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Publication Date

1984-11-01

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Submitted to Environment International. [BL-18874]

EEB-Vent 85-7

AN EXPERIMENTAL STUDY OF AIR WASHING FOR THE REMOVAL OF FORMALDEHYDE FROM INDOOR AIR

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November 1984

This work was supported by the Assistant Secretary for Conservation and Renewable Energy, Office of Building Energy Research and Development, Buildings Systems Division and the Assistant Secretary for Environmental Protection, Safety, and Emergency Preparedness, Office of Environmental Analysis of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098 and by the U.S. Environmental Protection Agency, Office of Research and Development.

Although the research described in this article has been funded in part by the U.S. Environmental Protection Agency (EPA) through Interagency Agreement Number AD-89-F-2A-062 to the Department of Energy, it has not been subjected to EPA review and therefore does not necessarily reflect the view of EPA and no official endorsement should be inferred.

ABSTRACT

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Formaldehyde is a common indoor air pollutant that is difficult to control. One potentially suitable control technique for indoor formaldehyde is air washing: the absorption of formaldehyde by a liquid. In this report we present a mathematical model of an air washer, describe tests of two air washers, and compare the energy required for controlling formaldehyde concentrations by ventilation and by air washing. The two experimental air washers tested employed water as the washing liquid and incorporated a refrigeration system to control the humidity of the outlet airstream. Air flow rates through the air washers were 100-160 L/s and inlet formaldehyde concentrations were 80-480 ng/L. The steady-state formaldehyde removal efficiencies of the two designs were 0.36-0.47 and 0.30-0.63 with water consumption rates of 1.7-7.9 L/hr and 0.5-2.3 L/hr, respectively. The power consumption of an air washer with a 140 L/s air flow rate is estimated to be 1500-1800W. Results show that an air washer with an acceptable water requirement can effectively remove formaldehyde from indoor air. The net energy required for air washing can be less than for mechanical ventilation with heat recovery when most of the energy consumed by the air washer provides useable space heat.

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INTRODUCTION '

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Particleboard, medium density fiberboard, some plywood, and ureaformaldehyde foam insulation are common building materials that are manufactured from resins of which formaldehyde is a major component. These materials typically emit formaldehyde, often for extended periods of time, into the surrounding air. Since they are frequently found in the built environment in substantial quantities, their emissions can lead to significant formaldehyde concentrations indoors. Because there is concern about the adverse health effects of exposure to formaldehyde and since human exposure occurs primarily indoors (National Research Council, 1981a) there has been a move to establish indoor formaldehyde standards. The American Society of Heating, Refrigeration, and Air-conditioning Engineers (ASHRAE) has recommended a 120 ng/L (25°C, 1 atm) maximum indoor concentration (ASHRAE,1981). (This concentration is equivalent to 97.8 parts per billion by volume.) Recent studies have, however, found that residential indoor formaldehyde levels frequently exceed this guideline (Hawthorne et al., 1983; Colombe et al., 1983). Formaldehyde concentrations above 1200 ng/L have been measured in some mobile homes (National Research Council, 1981b).

Indoor formaldehyde concentrations are best controlled by limiting the quantity of formaldehyde sources in the indoor environment or by reducing the emissions from these sources. Some effort has been

directed toward the development of coatings, for sources such as pressed wood products, that inhibit formaldehyde release (Fisk et al., 1984). Emission rates from particleboard have also been reduced by modifying the resins and the manufacturing process (Meyer and Hermanns, 1984).

At present, ventilation is the most readily available formaldehyde control technique for existing buildings with unacceptable formaldehyde concentrations. Ventilation, the exchange of indoor air with outdoor air, causes formaldehyde to be removed from the indoor space since the concentration of formaldehyde in outdoor air is normally very low. The amount of energy required to heat or cool the fresh, outdoor air may be quite significant, however, even when ventilation systems that incorporate heat recovery are used.

Another method of reducing high formaldehyde concentrations in existing buildings is air cleaning, the separation and removal of formaldehyde from the air. There are two major air cleaning processes potentially suitable for indoor·formaldehyde control: chemical adsorption and absorption. In a chemical adsorption process, formaldehyde attaches to the surface of a solid material and then reacts chemically with the material. The results of limited studies with commercially available chemical adsorbents indicate that they \cdot are capable of reducing indoor formaldehyde concentrations but that they quickly become saturated and require replacement (Fisk et al.,

1984). In an absorption air cleaning process, also referred to as air washing, formaldehyde is dissolved in a washing solution. The washing process takes place in an air washer, a device which provides a large interface area between the washing solution and a formaldehydecontaminated airstream. This technique is potentially suitable for use in residential as well as commercial and industrial environments.

To investigate the feasibility of air washing we designed and fabricated two full-scale air washers and evaluated their performance. In this report we present (1) a mathematical model of an air washer, (2) a description of the experimental air washers and our method of evaluating their performance, (3) the results of these tests, and (4) a comparison of the energy required for controlling formaldehyde concentrations by ventilation and by air washing. Additional information is presented in another report (Pedersen and Fisk, 1984a).

AIR WASHER MODEL

The model presented below reflects the design of the experimental air washers (described in the next section). The washing solvent was water, which has a very high capacity for dissolved formaldehyde. The experimental air washers contained a large quantity of water which was recirculated through an airstream so that the concentration of formaldehyde in this washing solution was essentially uniform throughout the air washer. A portion of the washing solution was

continuously replaced with fresh, formaldehyde-free water to prevent saturation of the solution with formaldehyde.

Figure 1 is a schematic of the control volume employed for derivation of the one-dimensional model. The formaldehyde mass balance equation for the element shown is

$$
Q C(x) - [Q C(x) + (dC/dx)dx] = [C(x)-C_e](h_dA/L)dx
$$
 (1)

where: $Q =$ volumetric air flow rate,

- $C(x) =$ concentration (mass/volume) of formaldehyde in air a distance x from the airwasher inlet,
- \mathbf{c}_{e} concentration of formaldehyde in air that would be = in equilibrium with the washing solution,

A = total air-solution interface area,

Ł length of air washer in the x direction, and

 $h_{\rm d}$ = mass transfer coefficient.

The term Ce accounts for the concentration of formaldehyde in the solution. We have assumed C_{e} to be a constant. This is equivalent to assuming that the solution in the sump is well-mixed and that the formaldehyde concentration of the solution does not increase significantly as the solution passes through the airstream. The experimental results and additional calculations have shown these to be valid assumptions for the two air washers tested.

The formaldehyde removal efficiency of an air washer, ε , is defined to be

$$
\varepsilon_1 = (C_{in} - C_{out}) / C_{in}
$$
 (2)

where: C_{in} = concentration of formaldehyde in air at the air washer inlet

> c_{out} = concentration of formaldehyde at the air washer outlet.

By solving Equation 1 for Cout we obtain

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$$
\varepsilon_{1} = \left[1 - \exp\left(-\frac{h_{d}A}{Q}\right)\right] \left(1 - \frac{C_{\rho}}{C_{in}}\right). \tag{3}
$$

From Equation 3 it may be seen that the formaldehyde removal efficiency is limited by C_{e} . The quantity C_{e} can be related to the concentration of dissolved formal dehyde in the washing solution, C_5 , by Henry's Law

> $C_e = K(T)C_s$ (4)

where K(T) is a proportionality constant dependent on temperature. Data for K(T) at five temperatures in the range 5-25°C (Anthon, Fanning, and Pedersen, 1985) was fit to the following expression (r^2) $= 0.96$)

$$
K(T) = 0.97 \exp(24.33 - 6560/T) \tag{5}
$$

where $K(T)$ and T have the units $(ng/L)/(mg/L)$ and degrees Kelvin, respectively. The quantity C_S depends on the rate at which formaldehyde is removed from the air and the rate at which the washing solution is replaced with fresh water

$$
C_{S} = \varepsilon_{1} C_{in} Q/R
$$
 (6)

... .

where R is the solution replacement rate.

It is useful to define a second, "air washer device efficiency" which is a function of the air washer design but independent of the solution replacement rate

$$
\varepsilon_{2} = (C_{in} - C_{out}) / (C_{in} - C_{e}). \qquad (7)
$$

Again from Equation 1 we have

$$
\varepsilon_{2} = 1 - \exp(-h_{d}A/Q). \tag{8}
$$

Note that this device efficiency is the first term in the formaldehyde removal efficiency expression (Equation 3). The second term in Equation 3 describes the effect of the driving potential for mass transfer on the formaldehyde removal efficiency.

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Air Washer Desian

The two experimental air washers utilized the same case to hold the air-solution contact arrangements and additional common components. The insulated stainless steel case, shown diagrammatically in Figure 2, included a chamber for air-solution contact (0.71 x 0.56 x 0.56 m) and a sump (30 L capacity). Washing solution was removed from the sump by a constant-flow, adjustable rate pump. Fresh, make-up water was provided by a city water connection to the case. The inflow of water was controlled by a float valve so that a constant solution level was maintained in the sump. The case also included the evaporator and condenser of a 4.0 kW (output) refrigeration system to control the humidity of the air washer airstream. Contact between air and water at room temperature would humidify the air, an undesirable consequence. By cooling the airstream prior to contact with the washing solution, however, the humidity of the exiting airsteam was controlled. The heat rejected from the refrigeration system's condenser was returned to the airstream after the washing process. In actual use this heat would, consequently, be provided to the indoor space.

The two air washers'were distinguished by their air-solution contact arrangements. The contact arrangement for Air Washer No. 1, shown in

Figure 3a, consisted of either three or four rotating mats through which the airstream passed. These porous foam mats were kept wet by rotating them through the solution in the sump. The contact arrangement of Air Washer No. 2, shown in Figure 3b, was based on a commercially available mass transfer media (Munters, Model GS XF 6560/15). A pump continuously circulated solution from the sump to solution distribution pipes above the media.

Test System and Procedure

The formaldehyde removal performance of the experimental air washers was evaluated by supplying an airstream with a controlled formaldehyde concentration to the air washer and measuring the inlet and outlet formaldehyde concentrations. The test system (including the formaldehyde measurement apparatus) is only briefly described here but is discussed in detail by Pedersen and Fisk (1984b). Gaseous formaldehyde was introduced to the airstream by continuous evaporation of a methanol-free aqueous formaldehyde solution that was delivered by a syringe pump. A blower supplied the temperature- and humiditycontrolled airstream (70-160 L/s) to the air washer through a duct (0.15 m diameter). The formaldehyde concentration of the air was measured by drawing sample airstreams through chilled, water-filled impingers and subsequently analyzing the water by the modified pararosaniline method (Miksch et al., 1981). This integrating formaldehyde measurement system was calibrated before and after the

series of tests with a formaldehyde calibration system (Geisling, Miksch, and Rappaport, 1982). The air flow rate through the air washer was measured with a calibrated orifice plate flow meter mounted in the duct upstream of the air washer (American Society of Mechanical Engineers, 1971). The air temperature and humidity of the inlet and outlet airstreams were meqsured by calibrated sensors and recorded on chart recorders. Sensor readings were compared periodically to precision wet and dry bulb thermometer measurements.

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The test procedure was designed to evaluate the air washers under steady-state conditions. Steady-state occurs when the rate of formaldehyde removal from the air equals the rate of formaldehyde removal from the sump by the washing solution replacement process. Prior to steady-state operation the formaldehyde concentration of the solution is lower than the steady-state value, thus, as may be seen from Equation 3, the formaldehyde removal efficiency is artificially high. To allow the concentration of formaldehyde in the washing solution to increase to approximately its steady-state value, the air washer was first operated for a period of time without any replacement of washing solution. The test was then initiated by starting the solution replacement process and the measurements of formaldehyde concentration in air. Typically, these formaldehyde measurements took place over an 8 to 16 hour period. A sample of washing solution was drawn from the sump at the beginning and end of each test and its formaldehyde concentration determined, also by the pararosaniline

method. These measurements permitted us to determine if the air washer was operating at steady-state and also made possible corrections of the measured formaldehyde removal efficiencies for tests performed when steady-state had not been attained.

For tests of both air washers, relevant parameters were varied to assess their impact on performance. To demonstrate that the materials from which the air washers were fabricated did not, at steady-state, remove formaldehyde from the air, background tests were run with each air washer. To conduct these tests, the washing solution was removed and the refrigeration system was not operated.

DATA ANALYSIS

The formaldehyde removal efficiencies of the two experimental air washers were calculated from Equation 1. However, since steady-state operation was not achieved for most tests, weal so calculated ε*, the corrected, i.e., steady-state, formaldehyde removal efficiency, by applying the theoretical model and data on C_{s} , the concentration of formaldehyde in the washing solution.

The model was also employed to calculate theoretical parameters from the experimental results including the device efficiency, the mass transfer coefficient-interface area product, h_dA . $\varepsilon_{\frac{1}{2}}$, and Three additional parameters calculated for each test are the ratio $\epsilon \nless \epsilon$,

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a measure of the impact of C_e on $\epsilon_1^{\star}; \, \mathbb{Q}_{\mathsf{C}},$ the effective clean-air flow rate; and M, the formaldehyde mass balance ratio. The effective clean-air flow rate is defined as the product of $\begin{smallmatrix} \varepsilon^{\star} & \varepsilon^{\star} \\ 1 & \end{smallmatrix}$ and the air flow rate, Q. This parameter represents the equivalent flow of formaldehyde-free air that is provided by an air washer. It is a particularly useful quantity for comparing the rate of formaldehyde removal by an air washer to the rate of formaldehyde removal by a given amount of ventilation. The numerator of the mass balance ratio equals the mass of formaldehyde contained in the solution within the air washer at the end of the test plus the total mass of formaldehyde in the air and the solution that exited the air washer during the test. The denominator of the mass balance ratio equals the mass of formaldehyde in the solution that is within the air washer at the start of the test plus the total mass of formaldehyde in the air that enters the air washer during the test. This mass balance ratio is an indicator of the quality of the formaldehyde concentration measurements and will be unity if all formaldehyde is "accounted" for.

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The estimated maximum uncertainties in the calculated parameters are shown in Table 1. The procedure employed to estimate uncertainty is described in another repott (Pedersen and Fisk 1984a).

RESULTS AND DISCUSSION

Test Results

The test results and test condition data are listed in Tables 2 and 3 for Air Washers No.1 and No.2, respectively. The corrected for- . maldehyde removal efficiencies of Air Washer No. 1 ranged from 0.36 to 0.47. There was little difference between the measured and corrected formaldehyde removal efficiencies for all but two of the tests. This indicates that, with two exceptions, the performance of this air washer was evaluated under essentially steady-state conditions. The effective clean-air. flow rates were 41 to 57 L/s. For all tests of this air washer, the values of $\varepsilon_1^{\star}/\varepsilon_2^{\star}$ were high (0.64-0.95) because the washing solution replacement rates were high. Thus, the efficiency of this air washer was not reduced substantially by the concentration of formaldehyde in the washing solution. Instead, the efficiency was limited by the physical design of the air washer, i.e., the limited interface area per unit air flow rate.

The corrected formaldehyde removal efficiencies for tests of Air Washer No.2 were 0.30 to 0.63. The measured formaldehyde removal efficiencies were considerably higher, 0.50 to 0.77, indicating that this air washer was not tested under steady-state conditions and that the solution replacement rates were too low for optimal performance. By increasing the solution replacement rate, however, steady-state

removal efficiencies equal to or even greater than the measured efficiencies could be achieved. This is illustrated in the next section by analysis with the theoretical model. The effective clean-air flow rates for the second air washer were 35 to 74 L/s. The various performance indicators had lower values for test 2-8 because one-half of the mass transfer media had been removed from the air washer.

The performance of Air Washer No. 2 was superior to that of Air Washer No. 1. It should be noted that the similiar or higher formaldehyde removal efficiencies were achieved with generally lower solution replacement rates. This was possible because the device efficiencies, ε , of Air Washer No. 2 were much greater, evidence that the product of hd and A for this air washer was greater than that for the first air washer.

The background formaldehyde tests of both air washers showed that there was negligible formaldehyde removal when water was not present in the air washer, therefore, all formaldehyde removal may be attributed to the air washing process.

The mass balance ratios, M, were within 11% of unity with only two exceptions. This is evidence, but not proof, that the measurements were generally accurate. The cause of the two instances of a poor mass balance ratio (0.81 and 1.40) was not determined.

Results of Analysis Using the Theoretical Model

While the theoretical model is relatively simple and cannot account for many complexities of air washer performance, further analysis using the model provides results that are useful for evaluation of air washing and for air washer design optimization. Figure 4, for example, shows the relationship between formaldehyde removal efficiency and solution replacement rate .at various solution temperatures. The curves are for an air washer with an air flow rate of 140 L/s and a device efficiency of 0.90. As the solution replacement rate, R, is increased, the formaldehyde removal efficiency, ε _,, increases. However, ϵ_{1} is limited to 0.90 since the device efficiency, ϵ_{2} , is 0.90. A solution replacement rate of 6 L/hr (a reasonable rate) is required for $\varepsilon_{_1}$ to be 0.80 when the solution temperature is 2°C.. Also shown by Figure 4 is the effect of solution temperature on solution replacement rates. In addition to permitting humidity control, reducing the water temperature also reduces water consumption substantially.

The relationship between the effective clean-air flow rate and the air flow rate through the air washer for various values of h_dA is shown in Figure 5. As the air flow rate increases, the effective clean-air flow rate asymptotically approaches the value of h_dA (assuming h_dA is constant). Increasing the air flow rate will also increase the power required to mmove and cool the air, however, so a compromise must be made to optimize efficiency and power consumption.

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Power and Water Consumption

The power requirement of an air washer with an air flow rate of 140 L/s is estimated to be 1500-1800 W. This includes power for operation of the refrigeration system, the solution circulation and replacement pump, and the fan. Based on the experiments performed and the theoretical analysis, such an air washer could potentially have a formaldehyde removal efficiency of at least 0.80. A removal efficiency of 0.80 would require a washing solution replacement rate of approximately 6 L/hr. With year-round operation, 5.2×10^4 L/yr of water would be required. This water usage would increase the water consumption of a typical residential consumer by 14% and cost an average of \$14/yr (American Waterworks Association, 1983).

Comparison of the Energy Requirements of Ventilation and Air Washing

At present, ventilation is the most readily available formaldehyde control technique for existing residences with unacceptably high formaldehyde concentrations. Ventilation of indoor spaces may be provided naturally, by leakage of air through cracks or other openings in the building envelope, or in a more energy-efficient manner with a mechanical ventilation system which incorporates an air-to-air heat exchanger (MVHX system). As noted previously, however, energy will be required to heat the ventilation air during the heating season. Additionally, in the case of the MVHX system, energy will be required

to operate the fans. A significant amount of energy will be required to operate an air washer, however, the air washer will reject heat to indoors and, therefore, descrease the amount of heat that must be supplied by the building's heating system.

To compare the energy requirements of natural ventilation, operation of an MVHX system, and operation of an air washer, a simple energy analysis was performed for residences located in two different climates, those of Minneapolis, Minnesota and Chicago, Illinois. Ventilation or air washing was assumed to be continous during a sevenmonth heating season. The ventilation and air washer effective cleanair flow rates were chosen to be. 90 L/s. For an air washer with a 140 L/s air flow, a 90 L/s effective clean-air flow rate is expected if the formaldehyde removal efficiency is 0.8 and the "ventilation efficiency" is 0.8. This so-called ventilation efficiency accounts for the imperfect mixing of air indoors which could cause the concentration of formaldehyde in air entering the air washer to be less than the average indoor concentration. This factor has also been accounted for in the energy requirement calculations for the two ventilation alternatives by assuming that heat and formaldehyde were removed from the building with the same efficiency. The energy requirements of the two ventilation strategies were calculated using data by Fisk and Turiel (1983). The power required to operate the air washer was taken to be 1800 W. The energy requirements of each control measure are listed in Table 4. The MVHX system requires the least energy of the 3

alternatives in both cities, however, this does not account for the heating load offset resulting from heat produced by the air washer.

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Using weather data (Nicholson, 1978) we calculated the fraction of the energy consumed, i.e., heat produced, by the air washer that would substitute for heat normally provided by the heating system in a wellinsulated residence. In Minneapolis and Chicago, these fractions are 0.82 and 0.76, respectively. As can be seen from Table 4, when this heating offset is accounted for, the net energy required for air washing is less in both climates than the energy required for either method of ventilation. This energy comparison is valid only for electrically-heated residences. In buildings which are heated using energy that is less expensive than electricity, the reduction in. heating load offset caused by air washer operation is less advantageous.

We have not assessed the economic feasibiblity of air washing. Fisk and Turiel (1983) have shown residential MVHX systems to be economically attractive (from a homeowner's perspective) compared to ventilation without heat recovery primarily in colder climates and in buildings heated using expensive forms of energy. While the net energy required for of an air washing may be less than that required by MVHX systems, an air washer would probably have higher initial and maintenance costs than an MVHX system. Further study is necessary before an accurate economic comparison can be made.

CONCLUSIONS

This study shows that an air washer can effectively remove formaldehyde from indoor air. Higher formaldehyde removal efficiencies could be achieved with future designs. Air washing is most attractive, compared to ventilation with heat recovery, when most of·the energy required by the air washer provides usable heat and when the structure is heated with electricity or some other expensive form of energy. The water consumption of an air washer is reasonable.

We have not attempted to predict the impact of air washer operation on indoor formaldehyde concentrations. The relationships between formaldehyde source strengths, removal rates, and indoor concentrations are complex and variable. In many cases, the formaldehyde source strength will increase significantly as the indoor concentration is reduced, therefore, large amounts of ventilation or air cleaning are required to substantially reduce indoor formal dehyde concentrations. Future investigations of air washing or other air cleaning techniques for formaldehyde control should be directed toward developing air cleaners with even larger clean-air flow rates, as well as lower power requirements, than the devices described here. Finally, study is needed to assess potential adverse effects of air washer operation, particularly, the possible formation of microorganisms in the air washer.

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ACKNOWLEDGMENTS

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This. work was supported by the Assistant Secretary for Conservation and Renewable Energy, Office of Building Energy Research and Development, Buildings Systems Division and the Assistant Secretary for Environmental Protection, Safety, and Emergency Preparedness, Office of Environmental Analysis of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098 and by the U.S. Environmental Protection Agency, Office of Research and Development.

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Table 1. Estimated Maximum Uncertainty in Test Results.

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Table 2. Results of Tests of Air Washer No. 1.

Footnotes are shown at bottom of Table 3.

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Table 3. Results of Tests of Afr Washer No. 2.

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 $\frac{\varepsilon}{1}$ is measured formaldehdye removal efficiency, $\frac{\varepsilon^{\star}}{1}$ is corrected formaldehyde removal efficiency, and 3 ε is device efficiency.

4 Mass transfer coefficient-interface area product.

5 Perfect value is unity. Not available for all tests.

 6 The results of these tests should be considered less accurate than the results of the other tests due to irregularities during the experiments.

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Table 4. Coaparison of the Energy Requirements of Ventilation and Air Washing.

¹Mechanical ventilation system with an air-to-air heat exchanger.

2 Fraction of air washer energy consumption that will offset building heat load.

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Figure 1.

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Control volume employed for derivation of air washer model. Nomenclature: Q is the air flow rate, C(x) is the formaldehyde concentration in air at position x , C_{e} is the concentration of formaldehyde that would be in equilibrium with the washing solution, hd is the mass transfer coefficient, and A is the total air-solution interface area.

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Figure 3a. Air-solution contact arrangement for Air Washer No. 1. Three or four rotating mats were employed simultaneously.

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Figure 3b. Air-solution contact arrangements for Air Washer No. 2. Except for one test, the media depth in the direction of air flowwas 0.61 m and four solution distribution pipes were employed ..

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Figure 4. Theoretical relationships between formaldehyde removal efficiency and washing solution replacement rate for various solution temperatures.

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Figure 5.

Theoretical relationship between effective clean air flow rate and air washer air flow rate for various values of the mass transfer coefficient-interface area product, h_{dA}.

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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