleboard is manufactured thicker than 3/4 inch.

Medium-density fiberboard - No data specific to this product were found. It is likely that applications similar to those for particleboard would be feasible. The NPA (1984) states only that limited experimentation has been performed with this resin-wood combination, but cites no technological factors that would render this use nonfeasible.

Hardwood plywood - Many manufacturers submitted comments on EPA's proposed 4(f) rule addressing this substitution. Hardwood plywood made with PF is said to exhibit a tendency to warp or expand in high humidity environments, necessitating very careful installation (MHI 1984); that problem may be solved by proper formulation of the resin with waxes and other fillers, according to Champion International (1984). The only situation in which PF is an inappropriate resin is in the veneering of light colored woods, such as oak, or when thin veneer of porous woods like elm, birch, hickory, and pecan are to be glued to the surface (HPMA 1984, Champion 1984).

Myers and Nagaoka (1981b) measured emissions from two sets of phenolic particleboards made with varying press times. Dynamic chamber tests were performed involving one ventilation rate (1.15 air changes per hour), a chamber loading of 19.2 m^2/m^3, and two sets of atmospheric conditions (25°C/75 percent RH and 40°C/75 percent RH). Results averaged only slightly above 0.1 ppm at 25°C and only approached 0.2 ppm at the higher temperature, despite the very high chamber loading.

Meyer (1981) evaluated formaldehyde emissions from four plywood samples, a 3/4 inch particleboard, and a waferboard, all manufactured with PF resins. Highest emissions (3.85×10^{-6} mg formaldehyde/ml test

solution, as measured by a perforator test were from the PF particleboard. A Douglas fir plywood released 2.5 x 10^{-6} mg/ml; that same type of wood product, after accelerated aging, released only 0.22 x 10^{-6} mg/ml. Releases from the waferboard, a southern pine plywood, and a fir/hemlock plywood were nearly equal at 1.45, 1.35, and 1.30 x 10^{-6} mg/ml, respectively.

Meyer used these data to predict equilibrium indoor air formaldehyde levels that could result from the use of these PF-resin pressed wood products in a home. At a loading rate of 1.18 m^2/m^3 and no air changes, the predicted formaldehyde level resulting from the use of the highest emitting product (particleboard) was 0.05 ppm; at 0.5 ACH, the predicted level was 0.0025 ppm.

Myers (1983) reported very low perforator and 24-hour desiccator measurements for four PF particleboards. Perforator values ranged from 1.1 to 1.4 mg/100g; 24-hour desiccator values ranged from 0.12 to 0.26 ug/ml. Myers (1984c) measured the formaldehyde concentrations resulting from different air exchange rate/loading combinations of a PF particleboard in chamber experiments. At a particleboard loading rate typical for mobile homes, Myers reported formaldehyde concentrations of 0.02 ppm for an air exchange rate of 0.5 per hour and 0.04 ppm for an air exchange rate of 0.25 per hour.

Matthews et al. (1982-1984) applied several test methods to phenolic hardboard and softwood plywood panels (as well as to other pressed-wood products) to investigate the dependence of emission rates on the background concentration of formaldehyde. Interpretation of the curves plotted indicates that averaged emission rates were essentially zero when background formaldehyde levels were about 0.1 ppm. The results of the tests indicate that these phenolic boards could produce, at most, an indoor air level of about 0.1 ppm formaldehyde regardless of the board loading rate or air exchange rate.

Manufacturers' data on phenolic pressed-wood products are also available. Champion International Corporation has published the results of large chamber tests for their phenolic plywood, waferboard, and particleboard. At a chamber loading of 0.43 m^2/m^3, a relative humidity of 50 ± 2 percent, a temperature of 24 ± 1°C (75 ± 2°F), and an air exchange rate of 0.5 changes per hour, formaldehyde levels in the test chamber are shown to be less than 0.1 ppm for all three types of phenolic panels. Even lower levels are shown for a ventilation rate of 1 air change per hour and for products covered with either a resilient floor cover or a pad and carpet (Champion International 1984).

Table 53 summarizes the results of a large study done on several phenolic products by the American Plywood Association (1984). In this investigation, formaldehyde emissions from most major types of phenolic panels were measured using large-scale dynamic chambers and two-hour desiccator tests. Table 54 summarizes additional emission data which have been furnished to the American Plywood Association (1984) by various phenolic panel manufacturers. Data from both large-scale dynamic chamber and two-hour desiccator tests are again provided. The data from both Tables 53 and 54 seem to confirm that phenolic products are not likely to contribute more than 0.1 ppm formaldehyde to indoor air levels.

5.3.2 Isocyanate Resins

Isocyanate resins contain no formaldehyde, making their use by definition an effective short and long term control measure. The presence of formaldehyde in isocyanate particleboard and in wood chips may be due to partial degradation of lignin or carbohydrates during the drying process (Roffael 1978). This incidental level of emission is demonstrated graphically in Figure 24. These resins are similar in cost to phenol-formaldehyde resins, and the major disadvantages to their use

Table 53. Summary of Formaldehyde Test Data from Various Phenolic-Bonded Panel Products Measured by Dr. W.F. Lehmann of Weyerhaeuser Co.

Products	2-Hour desiccator ug/ml	Large-Scale Test Chamber[a]					
		Initial test			Re-test[c]		
		Panel age (days)	W/panels (ppm)	Empty (ppm)	Storage time (Mo.)	W/panels (ppm)	Empty (ppm)
Southern pine plywood 13 mm, 4-ply	0.08	32	0.04[b]	0.01	4	0.03	0.02
Douglas-fir plywood 14 mm, 5-ply	0.18	<1	0.05	0.01	8	0.05	0.05
Oriented strand board No. 1 12 mm	0.14	19	0.07	0.01	-	—	—
Oriented strand board No. 2 (Sample No. 1) 12 mm	0.02	21	0.07	0.07	3	0.04	0.02
Oriented strand board No. 2 (Sample No. 2) 12 mm	0.09	21	0.03	--	-	--	—
Waferboard (Sample No. 1) 12 mm	0.17	55	0.08	0.03	8	0.01	0.01
Waferboard (Sample No. 2) 12 mm	0.03	21	0.06	0.03	-	--	--
Particleboard 19 mm, hot-melt coating	0.15	16	0.08	-	9	0.03	0.01

(a) Test conditions: 25 ± 1°C, 50 ± 5% RH, 0.5 AC/h, 0.43 m^2/m^3 loading.

(b) The sample was retested at 0 air changes per hour, and formaldehyde concentration was found to be 0.06 ppm.

(c) Re-test was performed after 3 months sample aging.

Source: American Plywood Association (1984).

Table 54. Results of Large-Scale Dynamic Chamber Tests and Two-Hour Desiccator Tests on Various Types of Phenolic Panel Products[a]

Product type[b]	Age at test (Days)	Chamber test parameters: Precondition time[c] (Days)	Loading rate[d] (m^2/m^3)	Temp. (C)	Relative humidity (%)	Formaldehyde levels: Two-hr. desiccator (ug/ml)[e]	Large chamber: Empty (ppm)	Large chamber: Loaded (ppm)
So. pine ply, 23/32", 4-ply	--	8	0.95	23 ± 0.5	48 ± 1	--	0.015	0.022
So. pine ply, 5/8", 5-ply	--	8	0.95	23 ± 0.5	48 ± 1	--	0.010	0.020
So. pine ply, 5/8", 5-ply	-	8	0.95	23 ± 0.5	48 ± 1	--	0.005[f]	0.011[f]
D.-fir ply, 1/2"	--	8	0.95	23 ± 0.5	48 ± 1	--	0.013	0.017
Particleboard, 5/8"	--	2	0.49	24 ± 1	50 ± 1	0.51	--	0.04
So. pine ply, 5/8", 5-ply	<30	2-3	0.49	24 ± 1	50 ± 5	0.09	0.03-0.04[g]	0.03
COMPLY, 5/8"	<30	2-3	0.43	24 ± 1	50 ± 5	0.13	0.03-0.04[g]	0.04
Waferboard, 5/8"	22	2-3	0.52	24 ± 1	50 ± 5	0.17	0.03-0.04[g]	0.03
Waferboard 5/8"	--	2-3	0.52	24 ± 1	50 ± 5	0.18	0.03-0.04[g]	0.05
Particleboard 3/4"	-	;[h]	0.43	24 ± 1	45 - 55	0.22	0.01	0.04
Particleboard, 3/4"	--	][i]	0.43	24 ± 1	42 - 48	0.17	0.01	0.05
Particleboard 3/4"	--	][j]	0.43	24 ± 1	42 - 50	0.20	0.01	0.04
So. pine ply, 15/32"	--	2	0.43	25 ± 1	42 - 56	0.34[k]	-	0.04[l]

Table 54. (Continued) - Footnotes

(a) Ventilation rate was 0.5 air changes per hour for all chamber tests. Chromotropic acid was used for formaldehyde analyses, unless noted otherwise. Different test chambers were used by each of the companies represented.

(b) So. pine ply = southern pine plywood; D-fir ply = Douglas-fir plywood.

(c) Specimens were preconditioned at the same temperature, and relative humidity as is given for the test chamber.

(d) Loading is given in terms of square feet of panel surface per cubic feet of air volume in the chamber.

(e) All desiccator tests were performed in accordance with the procedures given by the National Particleboard Association, Test Method FTM-1, with edges unsealed unless noted otherwise.

(f) Pararosaniline was used for formaldehyde analysis, rather than chromotropic acid, using the same panel specimens as those used in the test whose results are reported directly above.

(g) Range typically encountered at this test facility.

(h) Background formaldehyde level in conditioning area was 0.08 ppm.

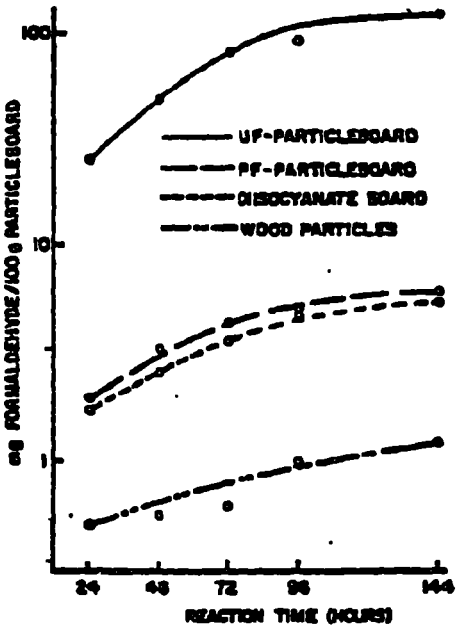
(i) Background formaldehyde level in conditioning area was 0.03 ppm.

(j) Background formaldehyde level in conditioning area was 0.05 ppm.

(k) Average of four tests involving samples from two separate panels; for each panel samples for one test had sealed edges, while those for the other tests were unsealed. Range = 0.29 - 0.43.

(l) Average of four measurements made on four consecutive days. Range was 0.02 - 0.05.

Source: American Plywood Association (1984).



Source: Roffael (1978).

Figure 24. WKI Method Test Results for Cured Resin/Wood Composites and Dried Wood Particles

are (as in the case of PF) related to increases in resin and finished product costs. An additional consideration cited by many in industry is the paucity of knowledge about the health effects of exposure to isocyanate resins and their byproducts; available data indicate potential problems with their use. Isocyanate resins include, as a class, a large number of compounds differing in cost and properties. Most investigators consider MDI (4,4'-diphenylmethanediisocyanate) to be the most promising as a wood adhesive.

Deppe (1977) was the first to publicize the feasibility of isocyanate binders for use in pressed-wood products in the U.S. He compared the use of isocyanate to PF resin in particleboard and reported an increase in shear strength, a decrease in thickness of swelling, and comparable properties for other parameters.

Wilson (1981) and Adams (1980) discuss three varieties of MDI isocyanate binders. Polymeric MDI, or PMDI, is the resin currently used by manufacturers that make isocyanate based pressed-wood products. Emulsifiable MDI (EMDI) and monomeric MDI are also discussed as potential adhesives. He found that PMDI and EMDI are approximately equal in strength properties, and performed better than UF resin pressed-wood products, but that polyols of those resins were actually the best adhesives. All other resins tested performed better than monomeric MDI.

Ellingson Lumber Co. presently manufactures a particleboard bound with isocyanate (PMDI) (Ellingson 1984). They state that the resultant product is competitive with traditionally-manufactured particleboard and medium-density fiberboard (MDF) for many applications, including as underlayment. This report supports the theory that there are no technological restrictions on the substitution of MDI for UF, at least in particleboard manufacture. It is likely that MDI would also be suitable

for use in MDF, and the NPA reported limited testing of that combination but no commercial manufacture (NPA 1984). Wilson (1980) states, however, that the low tack properties of MDI may render it unsuitable for many types of plywood.

5.4 Substitute Wood Products

EPA is considering several material substitutions as potential control measures for lessening formaldehyde exposures in residences. Some of these substitutes are products with no formaldehyde, while some are low-emitting products. These options are described below.

5.4.1 Hardboard

Hardboard is, in most instances, manufactured with PF resin and is a low-level formaldehyde emitter. It is used in the following applications: exterior siding, interior paneling, household and commercial furniture, and industrial board (American Hardboard Association 1984). It competes with both hardwood plywood and softwood plywood, but is not more extensively used at this time because of cost considerations. Hardboard currently makes up 23 percent of the market for residential paneling and is the leading component of exterior siding (AHA 1984).

It thus appears that the value of this control option lies in its potential use as a substitute for hardwood plywood that is formulated with UF resin or for MDF or particleboard used in furniture and cabinets. The potential utility of hardboard as a substitute for particleboard underlayment and similar applications is less clear.

5.4.2 Gypsum Board

Gypsum board is considered a viable alternative to hardwood plywood in the manufactured housing market, and is currently gaining favor (MHI 1984). Gypsum board contains no formaldehyde, which would lead one to believe that this substitution would be an entirely effective control option. Recent studies indicate that gypsum board is a strong absorber of formaldehyde in the atmosphere, and can emit that absorbed formaldehyde if other sources are temporarily controlled (Pickrell et al. 1984, Weyerhayser 1984). The effectiveness of this option therefore depends to a great extent on the presence of other formaldehyde sources within the home.

5.4.3 Other Substitutes

Softwood plywood is, as discussed previously, a potential substitute for hardwood plywood under some circumstances of use. It is viable for some interior paneling uses (those not involving hardwood veneering) and for use as decking. Like gypsum board, softwood plywood bonded with PF resin may be a formaldehyde absorber and can become a source under some circumstances. There are also formaldehyde emissions, though slight, associated with PF resin products like softwood plywood.

5.5 Increased Room Ventilation

Commenters on the Advanced Notice of Proposed Rulemaking (ANPR) for the formaldehyde 4(f) investigation mentioned that ventilation, or increasing a home's air exchange rate, may be an effective means of controlling formaldehyde levels (Weyerhauser 1984, NPA 1984). An increase in ventilation rate will reduce the formaldehyde concentration, though unlike many other indoor air pollutants the reduction will not be in direct proportion to the ventilation change (NPA 1984); the lowering

of the ambient concentration will trigger an increased emission rate. The NPA states that doubling the ventilation rate will cut the formaldehyde level by only one-third. A marginal increase in exposure reduction may be achieved with air-to-air heat exchangers when used with increased ventilation (MHI 1984, Weyerhauser 1984).

Myers (1984b) critically reviewed the available literature concerning the effect of ventilation rate and pressed-wood product loading on indoor formaldehyde concentrations. Although a large number of studies of formaldehyde levels have been reported for a variety of buildings, Myers found that very few of these studies report measurements of ventilation rates in the buildings. Of these few studies, Myers determined that only three (Jewell 1980b; Moschandreas and Rector 1981; and Singh et al. 1982b) provide sufficient data to permit even a semiquantitative evaluation of ventilation rate effects.

Based on the concentration data and air exchange rate and product loading information given in the studies, Myers fit the three data sets to the HBF equation* so as to have a basis for consistently evaluating the studies' results. Myers found that the calculated (and observed) changes in formaldehyde concentration with increasing ventilation rate were within the ranges seen in controlled chamber tests with pressed-wood products. A doubling of the air exchange rate from 0.25 to 0.50 ACH (air changes per hour) decreased the formaldehyde concentration by as little as 8 percent to as much as 37 percent.

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*The HBF equation is a model developed by Myer (1984b) based on research by Hoetger, Berge, and Fujii. The HBF equation, like the emission rate models being developed by ORNL for CPSC (see Section 7.1), is an expression that linearly relates the steady-state concentration of formaldehyde in a chamber to the air exchange rate and loading of the pressed-wood product.

Increasing the air exchange rate from 0.50 to 1.0 ACH decreased the concentration by 9 to 42 percent. Increasing the air exchange rate from 0.25 to 1.0 ACH decreased the concentration by 17 to 63 percent.

Thus, as has been found in chamber tests with pressed-wood products, increasing the ventilation rate will reduce formaldehyde concentrations, but the reductions achieved will typically not be as large as one might expect; that is, a simple doubling of the air exchange rate will not necessarily reduce the concentration by 50 percent, but rather by about one-third or possibly less. The emission rate of formaldehyde from a pressed-wood product is a function of the concentration of formaldehyde in the ambient air surrounding the board. The emission rate will decrease as the ambient air concentration increases, and it will increase as the ambient air concentration decreases.

5.6 Presale Storage (Board Aging)

Aging boards under conditions that promote formaldehyde emission, before they are sold, has been shown to be an effective option for controlling consumer exposure. Although this process is not under active consideration by EPA at this point, many data are available and reported in this section.

Kazakevics (1984) performed a five-year study on the effects of board aging on formaldehyde emission. Soon after manufacture, emissions were 10 to 100 times higher than after five years. Particleboards with emissions of 12 $mg/m^2/hr$ were tested five years later and emitted 0.1 to 1.1 $mg/m^2/hr$. Kazakevics found that emissions from boards of F:U 1.0 to 1.5 leveled off from high levels of "free formaldehyde" to lower levels of "hydrolysis products" at approximately 12 months. Fluctuations were attributable to changes in the climate in which the boards were stored.

Myers (1982b) presented the following testing results of aging on hardwood plywood board samples. After a 30-day aging at 75°C and 50 percent RH, the mean formaldehyde air concentration in a dynamic testing chamber was reduced by 90 percent (3.0 to 0.31 ppm). After two such aging periods, the mean concentration dropped by 100 percent (3.0 to <0.01 ppm or below the detection limit). Twenty-four hour desiccator tests on similar board samples after identical aging conditioning showed a 93 percent reduction in formaldehyde emission after only a 15-day aging period.

Testing results in Myers (1982b) also showed how presale board aging could be used effectively with other control options. For example, hardwood plywood board samples treated with a urea-containing surface coating were measured in dynamic testing chambers at a mean value of 0.3 ppm. After one 30-day aging period (75°C and 50 percent RH) measured formaldehyde concentrations were reduced to 0.039 ppm (87 percent improvement). In the same study, the combined effect of board aging and ammonia scavenging was evaluated. In a 24-hour desiccator test, aging had a much greater impact on the untreated plywood (3.9 to 0.3 ug/ml or 92 percent improvement) than on the ammonia treated plywood (0.06 to 0.05 ug/ml or 17 percent improvement). In the dynamic chamber test, ammonia-treated plywood samples, initially measured for very low formaldehyde emissions (<0.01 ppm), increased slightly after aging. It was suggested that this increase was due to a loss of sorbed ammonia during aging and, thereby, a loss of scavenging capability.

Forintek (1983) evaluated the effect of board aging on formaldehyde emission rates from particleboard and hardwood plywood. They performed desiccator tests daily for a period of 60 to 223 days, then plotted the $\ln(CH_2O)$ vs time. This confirmed that the half-life of particleboard emissions is around 8 to 9 months, and indicated that after 60 days of storage, emissions decline at the rate of 1 percent per day.

5.7 Approaches to Reducing Formaldehyde Emissions from UF Bonded Wood Products Based on Resin Chemistry

This section basically consists of excerpts from a recent draft report by H. Podall of EPA's Office of Toxic Substances entitled "A Review of the State-of-the-Art on Urea Formaldehyde Resins for Wood and Causes of Formaldehyde Release" (USEPA 1984).

Any approach regarding physical/chemical changes in the resin, treatment of the board, the curing step, or in the final board treatment or finishing, aimed at reducing initial formaldehyde emissions, must also take into account the long-term hydrolytic stability of the resin/wood composite. Although our understanding of the exact sources and mechanisms of the releases is not complete, the following approaches appear desirable for reducing the long-term, as well as the initial, releases of formaldehyde from UF-bonded wood products:

(1) Requirement of lower ratio F:U resins in imported hardwood plywood boards, comparable to those products manufactured in the U.S.

(2) Storage of resins in dry forms (where storage is required) to reduce the decomposition of the resin to formaldehyde.

(3) Moisture-proofing of furnishes in composition boards or of veneers in hardwood plywood to reduce affinity for moisture.

(4) Minimize or avoid addition of other acidic components (such as formic acid) to the resin formulation or to the wood, in order to minimize long-term hydrolysis of resin components to formaldehyde.

(5) Use of a minimum amount of NH_4Cl or $(NH_4)_2SO_4$ as acid catalyst (hardener) for cure.

(6) Production of tighter boards to reduce permeability of moisture in board and hence displacement of formaldehyde.

(7) Treatment of cured board with an appropriate base of sufficient strength (possibly sodium bicarbonate or triethanolamine) to reduce the free acid concentration in the board to a pH of about 7.

(8) Coating of edges of board (and both sides - particularly for composition boards) with a moisture-impervious coating to reduce diffusion and escape of formaldehyde, eliminate displacement of formaldehyde by moisture, and to reduce and/or eliminate mid- to long-term hydrolysis of various formaldehyde-releasing species.

(9) Use of low F:U mole ratio resins (e.g., 1:1) which do not require an acid hardener for curing.

(10) Use of veneers for hardwood plywood or furnishes for composition board whose wood is approximately neutral (pH 7).

(11) Use of appropriate external crosslinking agents, such as trimethoxymethylmelamine, added to the resin formulation to facilitate curing (to the desired three-dimensional network) and/or to obviate the requirement for an acid hardener.

6. FORMALDEHYDE STANDARDS FOR WOOD PRODUCTS AND INDOOR AIR

Many European countries, and recently the United States (through the Department of Housing and Urban Development), have promulgated standards that limit formaldehyde emissions from wood products and/or set maximum allowable indoor air concentrations for formaldehyde. Table 55 lists the standards and the associated analytical test methods for determination of emission potential.

As can be seen in the table, the analytical test methods upon which the standards are based include chemical extraction (i.e., perforator), static chambers (JIS and TNO), and dynamic chamber methods. The perforator methods vary little among European nations, and the results are in general directly comparable. The static chamber methods are difficult to interrelate because of variable testing conditions (Matthews et al. 1982 Report V.) The dynamic chamber methods are also difficult to compare because they differ in air exchange rate, loading factor, pre-conditioning of boards, and measurement procedures. In 1978, the European countries formed an official Technical Committee (CEN TC 91) in an attempt to develop standardized dynamic chamber test methods and preconditioning methods that would be used by all countries (Gaudert et al. 1983). The work of this committee apparently came to a halt in 1983 before a standardized method had been developed.*

Details on the standards of several countries are presented below.

6.1 Denmark

All pressed-wood products used for construction purposes (e.g., particleboard, plywood, waferboard, hardboard, MDF) are governed by the standards. The maximum allowed perforator value for any board is 25 mg/100 g. Any board that is to be used in places that people

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*Personal communication between G. Schweer (USEPA/OTS) and P. Gaudert (National Research Council of Canada) on October 30, 1984.

Table 55. International Indoor Air Standards for Formaldehyde and Standards for Formaldehyde Emission from Pressed Wood Products [a]

Nation	Product type	Use(s)	Standard or Guideline [c]	Standard test method	Indoor air Standard or Guideline[e]
Belguim	Particleboard	Indoor Use		Perforator	
	Class 1		14 mg/100g		
	Class 2		28 mg/100g		
	Class 3		42 mg/100g		
Denmark	All pressed wood products	Homes, schools, etc.	0.12 ppm	Dynamic chamber	0.12 ppm (law)
	All pressed wood products	Non-home, etc.	25 mg/100g	Perforator	
Finland	Particleboard	Indoor Use	30 mg/100 g (mean)	Perforator	0.12 to 0.24 ppm (guideline)[b]
			50 mg/100 g (mean)	Perforator	
France	Particleboard	Indoor Use	50 mg/ 100 g	Perforator	
Italy					0.10 ppm (guideline)
Japan	Particleboard		5.0 ug/ml	Static JIS method	----
Netherlands	Particleboard	Government-subsidized housing	10 mg/100 g	Perforator	0.10 ppm (guideline)
Norway	Particleboard	Indoor use	30 mg/100 g	Perforator	----
Spain	Particleboard	Indoor use	50 mg/100 g	Perforator	----
Sweden	Particleboard	Indoor use	40 mg/100 g	Perforator	0.4 ppm (guideline)
Switzerland	Particleboard	--	(under development)	--	0.2 ppm[d] (law)
United Kingdom	Particleboard	--	50 mg/100 g (mean)	Perforator	----
			70 mg/100 g (max.)	Perforator	----
United States	Particleboard	Mobile homes	0.3 ppm	Dynamic chamber	----
	Hardwood plywood	Mobile homes	0.2 ppm	Dynamic chamber	
West Germany	Particleboard (E1)	Indoor use	≤0.1 ppm	Dynamic chamber	0.10 ppm (guideline)
	Particleboard (E2)	Indoor use	>0.1 to ≤1.0 ppm	Dynamic chamber	
	Particleboard (E3)	Indoor use	>1.0 to <2.3 ppm	Dynamic chamber	

Table 55. Footnotes

[a]Based on information reported in Gaudert et al. (1983), Matthews et al. (1981-1982 Progress Reports IV, V), and Sundin (1985).

[b]See Section 6.2 for additional details.

[c]Listed values are standards in Denmark, Finland, Sweden and the U.S. Listed values for West Germany and the Netherlands are guidelines. The legal status of the listed values for the other countries is uncertain.

[d]Personal communication between G. Schweer (USEPA) and Dr. B. Gfeller (Novopan-Keller AG), January 22, 1985.

[e]The indoor air standards refer to measurements conducted under "normal indoor conditions". The criteria for determining "normal indoor conditions" vary from country to country but are within the following parameter ranges: temperature - 20 to 24°C; relative humidity - 40 to 60%; and air exchange - 0.5 hr^{-1} (Sundin 1982).

normally inhabit (e.g., homes, schools) must also either result in an air level of 0.12 ppm formaldehyde or less in the Danish Chamber Test or be treated in a manner approved by the Danish authorities (e.g., surfaces covered with PVC foils, melamine paper, veneers, or formaldehyde absorbing paints) so that the emission value does not exceed 0.32 ppm (Matthews et al. 1982 Report V) Denmark has also promulgated an indoor ambient air standard of 0.12 ppm formaldehyde (Gaudert et al. 1983).

6.2 Finland

Finland has established an indoor ambient air guideline of 0.12 ppm of formaldehyde for new buildings (i.e., constructed during or after 1983) and 0.24 ppm for old buildings (Niemela and Toppila 1984).

6.3 West Germany

West Germany has a graded product standard in place for particleboard used for construction and in kitchen cabinetry. The standard was initially a national guideline but has been adopted as local law throughout the country. Products with formaldehyde chamber values less than or equal to 0.1 ppm can be used, uncovered, in the home; these are termed E1 boards. Boards with chamber values ranging from >0.1 to $\leq$1.0 ppm, called E2 boards, must have exposed surfaces covered prior to use in homes. Boards with chamber values ranging from >1.0 to $\leq$2.3 ppm, E3 boards, can be used only with both surfaces and edges covered. Boards with chamber values in excess of 2.3 ppm cannot be used indoors (Gaudert et al. 1983).

6.4 Netherlands

The Netherlands has established an indoor ambient air formaldehyde limit value of 0.1 ppm (Gaudert et al. 1983). For schools and houses for rent the limit value has a legal base. The Netherlands is in the process of establishing a legally binding product standard for particleboard including furniture of 10 mg/100g (perforator).

6.5 Sweden

In June of 1977, the Swedish government proposed an indoor ambient air interim standard for formaldehyde of 0.4 ppm with a planned final standard of 0.1 ppm. Apparently, this standard is still a proposal and has not been promulgated. However, the interim standard is regarded as semi-official and remedial measures are required by local building boards when it is found to be exceeded (Gaudert et al. 1983).

6.6 United States

On August 9, 1984, the U.S. Department of Housing and Urban Development (HUD) published final regulations limiting formaldehyde emission from particleboard and hardwood plywood used for construction purposes in mobile homes (49 FR 31996). The regulations became effective on February 9, 1985. Hardwood plywood is limited to a maximum emission value of 0.2 ppm by a dynamic chamber method, and particleboard is limited to a value of 0.3 ppm.

The American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE) recommends a limit of 0.12 mg/m^3 (0.1 ppm) of formaldehyde in its "Ventilation Standard for Acceptable Indoor Air Quality" (Standard 62-1981).

7. MODELING FORMALDEHYDE RELEASE FROM PRESSED-WOOD PRODUCTS AND EXPOSURE IN RESIDENTIAL SETTINGS

The following sections discuss efforts by CPSC and EPA to model formaldehyde release from pressed-wood products and subsequent exposure. Different investigators and research projects have focused on different segments of the chain of events that lead to residential formaldehyde exposure. One investigator may, in addition, create multiple "models" of varying complexity and precision.

The first subsection (7.1) describes the ongoing research, funded by CPSC, designed to produce an accurate predictive model for estimating formaldehyde exposure due to emissions from pressed-wood products. The CPSC model is the most complex of those discussed herein; it incorporates algorithms to predict emission, absorption by (and subsequent emission by) formaldehyde sinks, the effect of numerous products, the decay of emissions over time, and the effects of varying environmental conditions to describe dynamic formaldehyde levels in homes.

Section 7.2 describes a simplified version of the CPSC model; in this report, the model is termed "Matthew's Steady-State Model." It incorporates emission predictions for one or more sources but neglects sinks. Matthews proposes this model as a tool to compare the relative impact of residential formaldehyde sources on indoor air levels.

Section 7.3 does not discuss a model per se; it describes the mathematical prediction of long-term levels resulting from "slow" formaldehyde decay. The substance of this section is a statistically-derived equation that represents the "best-fit" for a large collection of long-term formaldehyde monitoring data in mobile homes. Numerous statistical examinations of this data collection are described. This discussion is included here because it is a logical extension of the previously-discussed models. The use of a decay curve such as that described in Section 7.3 can provide integrated, long-term exposure estimates. However, the major disadvantage to this curve is that it is

based on historical monitoring data collected in mobile homes containing pressed wood products that were not manufactured with the low F/U ratio resins in use today. Thus, it may not accurately reflect the decay rates of these new resins.

7.1 CPSC Indoor Air Quality Model

The U.S. Consumer Product Safety Commission (CPSC), through interagency agreements with Oak Ridge National Laboratory (ORNL) and the National Bureau of Standards (NBS) has been developing a sophisticated, computerized indoor air quality model to accurately predict expected formaldehyde levels in housing, given the emission source characterization and the other physical parameters upon which the concentration is based. It is anticipated that this model, after its development and validation (expected in early 1985), will yield indoor formaldehyde estimations representative of pressed-wood product loading scenarios observed in the field. This is a requirement for accurate results in the subsequent exposure and risk analysis proposed for ongoing regulatory investigations.

CPSC has developed and is refining computer programs for a two-compartment model for indoor air quality based on the "mass balance" principle. This principle, simply stated, is that the mass flow into and out of the compartment must be equal and that the rate of change of the pollutant level is determined by the rate of generation and rates of removal. As it is described by Mulligan (1983)*, the formaldehyde mass balance equation calculates the rate of change of the air concentration in a single compartment of specific volume by considering the:

- Mixing Factor
- Formaldehyde contribution from outside air
- Filter efficiency of the ventilation system
- Air and pollutant removed from the compartment
- Emission source term
- Sink term

---

*Much of this Section (7.1) is excerpted directly from Mulligan's (1983) description of the CPSC Indoor Air Quality Model.

In the two-compartment model, one of the compartments only exchanges air with the other compartment. This would usually result in better prediction of the concentrations in one compartment as opposed to an average concentration for the whole house. The sophistication of the model is related to the additional factors augmenting the simple mass balance function. Because formaldehyde emission from pressed wood and other products is complex and because formaldehyde is in equilibrium in the indoor air column, one must incorporate influential factors into the overall model in order to obtain accurate estimates. Three such factors that will be addressed by the model are non-uniform air infiltration, mixing, and air exchange. Other factors that will allow the sophisticated model to better simulate real life situations (such as homes of varying ages, homes where sources have been covered or masked, or homes where disproportionate amounts of sources are found) are currently being researched in support of the model development.

The Oak Ridge National Laboratory (ORNL) has been conducting research on the formaldehyde emissions from pressed wood for the CPSC for the past two years. This research is specifically designed to support the development of portions of the CPSC model; for example, the emission model, barrier model, decay model, etc. The current efforts at ORNL are:

1. Emission characterization of particleboard, paneling and MDF (medium density fiberboard), which represents a random sampling of products produced at the end of 1982.
2. Sorption/desorption characterization of gypsum wallboard.
3. Decay studies of formaldehyde emission from pressed wood products.
4. The effects of permeation barriers on formaldehyde emission from particleboard.
5. The inter-laboratory testing of the formaldehyde surface emission monitor.

In order to predict formaldehyde concentrations in homes, it is necessary to have the data upon which a model of these emissions can be derived and verified. ORNL has developed a chamber test method which allows the emissions to be measured as a function of temperature, relative humidity, and background formaldehyde concentration. Using this method it is possible to develop sub-models (i.e., equations) for the emission characteristics of the various pressed wood products. ORNL has developed two emission models. The current testing is intended to confirm the physical bases for these models and also yield coefficients representative of the pressed wood products on the market at the end of 1982.

While the pressed wood in homes can act as the source of formaldehyde, the gypsum wallboard can act as a sink which removes formaldehyde from the air. However, under certain circumstances (e.g., reduced background levels of formaldehyde) the sink can itself become a source releasing previously absorbed formaldehyde. These actions combine to make prediction more complex and interactive, but they make the resulting real room concentrations less subject to wide variations than could theoretically result from short- term variations in environmental conditions. ORNL has developed an experiment to characterize the behavior of wallboard both in the sorption and desorption cycles.

The emission characteristics of pressed wood vary with time, and ONRL is therefore measuring the emission rate change. Two experiments are being undertaken. The first, referred to by ORNL as the "fast" decay, is conducted under a low background concentration of formaldehyde (~0.1 ppm). It is expected that this will result in rapid decay of the emission rate. The second experiment is being conducted under a high (approximately 0.5 ppm) background concentration of formaldehyde. This experiment is expected to yield information on the slow decay of emission rates.

The emission of formaldehyde from pressed-wood products is affected by any coverings over the pressed wood. The effect of these barriers is being measured at ORNL in a two-part experiment. The first part, which has already been completed, was a dual desiccator setup for measuring the transport coefficient. The resulting data indicate that the rate of emission from pressed-wood products can be expected to be lowered by the presence of barriers. The second, ongoing tests are chamber tests using pressed wood as the source of formaldehyde. In these tests, a teflon lined chamber is placed over the rug/pad/particleboard combination, and the concentration in the chamber is measured. Initial results from these dynamic tests indicate that in actual use the transport of formaldehyde through carpets and padding will result in emission rates slightly lower than indicated by the desiccator tests. This is probably due to a suppression effect at the particleboard/pad interface.

A major element of the successful development of the CSPC indoor air quality model is its validation. The National Bureau of Standards (NBS) has been contracted (under interagency agreement) to support the research necessary to validate the model. The focus of their effort is to investigate, experimentally and theoretically, the behavior of formaldehyde-emitting pressed-wood products in simulated and real homes. This effort will be used to validate both the ORNL developed pressed-wood product emission sub-models and the overall indoor air quality computer model.

The NBS controlled experiment consists of emission rate measurements, in 4 x 8 x 2 ft teflon chambers, of the various pressed-wood products to be used in a two compartment, 10 x 20 x 8 ft test chamber. Both of these measurements are being made in a large environmental chamber capable of maintaining the requisite temperatures and humidities. Once the emission rate has been determined, the products will be installed in the two-compartment chamber, in a manner that simulates their use in a home. A prediction of the expected formaldehyde

concentration will be made, and this prediction will be compared to the actual measurements. Final modeling adjustments will be made in accordance with these experiments.

7.2 Matthews et al. Simple Steady-State Model for Indoor Formaldehyde Concentrations

Matthews et al. have simplified the CPSC model described in Section 7.1 to evaluate the relative importance of a variety of formaldehyde emission sources in a single compartment. The following discussion is in large part excerpted from Matthews et al. (1983b).

At steady-state, the formaldehyde concentration in a single compartment may be expressed as:

$$[CH_2O]_{SS} = [CH_2O]_O + CH_2OER/(C \times ACH \times VOL) \quad (1)$$

where

$[CH_2O]_{SS}$ = steady-state concentration inside the compartment (mg/m^3),

$[CH_2O]_O$ = steady-state concentration outside the compartment (mg/m^3),

CH_2OER = the emission rate of formaldehyde sources inside the compartment (mg/h),

C = the fraction of air coming into the compartment that mixes within the volume (i.e., the mixing factor),

ACH = the flow rate of air through the compartment in compartment volume per time (hr^{-1}), and

VOL = the volume of the compartment (m^3).

The multiplicative product of C and ACH is termed by Matthews et al. as PEX, the effective pollutant exchange rate (in units of hr^{-1}). Equation 1 therefore becomes

$$[CH_2O]_{SS} = [CH_2O]_O + CH_2OER/(PEX \times VOL) \quad (2)$$

Application of the model as expressed in Equation 2 is simplified by assuming that all parameters in the equation remain constant (at steady-state) and that there are no permanent losses of formaldehyde due to irreversible sorption to sinks.

Equation 2 must be rewritten to accomodate the different characteristics of various sources of formaldehyde emissions; emission rate expressions are substituted for CH_2OER in the equation. The three types of formaldehyde emissions, each of which is treated somewhat differently by the model, are:

(1) Solid emission sources in direct contact with indoor air (such as hardwood plywood paneling).

(2) Solid emission sources that have a barrier, reducing emission rate, between the source and the indoor compartment (for example, particleboard underlayment with a carpet barrier).

(3) Combustion sources (cigarettes, gas appliances, etc.)

The first two types of sources listed above are area-dependent in that the magnitude of the emission is a direct function of the surface area of the source in the compartment. The equivalent of Equation 2 for area-dependent sources is

$$[CH_2O]_{SS} = [CH_2O]_0 + CH_2OER' \cdot Area/(PEX \times VOL) \qquad (3)$$

with CH_2OER' in units of mg/m^2hr and area in m^2.

The third, combustion sources, may be modeled with Equation 2 (assuming that the emission rate is constant over time). The emissions expressions for the area-dependent sources, both with and without barriers, are discussed below.

Fick's Law describes the bulk-vapor interphase at the surface of a solid emission source. If one assumes that the mass transport coefficient and the formaldehyde concentration in the bulk phase are independent of the formaldehyde concentration in the vapor phase, the emission rate of a solid source is:

$$CH_2OER' \simeq -m\,[CH_2O]_V + b \qquad (4)$$

where

m = the mass transfer coefficient (m/hr)

$[CH_2O]_V$ = the CH_2O concentration in the vapor phase (mg/m^3)

b = a constant; the emission rate at zero CH_2O concentration in the air (mg/m^2hr)

Therefore, Equation 5 (Equations 3 and 4 combined) is used to calculate the concentration inside a single compartment with a solid emission source in direct contact with the air:

$$[CH_2O]_{SS} = (Area/(PEX \times VOL) \times b + [CH_2O]_0)/(1 + m\,(Area/(PEX = VOL))$$

The third type of formaldehyde source is a solid, area-dependent source with an emission barrier (such as carpet or a vinyl laminate). Again, Fick's Law describes formaldehyde transport across such permeation barriers:

$$CH_2OER' = K([CH_2O]_{BB} - [CH_2O]_{SS}) \qquad (6)$$

where

K = mass transport coefficient for the permeation barrier (m/hr)

$[CH_2O]_{BB}$ = formaldehyde concentration below the barrier (mg/m^3)

$[CH_2O]_{SS}$ = formaldehyde concentration above the permeation barrier (mg/m^3)

If it is assumed that there are no concentration gradients above or below the barrier, Equation 7 applies:

$$CH_2OER' = -m\,[CH_2O]_{BB} + b \qquad (7)$$

This simplifying assumption results in an overestimation of CH_2OER' because the presence of the barrier will in fact cause concentration gradients near the surface of the source, which would reduce CH_2OER'. The emission model for a solid source and a permeation barrier is, by combining Equations 6 and 7,

$$CH_2OER' = (b - m \times [CH_2O]_{SS})/1 + m/k) \quad (8)$$

The concentration inside a single compartment with this type of source is calculated via Equation 9:

$$[CH_2O]_{SS} = \frac{(Area/(PEX \times VOL)) \times b + [1 + (m/K) \times [CH_2O]_o]}{1 + (m/K) + m\,(Area/(PEX \times VOL))} \quad (9)$$

Equation 9 holds for barriers of intermediate efficiency in emission reduction; for very inefficient barriers (i.e., K>>1), Equation 5 applies. For extremely efficient barriers (K~0), the source may be disregarded by assuming that $[CH_2O]_{SS}$ is approximately equal to $[CH_2O]_o$.

Equations 2, 5, and 9 describe the calculation of $[CH_2O]_{SS}$ for indoor compartments with combustion, direct, and source-barrier combination sources alone. For a single compartment with multiple formaldehyde sources, Equation 10 may be used to derive the steady-state concentration $[CH_2O]_{SS}$:

$$[CH_2O]_{SS} = [CH_2O]_o + \sum_{i=1}^{n} (CH_2OER/(PEX \times VOL) + \sum_{i=1}^{v} (CH_2OER' \times Area)\,(PEX \times VOL) \quad (10)$$

with proper emitter equations (such as Equations 4 and 7) substituted for CH_2OER'.

Matthews et al. have applied this model to predict the relative importance of a variety of formaldehyde sources in an indoor compartment. The results of these analyses were discussed in Section 3.6 of this report.

7.3 Derivation of a Best-Fit Decay Curvey to Predict Long-term Levels of Formaldehyde in Homes

Numerous investigators have observed that there is a relationship between the age of a home (i.e., the age of the pressed-wood products within it) and the formaldehyde level in the air of that home. In a plot

of formaldehyde levels versus time (with time in weeks, months, or years), an exponential type of decrease, or decay, is exhibited (Meyer and Hermanns 1984a,b; Myer 1984b). This is termed slow decay, and a mathematical description of the correlation can be a valuable predictive tool for long-term estimates of exposure in residential settings. Models such as the CPSC model (discussed in Section 7.1) provide a starting point, or an initial formaldehyde level; equations derived from the decay curve function can integrate levels over extended time periods.

This section discusses the development of a best-fit decay curve for a combined data set of formaldehyde observations over time for representative (i.e., noncomplaint) homes. The combined data set is composed of two surveys, known as the Clayton study and the Wisconsin study. The following sections describe each data set and present results of statistical data analysis on each set and on the combined data set.

The best representation of the decay function is described by an exponential equation fitted to the combined data set. As described below, correlation coefficient is only one of the indicators of an equation being representative of the data. Mean square error is an important determinant as well.

7.3.1 Description of Data Sets

(1) Clayton Survey Data. This data set contains 259 observations from 259 mobile homes, in which the formaldehyde levels have been standardized for constant temperature and relative humidity. The standardized concentrations range from 0.2 to 2.9 ppm. The mean for this distribution is 0.62 ppm, and the median is 0.38. The 75th percentile value is 0.53, and the coefficient of variation is 93 percent. The distribution exhibited a marked positive skewness; taking the log transformation of the standard concentration resulted in a symmetrical normal distribution.

The home age ranges from 0 to 2894 days. Homes less than three years old had an average formaldehyde level of 0.73 ppm, while homes over three years old averaged 0.18 ppm. Information was provided on temperature and humidity conditions. Homes tested were primarily "non-complaint" homes, and QA/QC was reportedly good. Figure 25 presents a plot of the data points as mobile home age (X) versus formaldehyde concentration (Y).

(2) Wisconsin Survey Data. This data set contains 920 data points from 137 mobile homes; 56 of the original 976 observations were missing values from lost samples and similar experimental problems. The formaldehyde levels have been standardized for constant temperature and relative humidity by the Wisconsin Division of Health. The standardized concentrations ranged from 0.02 to 2.26 ppm. The mean for the distribution is 0.38 ppm, and the median is 0.3 ppm. The 75th percentile is 0.51 ppm while the 90th percentile is 0.72 ppm. The standard deviation is 0.3 ppm, and the coefficient of variation is 79 percent.

The home ages range from 30 to 3360 days. Homes less than three years old had an average formaldehyde level of 0.45 ppm, and homes over three years old averaged 0.15 ppm. Homes tested were primarily non-complaint homes. Figure 26 presents a plot of the Wisconsin data points as mobile home age (X) versus formaldehyde concentration (Y).

(3) Aggregated Data (Clayton and Wisconsin). The two data sets (Clayton and Wisconsin) were combined to obtain a new data set that has 1179 values. The concentrations range from 0.02 to 2.9 ppm. The mean for this distribution is 0.43 ppm, the median is 0.31 ppm, the 75th percentile is 0.55 ppm, and the 90th percentile is 0.89 ppm. The coefficient of variation is 90.6 percent.

The home age ranges from 0 to 3360 days. Homes less than 3 years old have an average formaldehyde level of 0.51 ppm, and homes over three years old averaged 0.15 ppm.

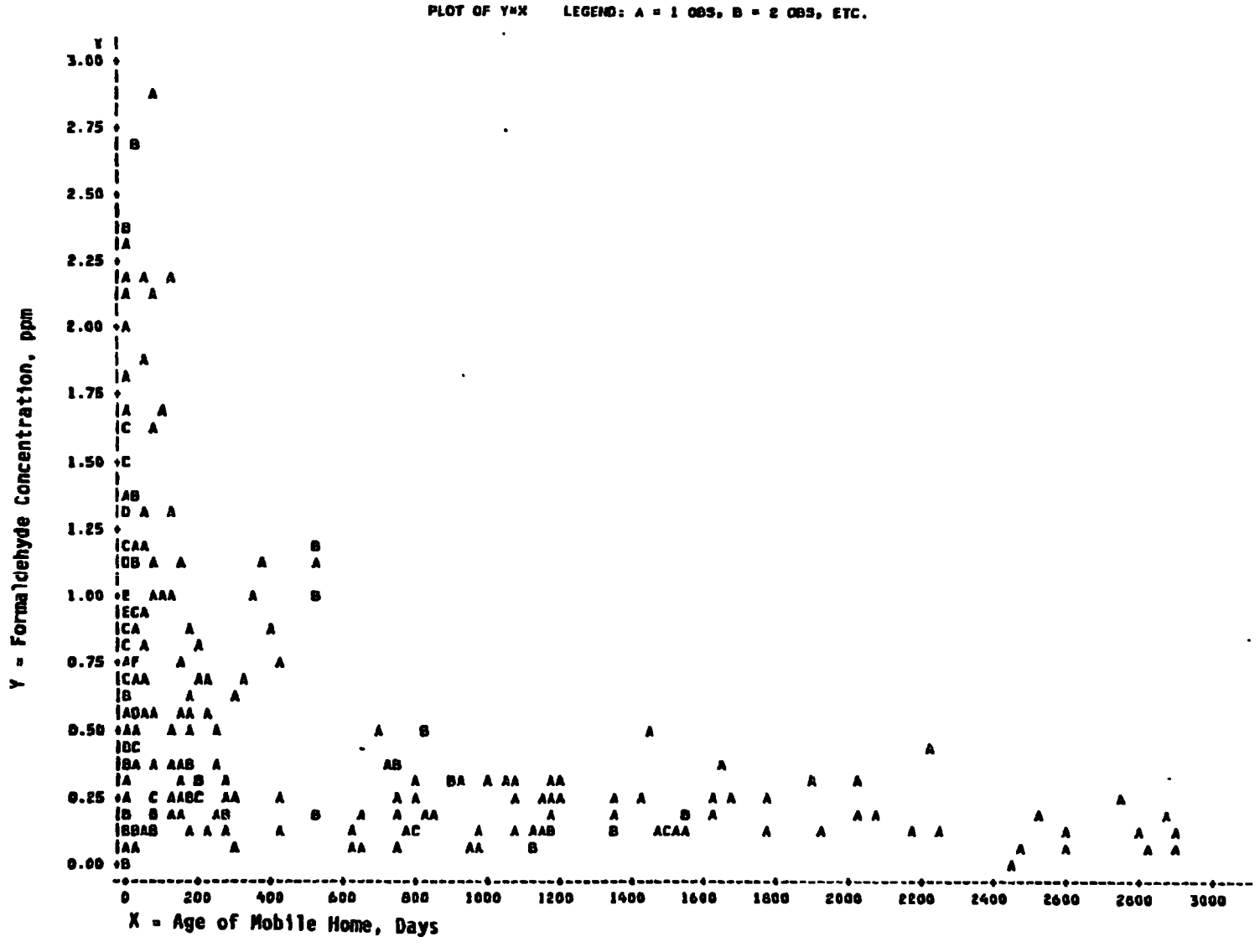

Figure 25. Plot of the Clayton Data

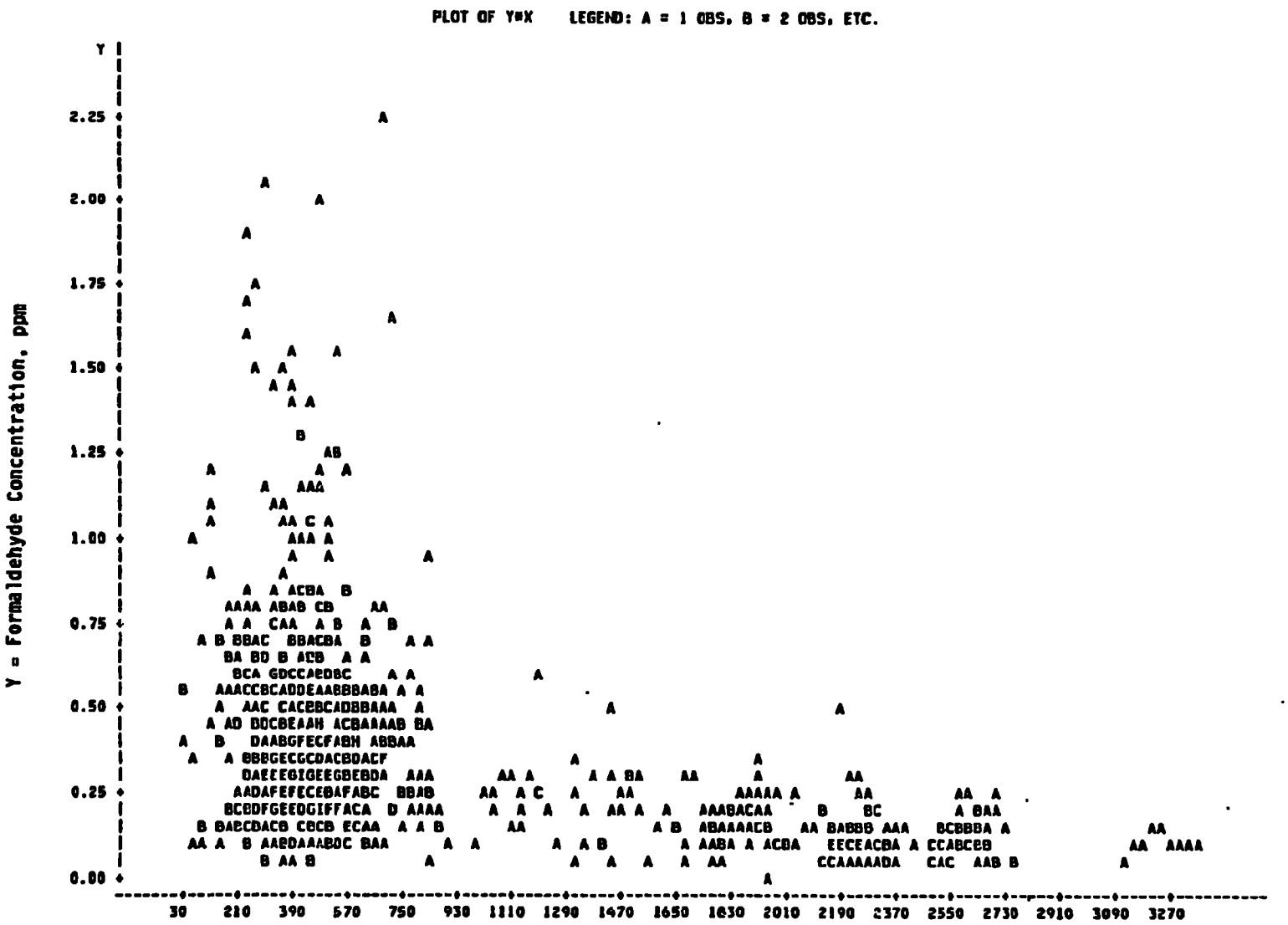

Figure 26. Plot of the Wisconsin Data

Each of the data points has the following characteristics:

- Formaldehyde was measured by the chromotropic acid method or by the pararsoaniline method.
- Each data point is the average of measurements taken on a particular day, and the age of the mobile home at the time of measurement is given.

Figure 27 presents a plot of the aggregated data points as mobile home age (X) versus formaldehyde concentration (Y).

7.3.2 Results of Statistical Analysis of the Decay Function

Based on the three plots (Figures 25, 26, and 27), it is clear that the initial average level of formaldehyde declines with the increase in the ages of homes. Consequently, the exponential and power mathematical models were evaluated to determine the function of the curve that best describes (or fits) the data.

The evaluation of these models involved developing several different statistical approaches for manipulating the data under the exponential and power laws. The description and results of these statistical evaluations are detailed in Section 7.3.3 of this report. The final determination was to select the exponential model as the best fit for the combined data. The resulting decay function follows:

$$\ln Y = -.684 - .00065X$$

$$Y = e^{-.684} e^{-.00065X}$$

$$Y = .504\, e^{-.00065X}$$

7.3.3 Statistical Analysis of Separate and Combined Data Sets

The two models considered to best fit the available data sets are the exponential and the power mathematical models.

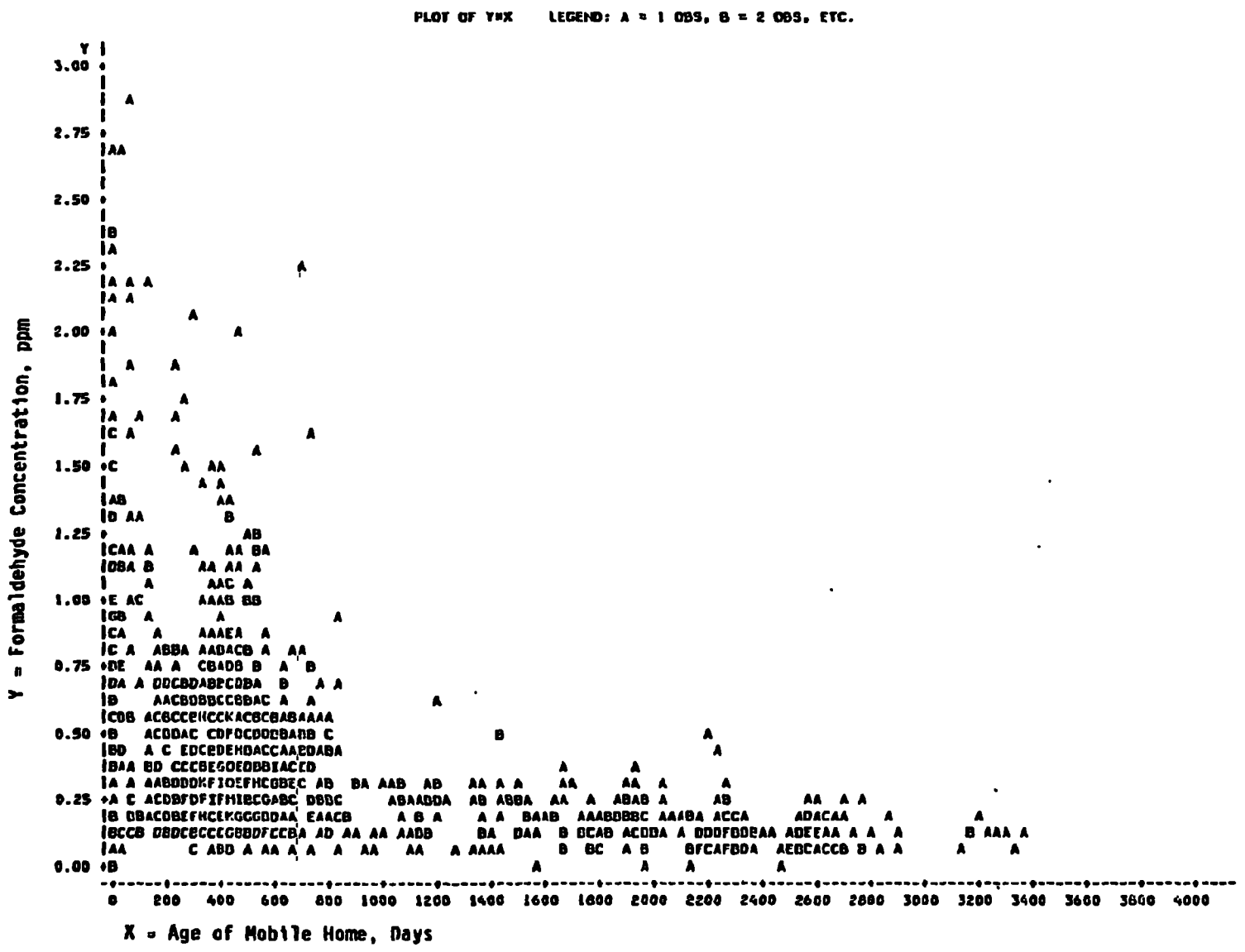

Figure 27. Plot of Combined Data Set

An exponential model for the regression of Y on X has the form:

$$y = a\, e^{bx}$$

which is transformed by taking logarithms to obtain

$$\ln y = \ln a + bx$$

setting $Y = \ln y$, $A = \ln a$, $B = b$, and $X = x$, produces the linear expression:

$$Y = A + BX$$

used in the analyses.

The power model has the form

$$Y = a\, x^{b},$$

which is transformed by taking logarithms to obtain

$$\ln y = \ln a + b \ln x$$

setting $y = \ln y$, $A = \ln a$, $B = b$, and $X = \ln x$ results in the linear expression:

$$Y = A + BX$$

used in the analyses.

In both the power and exponential forms, x equals the age of the home (in days) and y equals the measured level of formaldehyde in the indoor air (in ppm).

The nature of the data and the models considered indicated, that the data points for days 0 and 1 significantly deviated from an otherwise good fit. Consequently, both models were tested with a data set minus one or both of these days. Two basic types of statistical analyses, described below, have been performed.

(1) Analysis of Individual Observations. For all observations reported in the three data sets (Clayton, Wisconsin, and the combined data) the following three analyses were performed:

Analysis 1 - The observations that had 0 age values were excluded.

Analysis 2 - The observations that had 0 or 1 age values were excluded.

In addition, the original plot of the data set indicated that if the data set was split at what appeared to be the elbow in the curve, two linear functions, one for each subset, might best describe the entire data set. Moreover, this split could reflect the cutoff point between the two types of formaldehyde emission from pressed-wood products. Consequently, a third analysis would test both curve models and a linear model with the data split at the value X = 575.

Analysis 3 - The data set (0 and 1 omitted) was split into two parts, with regression analysis performed on each.

The results of these three analyses on the various model options are summarized below and in Table 56 and Figure 28 for the Clayton data; in Table 57 and Figure 29 for the Wisconsin data; and in Table 58 and Figure 30 for the combined data set.

- Analysis of the Clayton Data (zero age values excluded). The R^2 (the coefficient of determination) values and the mean square errors are very close for both models. The probability (0.1499) for testing the first parameter of the power model shows that it is not significantly different from 0. This means that parameter A = 1.
- Analysis of the Clayton Data (age of zero and 1 excluded). The R^2 values and the mean square errors are very close for the two models. The probabilities of each model show the significance of the two parameters.
- Analysis of the Split Clayton Data. For the first data region, the power model fits slightly better than the others. For the second data region, all the models fit equally well. The estimates for the model parameters (A, B) change dramatically from first to second data regions.

Table 56. Results of Statistical Analyses of Clayton Data

Model	Parameter A	Significance[a]	Parameter B	Significance	Goodness of fit MSE[b]	Goodness of fit R^2[c]
Analysis 1 (zero excluded)						
Exponential	-0.533	Yes	-0.00076	Yes	.66	.32
Power	0.172	No	-0.24	Yes	.67	.32
Analysis 2 (zero and 1 excluded)						
Exponential	-0.53	Yes	-0.00076	Yes	.66	.33
Power	0.47	Yes	-11.41	Yes	.63	.36
Analysis 3 (split data set)						
First data region:						
Exponential	-0.37	Yes	-0.001	Yes	.76	.05
Power	0.09	No	-0.16	Yes	.73	.09
Linear	0.44	Yes	-0.001	Yes	.35	.06
Second data region:						
Exponential	-1.41	Yes	00.0002	Yes	.34	.07
Power	0.55	No	-0.32	Yes	.34	.06
Linear	0.27	Yes	-0.00005	Yes	.01	.07

[a]Significance determined by the probability level using the T-test compared at the 95% level.

[b]Mean square error.

[c]Percent of variation explained by model.

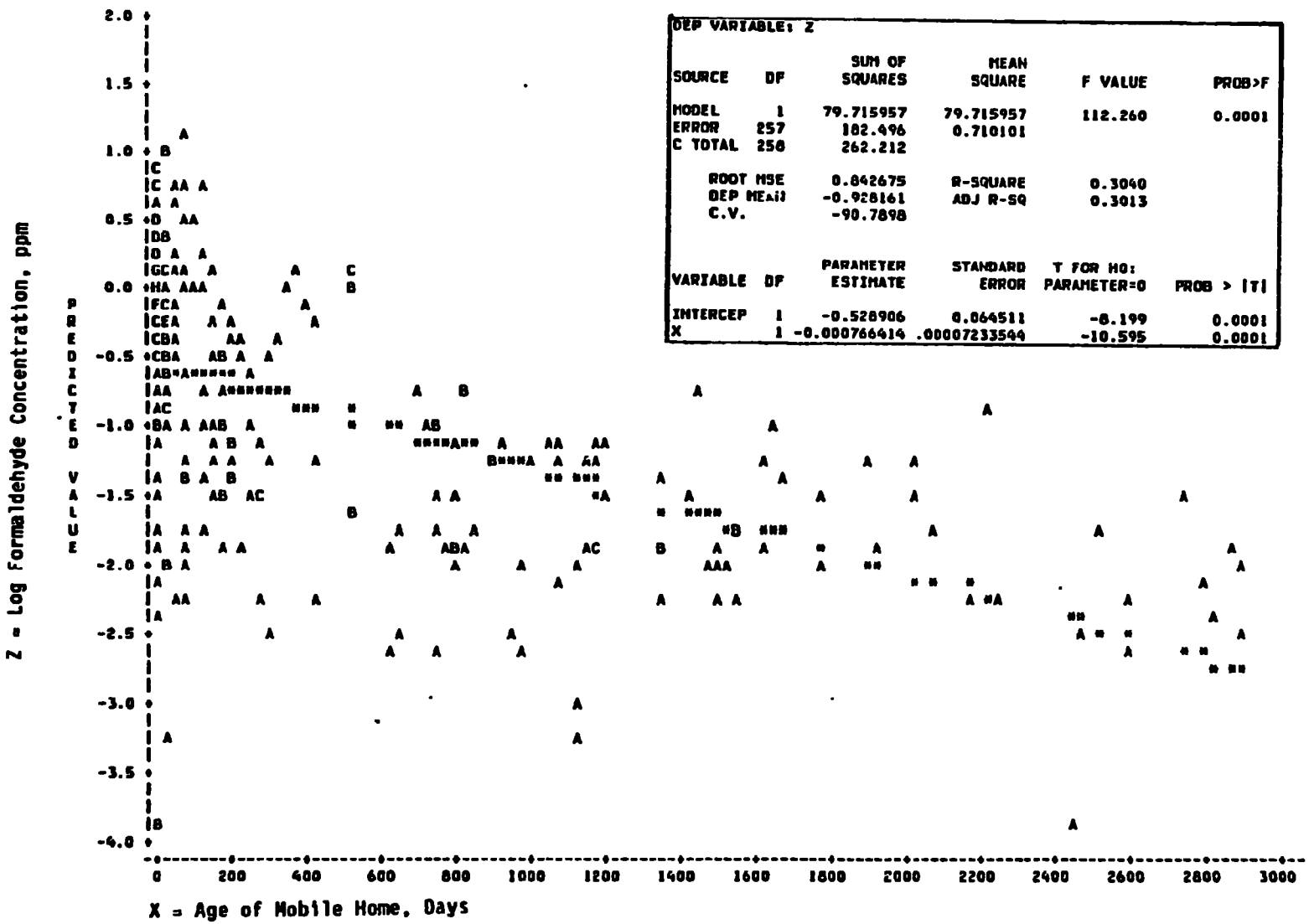

Figure 28. Regression Analysis of Clayton Data - Exponential Model

Table 57. Results of Statistical Analyses of Wisconsin Data

Model	Parameter A	Significance[a]	Parameter B	Significance	Goodness of fit MSE[b]	Goodness of fit R^{2c}
Analysis 1 (zero excluded)						
Exponential	-0.748	Yes	-0.0006	Yes	.38	.4
Power	2.14	Yes	-0.531	Yes	.41	.3
Analysis 2 (zero and 1 excluded)						
Exponential	-0.748	Yes	-0.0006	Yes	.38	.4
Power	2.14	Yes	-0.531	Yes	.41	.3
Analysis 3 (split data set)						
First data region						
Exponential	-0.887	Yes	-0.00027	No	.42	.002
Power	-0.568	No	-0.0700	No	.42	.001
Linear	0.524	Yes	-0.00017	No	.09	.002
Second data region						
Exponential	-0.732	Yes	-0.0006	Yes	.3	.4
Power	4.57	Yes	-0.869	Yes	.3	.5
Linear	0.468	Yes	-0.0001	Yes	.03	.3

NOTE: Analyses 1 and 2 are the same for the Wisconsin data; there were no 0 or 1 age values .

[a]Significance determined by the probability level using the T-test compared at the 95% level.
[b]Mean square error.
[c]Percent of variation explained by model.

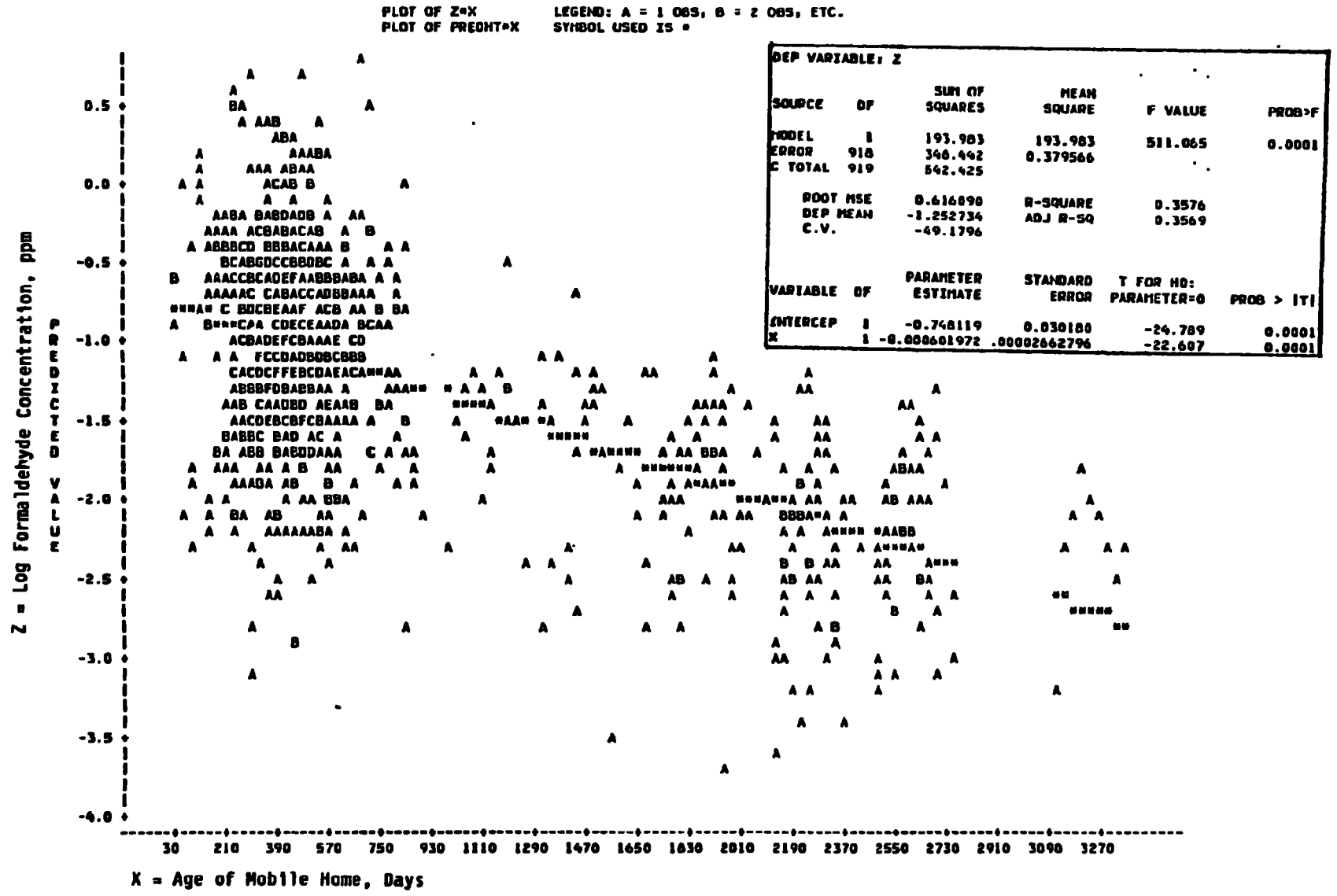

Figure 29. Regression Analysis of Wisconsin Data - Exponential Model

Table 58. Results of Statistical Analyses on the Aggregated Data Set

Model	Parameter A	Significance[a]	Parameter B	Significance	Goodness of fit MSE[b]	Goodness of fit R^{2}[c]
Analysis 1						
Exponential	-0.690	Yes	-0.00064	Yes	.44	.4
Power	0.598	Yes	-0.296	Yes	.50	.3
Analysis 2						
Exponential	-0.693	Yes	-0.00064	Yes	.43	.4
Power	0.92	Yes	-0.346	Yes	.48	.3
Analysis 3						
First data region						
Exponential	-0.54	Yes	-0.001	Yes	.50	.1
Power	0.096	No	-0.18	Yes	.49	.1
Linear	0.447	Yes	-0.00027	No	.33	.001
Second data region						
Exponential	-0.85	Yes	-0.0006	Yes	.32	.4
Power	4.03	Yes	-0.797	Yes	.32	.4
Linear	0.266	Yes	-0.000046	Yes	.07	.06

[a]Significance determined by the probability level using the T-test compared at the 95 percent level.

[b]Mean square error.

[c]Percent of variation explained by model.

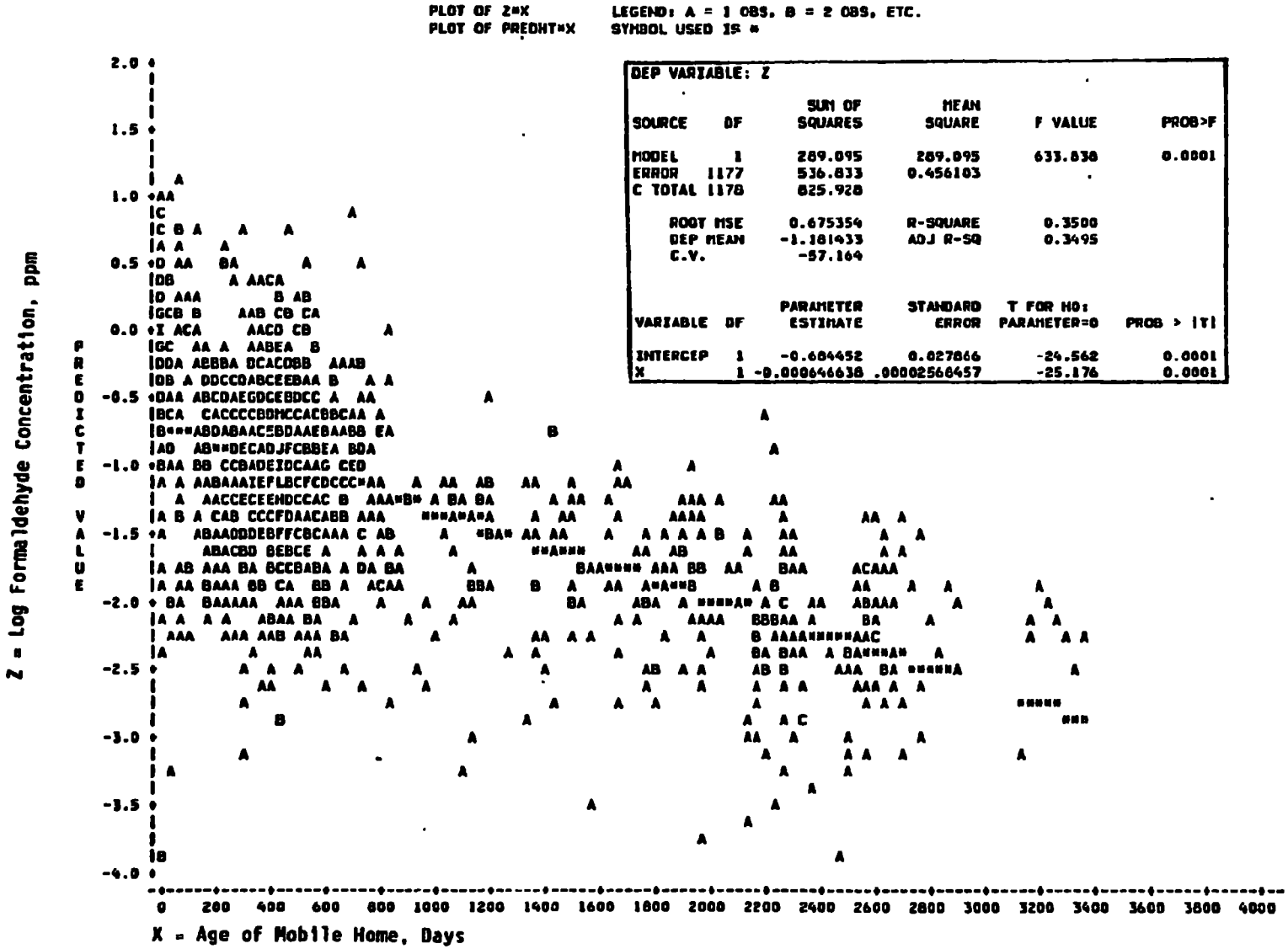

Figure 30. Regression Analysis of Combined Data Set - Exponential Model

- Analysis of the Wisconsin and Aggregated Data (zero age values excluded). The R^2 value for the exponential model of 0.4 ($r = -.6$) is larger than the R^2 for the power model of 0.3 ($r = -.5$). The mean square error for the exponential model is less than the mean square error for the power model.

- Analysis of the Wisconsin and Aggregated Data (age of zero and 1 excluded). The R^2 value for the exponential model is larger than the R^2 for the power, and the mean square error for the exponential model is less than the mean square error for the power model.

- Analysis of Split Wisconsin and Aggregated Data. For the first data region, both the models fit equally well. For the second data region, the power model fits better than the exponential.

Splitting the data set did not improve the fit for any of the models, even through using two equations for two regions should better describe the data set (the R^2 values are very small for both regions in the Clayton data and in the first region in the Wisconsin data).

From the above discussion and the results of the three analyses, the exponential curve is determined to best describe the decay of the average formaldehyde level in the mobile homes for the entire period.

(2) Analysis of Data Aggregated by Range of Age of Homes at Observation. An approach to analyzing formaldehyde decay by aggregating data was reported in Cohn (1984). In Cohn's approach, the data were grouped and the average formaldehyde level (or the mid point of each group) was used in subsequent equation derivaton. The results of this approach, as expected, showed a high value of R^2 (good fit) because variability (which decreases R^2) was reduced by averaging data ranges.

Similar analyses of the Clayton, Wisconsin, and combined data are described herein. The data are grouped by 10, 25, 50, 100, and 200 day intervals, and the average of concentration over these periods is used in the mathematical analysis (Table 59 summarizes the results). It is obvious that grouping the data creates a better fit line.

Table 59. Analysis of Data Grouped in Two Intervals

	EXPONENTIAL							POWER			
			Estimate		Std error of estimate			Estimate		Std error of estimate	
Interval, days	N	R^2	a	$b*10^{-4}$	a	$b*10^{-4}$	R^2	a	b	a	b
0	1178	.4	-0.684	-6.47	.027	2.57	.3	0.598	-.296	.089	.014
10	159	.6	-0.683	-5.96	.066	4.05	.6	1.8	-0.486	.215	.031
25	117	.7	-0.696	-5.66	.067	3.70	.7	2.185	-.535	.234	.033
50	64	.8	-0.675	-5.77	.078	4.17	.8	2.449	-.537	.276	.038
100	32	.8	-0.641	-5.74	.085	4.53	.8	2.54	-.581	.302	.042
200	17	.9	-0.611	-5.78	.099	5.04	.9	2.943	-.634	.296	.041

Table 59 shows that grouping the data reduces the number of data observations and greatly increases the standard errors of the estimates. A model with high standard errors is unreliable; furthermore, the R^2 and the estimates change with the changing of the grouping structure which shows the instability of modeling by this approach. The effect of data grouping is especially pronounced at the low age values, where formaldehyde levels range from 0 ppm to 2.9 ppm.

7.3.4 Conclusion

The exponential model of the full data set, chosen as the best description of the decay curve, has an R^2 value of 0.35. This R^2 is consistent with that shown for analysis of each data set. Since R^2 denotes the amount of variability in the values that is attributable to the variables modeled, it is apparent that the age of the home determines 35 percent of the home formaldehyde level. Other variables (type and amount of product, other formaldehyde sources, measurement error, etc.) account for the remaining 65 percent of the level. This confirms the complexity of formaldehyde emission and the factors that control it.

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