Effect of ventilation rates on indoor formaldehyde concentrations in residences

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SUMMARY
Chronic residential exposure to formaldehyde constitutes a human health risk. In this paper we examine the relationship between ventilation rates and indoor concentrations of formaldehyde (C_{in}) in existing homes. We analyzed data from the Relationship of Indoor, Outdoor, and Personal Air (RIOPA) study, where 303 non-smoking residences were monitored. The factors we considered in our analysis include house air exchange rate (AER), volume, indoor temperature, building type and age. In this sample of homes, C_{in} showed limited dependence on any of the studied factors. It is possible that the effectiveness of AER in reducing C_{in} is lessened by a positive relationship between formaldehyde emission rates and ventilation rates. Furthermore, this effectiveness appears to decline with time as the equilibrium concentration in emitting materials decreases. Consequently, strategies to lower population chronic exposures to formaldehyde will likely require lowering the formaldehyde content of building materials.

KEYWORDS
Formaldehyde, ventilation rates, emission factor

INTRODUCTION
Indoor residential formaldehyde exposure is widespread and can constitute a significant human health risk. Indoor concentrations of formaldehyde are generally higher than outdoor values because of the prevalence of sources within homes, including pressed-wood materials that are commonly used in home construction, cabinetry and furniture. Many of these sources emit formaldehyde throughout their lifetimes, which could lead to cancer risks that are two orders of magnitude greater than the US Environmental Protection Agency (EPA) benchmark of 10^{-6} (Hun et al., 2008). Increasing air exchange rates (AER) can mitigate formaldehyde concentrations, but this strategy is mostly based on short-term measurements from chamber experiments (i.e., Myers, 1984). Therefore, the objective of this investigation is to assess the effect of higher AERs on formaldehyde concentrations in existing homes. To this end, we analyzed data from 303 non-smoking households that participated in the Relationship of Indoor, Outdoor, and Personal Air (RIOPA) study.

METHODS
This research is based on the analysis of data from the RIOPA study, which were made available by the Health Effects Institute (HEI) at the website http://riopa.aer.com. Weisel et al. (2005) provide a detailed description of the RIOPA field and measurement protocols. Briefly, from 1999 to 2001, RIOPA recruited 303 non-smoking residences in Los Angeles County, California, Elizabeth, New Jersey, and Houston, Texas. Participants and their homes were monitored during two 48-hour periods that were approximately three months apart. In each of these sessions, personal, indoor and outdoor air were sampled. Formaldehyde

concentrations were collected using passive samplers. Building characteristics and household and personal activity patterns were also collected with questionnaires and walkthrough surveys. AER was measured using the perfluorocarbon tracer (PFT) method.

Several conventions were followed throughout this research. Data from households where someone smoked during a sampling session were excluded. Measurements from homes that were monitored twice were used as independent measurements. AERs greater than 5 hr^{-1} were excluded because the PFT method is unreliable with such values.

Nonparametric statistical analyses were utilized because the data were not normally distributed. The Kruskal-Wallis test was used to evaluate differences among independent samples, such as indoor concentrations from homes in three age categories. The Wilcoxon sign-rank test was used to assess the difference between paired samples, such as indoor and outdoor concentrations that were monitored concurrently. Differences were considered statistically significant at $p \leq 0.05$.

RESULTS

A summary of the measurements is presented in Table 1. Indoor concentrations of formaldehyde (C_{in}) were similar throughout the study, with mean and median values of 21 and 20 µg/m^3, respectively. Furthermore, C_{in} measurements were statistically higher than outdoor values, reflecting the importance of indoor sources for this pollutant. Linear correlations were not observed between C_{in} and AER, house volume or indoor temperature ($R^2 < 0.01$; β_1 not statistically different from 0).

A deeper assessment on possible associations between C_{in} and AER was performed because there are building characteristics that could influence their relationship. To this end, the AERs were divided into three categories (< 0.5, 0.5 – 1.0, > 1.0 hr^{-1}). Indoor concentrations for these AER ranges were not statistically different despite these variations in ventilation rates. Similar results were obtained when considering the effect of building type and age. Figure 1a shows that median values of C_{in} for single-family detached homes, apartments and manufactured homes varied slightly within and between AER categories. Moreover, there were no statistical differences in C_{in} among the three building types within each of the AER categories. Figure 1b indicates that median C_{in} values also do not appear to be affected by building age (< 5, 5 – 15, > 15 years). Differences in C_{in} among the building age categories within each of the AER ranges were not statistically significant.

Table 1. Subset of the RIOPA measurements.

Measurement	N	Mean	SD	Median	% > MDL
Indoor concentration (µg/m^3)	331	21.3	6.75	20.4	99
Outdoor concentration (µg/m^3)	331	6.69	3.65	6.79	99
Air exchange rate (hr^{-1})	331	1.04	0.90	0.72	-
Volume (m^3)	326	234	128	206	-
Indoor temperature (°C)	325	23.8	2.78	23.9	-

Abbreviations: SD, standard deviation; MDL, Method Detection Limit.

DISCUSSION

Indoor concentrations of formaldehyde showed no correlation with ventilation rates for the studied sample of existing residences. This lack of correlation remained even after we examined variables that could have influenced the relationship between AER and C_{in}, i.e., building volume, indoor temperature, residential type and age. To explain these observations, we used a steady-state equation for indoor concentration assuming a well-mixed house (1) and an emission rate equation that is based on mass transfer away from a source for which an equilibrium concentration can be defined at the source-air interface (2). We combined these equations to determine the derivative of C_{in} with respect to AER (3), the derivative of the emission factor with respect to AER (4), and the mass transfer coefficient (5).

$$C_{in} = C_{out} + EF \times (L/\lambda) \tag{1}$$

$$EF = k_g (C_{eq} - C_{in}) \tag{2}$$

$$\frac{dC_{in}}{d\lambda} = \frac{-k_g L(C_{eq} - C_{out})}{(\lambda + k_g L)^2} \tag{3}$$

$$\frac{dEF}{d\lambda} = \frac{k_g^2 L(C_{eq} - C_{out})}{(\lambda + k_g L)^2} \tag{4}$$

$$k_g = \frac{-1}{L}\left\{\left[(C_{in} - C_{out}) \big/ (dC_{in}/d\lambda)\right] + \lambda\right\} \tag{5}$$

Here, C_{out} is the outdoor concentration (µg/m^3), EF is the emission factor (µg/(m^2×h)), L is the loading factor or the ratio of the emitting surface area and space volume (m^{-1}) and was assumed to be 0.41 m^{-1} by using the house floor area as a substitute for the emitting area, λ is the air exchange rate (h^{-1}), k_g is the mass transfer coefficient (m/h), and C_{eq} is the equilibrium concentration at the surface of the emitting material (µg/m^3).

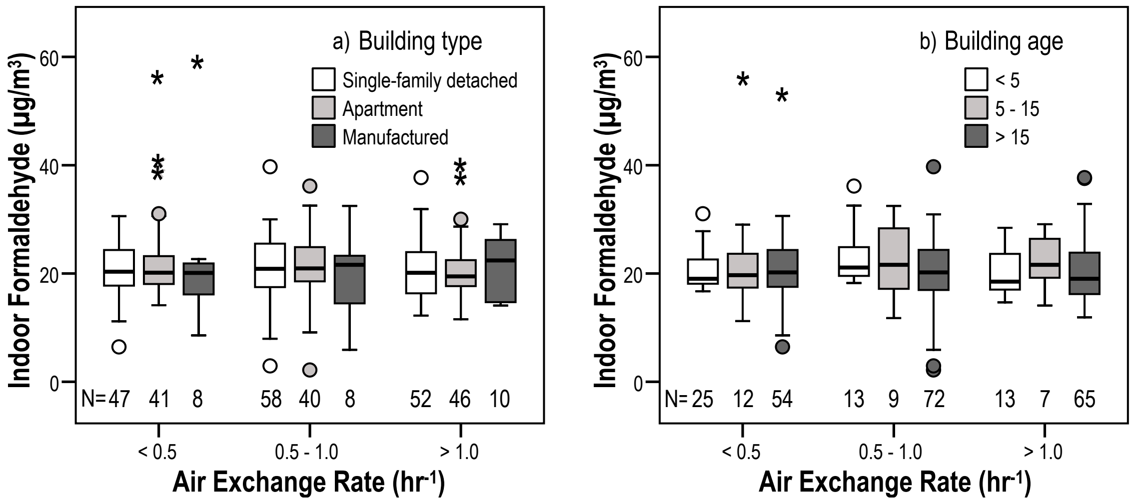

Figure 1. Indoor concentrations of formaldehyde for various air exchange rates and a) building type and b) building age. Box plots summarize the median, lower quartile, upper quartile, lower range and upper range. 'o' and '∗' are values between 1.5 and 3, and more than 3 box lengths from the upper or lower edge of the box, respectively.

Equation 3 shows that C_{in} should decrease with increases in AER, but this may diminish with time as C_{eq} declines. We confirmed this by estimating $dC_{in}/d\lambda$ for new (-54.9 µg×h/m^3, p = 0.105) and existing homes (-0.4 µg×h/m^3, p = 0.334) through linear regressions of data from Sherman and Hodgson (2004) and RIOPA, respectively. Equation 4 indicates that EF and

AER are positively associated, which would lessen the effects of ventilation on C_{in}. Moreover, the rates at which EF and C_{in} are affected by AER differ by a factor equal to k_g. Thus, when k_g is larger than 1 m/h and AERs rise, $dEF/d\lambda$ will be greater than $dC_{in}/d\lambda$. Equation 5 shows that the negative relationship between k_g and $dC_{in}/d\lambda$ may increase with time. This is reflected by our k_g estimates for new (1.1 m/h) and existing (66 m/h) houses, which we calculated using median values for L, ($C_{in} – C_{out}$), and λ. However, the k_g for the RIOPA homes was not within the expected range of 0.2 – 4 m/h (Lehman, 1987). This suggests that our estimate of L = 0.41 m^{-1} is not accurate, or that our assessment should consider other indoor sources of formaldehyde such as chemical reactions involving ozone and unsaturated organic compounds with a terminal double bond.

Our results partially agree with those from other investigations. Myers (1984) reviewed the literature published up to the 1980s on the effect of AER and L on C_{in}. In general, short-term chamber tests indicated that steady-state concentrations were proportional to L/λ. However, Myers noted that this dependence could be affected by decreases in C_{eq} with time. Zinn et al. (1990) confirmed that C_{in}, and consequently C_{eq}, declined with time based on chamber tests where L/λ was held constant. Our results complement this information because our analysis of a sample of existing homes and Equation 3 suggest that decreases in C_{eq} could reduce the mitigation effects of AER on C_{in}.

CONCLUSIONS

Our analysis of the RIOPA database suggests that in a sample of existing homes, higher ventilation rates may not be an indicator of lower indoor concentrations of formaldehyde. The effect of higher AERs is probably lessened by the concurrent positive relationship between formaldehyde emission factors and ventilation rates, and the decrease of the equilibrium concentration of the emitting material with time. Therefore, the most effective strategy to lower population exposure to formaldehyde likely involves regulations that reduce the formaldehyde content in building materials.

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