Impact of simulated climate and building features on the penetration of toxic gases from the ambient into the indoor environment

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This research is a combination of experimental results and analysis of formaldehyde penetration across a residential building envelope with the objective of developing an understanding of the factors that govern indoor air concentrations of air toxics and to provide linkages between EPA's research on indoor and outdoor air toxics and human exposure modeling. Experiments were conducted at an EPA research house. Air flow rates between zones of the research house were determined by injecting tracer gases and measuring the decay rates with time. This was done over a range of imposed pressure differences between the indoors and outdoors, simulating natural ventilation caused by weather effects. Formaldehyde was introduced from the exterior of the house and measured in the same zones as the tracer gas measurements using multiple measurement techniques. The outdoor zone was simulated by an external structure that was dosed with formaldehyde immediately prior to measurements.

KEYWORDS

Building penetration, air toxics, ventilation, hazardous air pollutants

INTRODUCTION

The EPA is moving forward with development of models to represent population-based human exposures that integrate exposures in both indoor and outdoor environments (Isakov et al. 2009). While pollutants in the outdoor environment are relatively well characterized, the same is not true for indoor environments. Pollutants in the indoor environment have major contributions from both indoor and outdoor sources. A better understanding of the relationship of indoor and outdoor contaminants is needed to categorize both individual and population exposures because most individuals spend a large fraction of time indoors. In order for models to properly account for exposures resulting from both indoor and outdoor sources, a better understanding of the penetration of ambient pollutants into the indoor environment is needed.

Development of strategies for reducing indoor exposures to certain toxic contaminants also requires a better understanding of the contributions from similar contaminants in the ambient air. To address this issue, a study of the penetration of selected air toxics from a simulated ambient compartment into the EPA research house (Sparks et al. 1991) has been conducted. The principle of mass conservation is applied in order to measure rates of adsorption and desorption of the contaminants of interest on the interior surfaces of the zone of injection while also accounting for losses due to ventilation. Outer wall assembly penetration was achieved by inducing a negative pressure in the house while zone to zone flow rates were measured using multiple tracer gases.

METHODS

Shown in Figure 1, a building pressurization system was used in addition to tracer gas measurements. An outer "zone" was attached to the back of the building to simulate the desired ambient conditions.



 \ddagger HCHO, SF₆ injection zone

 ϕ CF₃CHFCF₃ injection zone

Figure 1: Layout of the research house. Tracer gases were measured in Zones 1 and 2.

Main building de-pressurization was conducted at 0, 2.5, 5, 7.5 and 10 Pa lower than the simulated outer zone to induce infiltration via common weather pressure differentials. Results from the 2.5 Pa pressure difference case are presented here. For comparison, average pressure differences due to weather effects are of the scale 1-4 Pa and so the values used here can be considered representative of a real world setting. To create this pressure drop in the house, a filtered blower was used which allowed for measurement of the ambient to house pressure drop as well as an estimate of the house air change rate. Flow rates were determined using tracer gas concentrations and a mass balance approach, with derived equations for each predefined zone. The house (excluding garage) was defined as Zone 1, the simulated ambient environment Zone 2, and the ambient Zone "a". The following equations represent these defined zones. The flow rates entering and leaving Zone 1 and Zone 2 were solved through an iterative technique using Equations 1 and 2 in the MicroMath Scientist software including a least squares curve fit with experimental data.

For Zone 1:
(1)
For Zone 2:

$$\frac{d[Z]_{1}}{dt} + \frac{Q_{1a} * [Z]_{1}}{V_{1}} = \frac{Q_{21} * [Z]_{2}}{V_{1}}$$

$$\frac{d[Z]_{2}}{dt} + \frac{Q_{21} * [Z]_{2}}{V_{2}} = 0$$
(2)

Where

 $[Z]_i$ is defined as a combination of measured tracer gas concentrations for Zone i e.g. in Zone 2 where [] denotes concentration: $[Z]_2 = [SF_6]_2 + [CF_3CHFCF_3]_2$ V_i is the volume of Zone i Q_{ij} is the to-be-determined flow rate from Zone i to Zone j

Where the "Z" term is used to minimize the number of equations solved and has no direct effect on the computed results. Although the quantity of data did not allow for a statistical analysis of the results based on outdoor conditions or building to building differences, the data available does provide an initial view of HCHO penetration into the indoor environment for a specific case.

In each tracer gas experiment, the sulphur hexafluoride (SF₆) and heptafluoropropane (CF₃CHFCF₃) tracer gases were injected into different zones to allow for measurement of the flow rates between zones. In addition to air flow measurements, formaldehyde (HCHO) was injected into Zone 2 and its concentrations were measured in Zones 2 and 1. Vapor phase HCHO was dosed by way of a heated gas collection bulb and paraformaldehyde-water solution. The bulb and bulb outlet line were wrapped with heating tape and had a syringe sampling port at the side. The temperature of the heating tape was approximately 140 °C. Total HCHO mass injected was 0.46 ml, resulting in an initial nominal concentration of 2 ppm in Zone 2.

The outdoor to house pressure difference was set to a nominal 2.5 Pa, and a measure of the adsorption and desorption characteristics of the Zone 2 surfaces (the zone of injection) was acquired. Mass loss of HCHO due to its relatively short half life was not considered. The following mass balance equations were used (Tichenor et al. 1991, Singer et al. 2004) to determine the adsorption and desorption coefficients. HCHO concentration measurements included high performance liquid chromatography (HPLC) using 2,4-Dinitrophenylhydrazine (DNPH) cartridges in Zone 1 and an Innova optical sensor in Zone 2. Equations 3, 4 and 5 are HCHO mass balance equations with the Langmuir adsorption processes assumed. These equations are solved with the MicroMath Scientist software using the flow rates that were determined using equations 1-2. In Zone 2, the mass balance equations are presented in Equations 3 and 4, where Equation 4 is written in a general form as it can be applied to either Zone 1 or Zone 2 (with the zone number represented by i).

$$V_2 \frac{dC_2}{dt} = -Q_{21}C_2 - k_a S_a C_2 + k_d S_d M_2$$
(3)

$$\frac{dM_i}{dt} = k_a C_i - k_d M_i \tag{4}$$

These equations are solved in each zone, where

V is zone volume [m³]

C is zone formaldehyde concentration $[mg/m^3]$

Q is flow rate $[m^3/hr.]$ between zones, determined by solving equations 1 and 2.

- k_a is the adsorption rate coefficient [m/hr]
- k_d is the desorption rate coefficient $\left[1/hr\right]$
- M is the sorbed-phase concentration $[mg/m^2]$

S values are the surface areas $[m^2]$ available for adsorption and desorption.

The equation representing Zone 1 is given by Equation 5, which is similar to Equation 3 but includes two additional terms. The first, appearing as the second to last term in Equation 5, is an effective emission rate [mg/hr] from indoor flooring materials based on stainless steel

chamber measurements of similar loading. This assumption of emission only from flooring is based on chamber measurements of this flooring material showing the dominant effect of desorption over adsorption and has been shown to be the case for indoor wooden material of similar age (Park et al. 2006). The Equation 5 terms involving adsorption and desorption relate to HCHO interaction with the drywall surfaces only. The final term appearing in Equation 5 is associated with the mass entering from the Zone 2, which is equal to $P*Q_{21}*C_2$ where Q_{21} is the flow rate from Zone 2 to 1, C_2 is the concentration in Zone 2 and P is the penetration factor. Equation 4 remains unchanged in this zone.

$$V_1 \frac{dC_1}{dt} = -Q_{1a}C_1 - k_a S_a C_1 + k_d S_d M_1 + ER_{flooring} + PQ_{21}C_2$$
(5)

Using the same outdoor to house pressure difference of nominal 2.5 Pa, Equations 4-5 were then solved at this pressure difference, providing a measure of HCHO penetration into the building.

RESULTS

The house to ambient pressure differences imposed by the filtration blower resulted in a building exhaust flow rate and, consequently, air intake from Zone 2. Although this intake was inevitably from multiple origins (attic, crawlspace, Zone 2, etc), HCHO mass was introduced solely through Zone 2 and so incoming HCHO mass from other zones does not appear in the equations. If the main rooms within the house are taken as a single well mixed zone, a correlation can be made between the building indoor to ambient pressure difference and the air flow based on tracer gas measurements for Zone 2 to house flow rate, presented in Figure 2. The adsorption rate coefficient for Zone 2 for the 2.5 Pa pressure difference case was found to be 0.14 ± 0.07 [m/hr], and the desorption rate coefficient was 0.12 ± 0.06 [hr⁻¹]. The adsorption and desorption rate coefficients used for the interior building painted drywall surfaces are those reported by Liu et al. (2009) and the HCHO emission rate for the wood flooring was experimentally determined to be 2.0 mg/hr. Using these values in the 2.5 Pa building to outdoor pressure difference case resulted in a building penetration factor of 0.87for HCHO at this pressure difference over the initial hour of the experiment, when the measurement uncertainty was at a minimum. This penetration factor, appearing in Equation 5, relates the amount of contaminant that is able to cross the building envelope.



Figure 2: Flow rate from outdoor simulated Zone 2 to Zone 1 (left figure), HCHO concentration measurements for the 2.5 Pa case and least square curve fits (right figure) where the HPLC bars represent the time integrated measurement period.

DISCUSSION

An initial review of the data results in a penetration factor for formaldehyde for the 2.5 Pa outdoor to indoor pressure difference. Additional analysis of the existing and additional data is needed to quantify the adsorption and desorption from the indoor building materials as the real world use of materials and weather conditions may result in numbers which differ from those determined in chamber tests. Liu et al. (2009) present values for the desorption and adsorption coefficients of drywall surfaces, which account for some of the interior material used here. Use of mass balance equations has shown to be an efficient method of estimating flow rates, initial mass transfer, etc. This paper is considered a preliminary analysis of results from an extensive research program involving many aspects of penetration to the indoor environment from the ambient for a particular residential building in addition to indoor surface contaminant sink effects. Future work will include a continuation of this analysis of the experimental results.

CONCLUSIONS

The research presented is a first look at contaminant penetration into the indoor environment from ambient. While a penetration factor has been presented for a specific case of outdoor to indoor formaldehyde transport, analysis of additional data is needed to quantify the loss and re-emission from building materials in addition to a broadening of outdoor to indoor pressure differences. The included results are not to be generalized across building types, regions, etc. and so care must be taken in interpreting and using these results. This and future research is expected to aid in the continued development of models that require input on both indoor and outdoor contaminants in estimating risk and the potential for exposure towards developing options for risk management.

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