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ABSTRACT

When infiltration and ventilation of a building are reduced in order to achieve energy savings, indoor air quality might deteriorate. Formaldehyde and other aldehydes, important indoor air contaminants, were studied as a function of building air exchange rates in public buildings and energy-efficient research houses. Sequential gas bubbling systems were used in conjunction with a pneumatic flow control system for field sampling. Samples were collected simultaneously for formaldehyde and total aliphatic aldehydes, and were analyzed in the laboratory using modified chromotropic acid, pararosaniline and MBTH methods. At public buildings, it was found that concentrations of formaldehyde and aldehydes were about the same in indoor and outdoor air due largely to traditionally high ventilation rates in these buildings. However, it is evident that indoor air in general has higher formaldehyde and total aliphatic aldehyde levels than outdoor air. Residential buildings and office trailers, in particular, have formaldehyde and total aliphatic aldehyde concentrations that can exceed the promulgated European indoor formaldehyde standard of 120 $\mu g/m^3$ ($\sim$100 ppb). Since these high air contaminant levels might have adverse health and comfort effects on building occupants, further study is needed to establish ventilation requirements for energy-efficient building designs.

Keywords: air sampling, aldehydes, building materials, chromotropic acid, energy conservation, energy-efficient, formaldehyde, houses, indoor air quality, infiltration, MBTH, pararosaniline, public buildings, residential buildings, ventilation, ventilation rate

INDOOR/OUTDOOR MEASUREMENTS OF FORMALDEHYDE AND TOTAL ALDEHYDES

INTRODUCTION:

Substantial savings in building energy use can often be achieved by reducing infiltration and ventilation rates. In doing so, however, indoor air quality might deteriorate because reductions in infiltration and ventilation can cause a buildup of indoor generated air contaminants. Since people typically spend more than 80% of their time indoors, the potential deterioration of indoor air quality might have adverse effects on their health and comfort. Therefore, indoor and outdoor air was monitored as a function of building air exchange rate. Formaldehyde (HCHO) and other aldehydes are significant indoor air contaminants since they are generated by occupant activities, such as cooking and smoking, as well as by outgassing from building materials [1]. Techniques for sampling and analyzing HCHO and total aliphatic aldehydes are discussed, as well as the initial results of measurements taken of indoor and outdoor air.

SAMPLING:

A simple, but effective, flow control system has been developed for air sampling [2]. The system (see Figure 1) provides a constant and precise (better than +3%) air flow rate as low as 0.1 standard liters per minute (slpm). This was achieved by using a pneumatic flow controller which maintains a constant pressure difference across a fine needle valve opening. By adding a constant vacuum regulator downstream, adjacent to the needle valve, the immediate upstream and downstream valve pressures are kept constant. Thus, at a given temperature and at a preselected valve opening, the system samples air at a constant mass flow rate, regardless of any changes in pressure associated with the sample collecting device. Such pressure changes often occur in the field, e.g., due to the clogging and/or non-uniformity among gas dispersing frits during sequential sampling, and can give erroneous results when a simple critical orifice alone is used to control the flow rate.

In the field, air samples are collected with four automatic sequential sampling systems using gas bubblers (shown in Figure 1). Each polypropylene bubbler (100 ml capacity) contains either 35 ml of 0.05% MBTH (3-methyl-2-benzothiazolinone hydrazone) solution or distilled water. Air is drawn into the bubblers through a coarse, sintered glass frit (70 to 100 micron pore size) at a flow rate of 0.94 slpm. The air is then passed successively through a cold trap, a pair of 47 mm filters and 3/8" tubing, into the flow control system described above. This system is located in a mobile laboratory where it is maintained at a constant temperature. Samples are collected simultaneously in water and

MBTH solutions at one indoor and one outdoor location. Air is sampled continuously for an eight-hour sampling period during the day, and a sixteen-hour period during the night.

ANALYSIS:

Samples are sealed after collection in the bubbler tubes and refrigerated until they are returned to the laboratory for analysis. Samples in MBTH solutions are oxidized to form a blue dye and measured spectrophotometrically for the total aliphatic aldehyde concentration using the MBTH method, recommended by the Intersociety Committee [3]. In order to compare different techniques for measuring formaldehyde and total aliphatic aldehydes, each sample collected in water was analyzed using the MBTH*, chromotropic acid (1,8-dihydroxynaphthalene-3,6-disulphonic acid)[4], and pararosaniline [5,6] methods. Using these three methods, the results agreed to within 10% for the same water sample. Samples collected simultaneously in MBTH gave higher concentrations (about 37% higher). It appears that MBTH, through its chemical reactivity, collects total aliphatic aldehydes as they pass through the solution, while water efficiently captures only formaldehyde. When taking an eight-hour sample using 1 slpm flow rate, these wet chemical analytic techniques are applicable to the analysis of formaldehyde (pararosaniline and chromotropic acid methods) and total aliphatic aldehydes (MBTH method) in indoor air, where concentrations are generally greater than 30 micrograms per cubic meter of air ($\mu g/m^3$). This sampling period can be shortened if a higher concentration is expected. The sampling period could also be shortened by increasing the sampling flow rate; however, this might result in a reduction in collection efficiency**.

Currently, the wet chemistry techniques are the only practical methods for determining formaldehyde and total aldehydes in air. However, there are disadvantages in using these techniques. The MBTH method is not specific for the analysis of formaldehyde although it is extremely sensitive. The chromotropic acid method, recommended by the National Institute of Occupational Safety and Health (NIOSH)[4], is the most widely used technique in the U.S. However, this technique often produces both nonspecific and nonreproducible results. The pararosaniline method is extremely inconvenient and somewhat hazardous, because of the difficulties involved in the disposal of the mercury reagent. Improvements on analytic techniques are urgently needed. Studies in progress include the comparison of various techniques for the analysis of formaldehyde and total aliphatic aldehydes [7], and optimization of wet chemistry techniques for formaldehyde analysis [8].

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*MBTH is added after the sample is collected in water.

**formaldehyde collection efficiency is about 80% in each bubbler at 1 slpm.

RESULTS AND DISCUSSION:

Total aliphatic aldehydes and formaldehyde have been measured at several occupied and unoccupied energy-efficient research houses at various geographic locations in the U.S. It has been determined that at low ventilation rates of 0.3 air changes per hour (ach) or less, the indoor formaldehyde and aldehyde concentrations can exceed the promulgated European indoor formaldehyde standard [9] of 120 $\mu g/m^3$. Figures 2, 3 and 4 illustrate these results. The outdoor concentrations were typically 20 $\mu g/m^3$ (16 ppb) or less.

At a public school in Columbus, Ohio, and a large medical center in Long Beach, California, indoor and outdoor formaldehyde and aldehyde concentrations were similar, being well below the 120 $\mu g/m^3$ standard. The results are shown in Figures 5 and 6. High ventilation rates in these public buildings are the probable reason for the similar, low indoor and outdoor concentrations.

Indoor and outdoor air has also been sampled at several new office trailers. Occupants of these units have reported odor problems. Formaldehyde concentrations in these trailers ranged from 137 to 251 $\mu g/m^3$, compared to 10 $\mu g/m^3$ or lower outdoors under typical spring weather conditions. In one of the trailers studied, the ventilation rate was increased from 0.16 to 0.35 ach, and the formaldehyde concentration was reduced from a range of 190 to 212 $\mu g/m^3$ to a range of 99 to 126 $\mu g/m^3$ after this increase in the ventilation rate. Outgassing of HCHO from construction materials (particleboard and plywood) using formaldehyde resins are thought to be the cause of high indoor pollutant levels in these office trailers.

CONCLUSIONS:

It is evident that indoor air, in general, has higher formaldehyde and aldehyde levels than outdoor air. The results show that residential buildings and office trailers have indoor formaldehyde and aldehyde concentrations that can exceed known health effect thresholds [10]. Formaldehyde and other aldehydes, like other indoor air pollutants, are functions of indoor activities, building materials, and building design—especially the ventilation rate. Further study is required to establish energy-efficient building designs that will assure indoor air quality adequate to preserve the health and comfort of building occupants.

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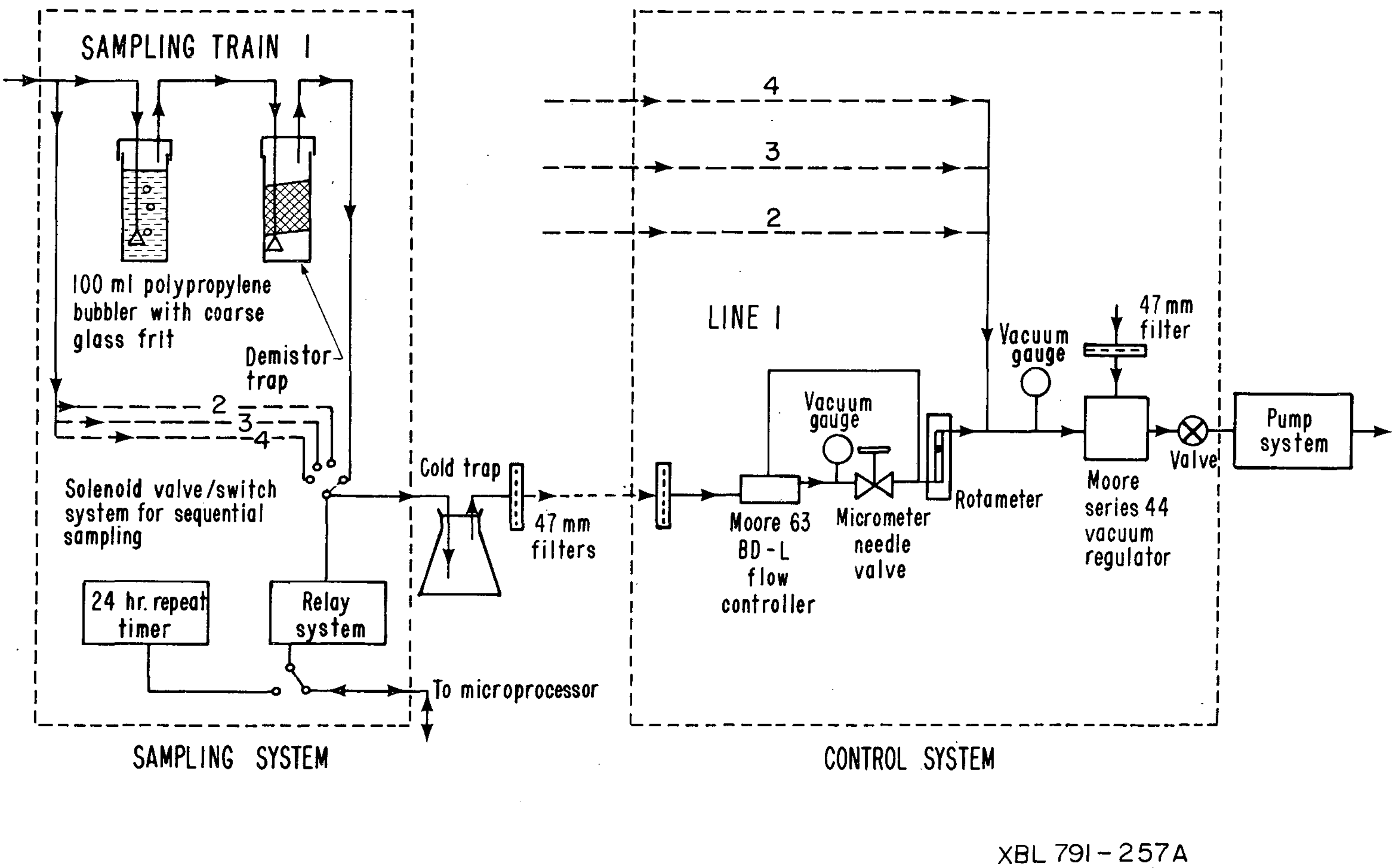

Figure 1. Sampling and control system for sequential sampling of formaldehyde and total aliphatic aldehydes. The unit illustrated contains four automatic sequential sampling systems.

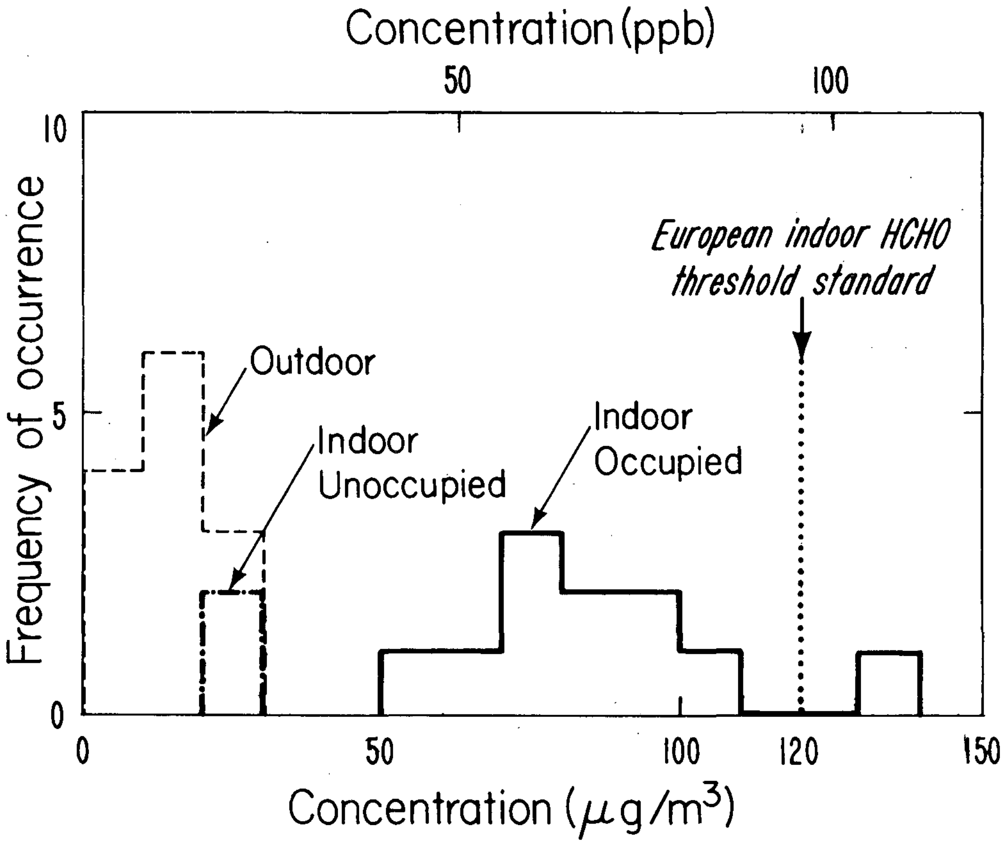

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Figure 2. Histogram of indoor and outdoor total aliphatic aldehyde concentrations measured at the occupied and unoccupied Minimum Energy Dwelling-I houses, Mission Viejo, California, August and September, 1978. Both houses have an air exchange rate of about 0.2 ach.

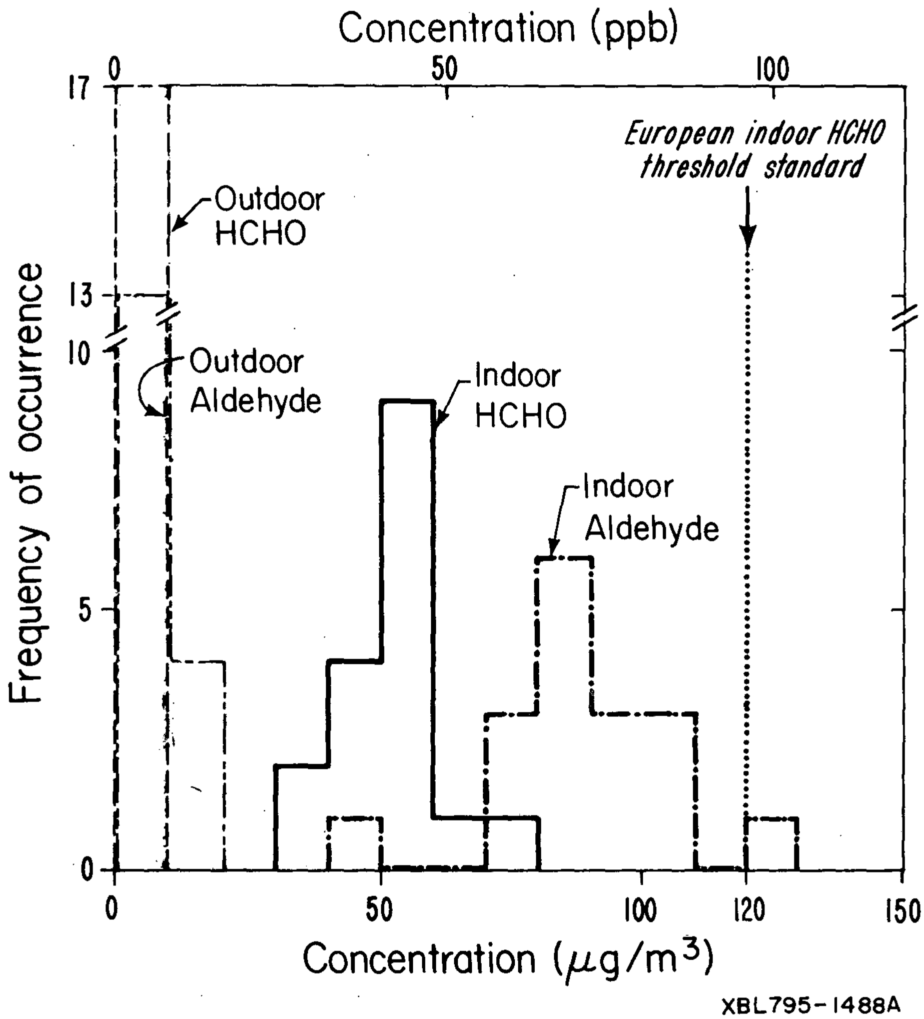

Figure 3. Histogram of indoor and outdoor formaldehyde and total aliphatic aldehyde concentrations in the Iowa State University Energy Research House, December, 1978. The house has an air exchange rate of about 0.2 ach.

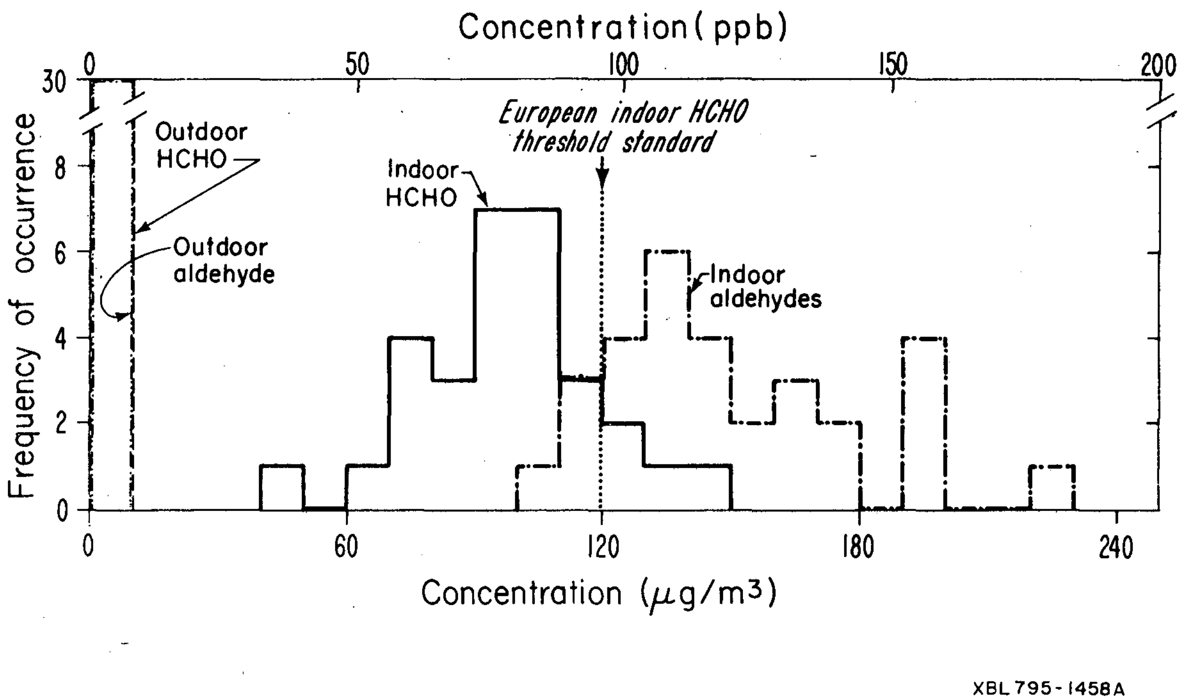

Figure 4. Histogram of indoor and outdoor formaldehyde and total aliphatic aldehyde concentrations measured at an energy research house in Maryland, March and April, 1979. The air exchange rate of the house is about 0.2 ach.

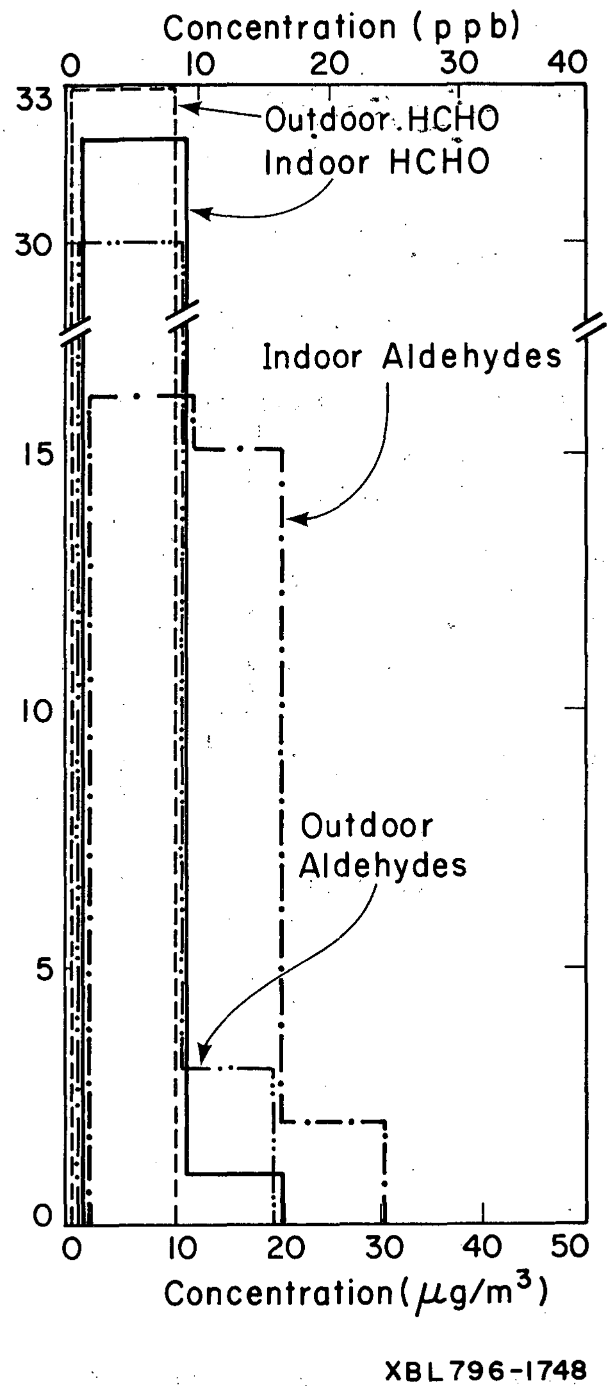

Figure 5. Histogram of indoor and outdoor formaldehyde and total aliphatic aldehyde concentrations measured in a 20-year old school classroom in Columbus, Ohio, January and February, 1979. The air exchange rates of the classroom were varied from about 2.5 down to 0.3 ach during these measurements. The concentration of HCHO did not vary significantly with air exchange rate.

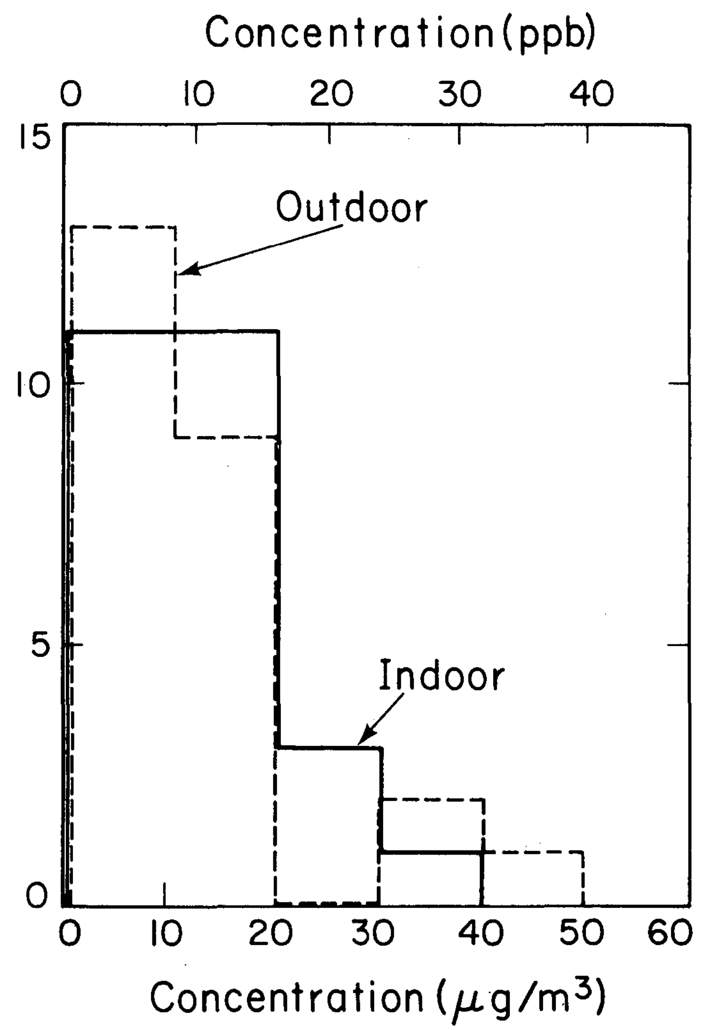

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Figure 6. Histogram of indoor and outdoor total aliphatic aldehyde concentrations measured at Long Beach Naval Regional Medical Center, California, September and October, 1978. The hospital maintains an air exchange rate well above 1.0 ach.

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