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Abstract: Measurements were taken in new US residences to assess the extent to which ventilation and source control can mitigate formaldehyde exposure. Increasing ventilation consistently lowered indoor formaldehyde concentrations. However, at a reference air exchange rate of 0.35 h^{-1}, increasing ventilation was up to 60% less effective than would be predicted if the emission rate were constant. This is consistent with formaldehyde emission rates decreasing as air concentrations increase, as observed in chamber studies. In contrast, measurements suggest acetaldehyde emission was independent of ventilation rate. To evaluate the effectiveness of source control, formaldehyde concentrations were measured in Leadership in Energy and Environmental Design (LEED) certified/Indoor airPLUS homes constructed with materials certified to have low emission rates of volatile organic compounds (VOC). At a reference air exchange rate of 0.35 h^{-1}, and adjusting for home age, temperature and relative humidity, formaldehyde concentrations in homes built with low-VOC materials were 42% lower on average than in reference new homes with conventional building materials. Without adjustment, concentrations were 27% lower in the low-VOC homes. The mean and standard deviation of formaldehyde concentration were 33 $\mu g\ m^{-3}$ and 22 $\mu g\ m^{-3}$ for low-VOC homes and 45 $\mu g\ m^{-3}$ and 30 $\mu g\ m^{-3}$ for conventional.

Keywords: Formaldehyde; Acetaldehyde; Indoor air quality; LEED; Indoor airPLUS; VOC

Practical Implications: Results suggest that both increased ventilation and home certification programs that require low-emitting building materials can significantly reduce residential formaldehyde exposures. Concentration-dependent formaldehyde emission rates can make ventilation 20-60% less effective at

reducing short-term concentrations than ventilating a pollutant emitted at a constant rate. But the increase in emissions can lead to quicker depletion and similar long-term reduction benefits. Achieving California

Office of Environmental Health Hazard Assessment chronic and 8-h reference exposure levels of 9 µg m^{-3} (OEHHA 2013) may require combining source control with ventilation. Ventilation rates could be increased during the first year after construction when VOC concentrations tend to be highest.

Introduction

Formaldehyde has among the largest total health impacts of chemical air pollutants in US residences, as quantified by disability adjusted life years (Logue et al. 2012). Recent studies report associations between in-home formaldehyde concentrations and childhood asthma (McGwin et al. 2010; Dannemiller et al. 2013), and formaldehyde is classified as a human carcinogen (NTP 2011). Formaldehyde concentrations in US residences regularly exceed benchmarks established for health protection (Logue et al. 2011). In a study of California new homes (Offermann 2009), formaldehyde concentrations measured over 24 h exceeded the California OEHHA chronic and 8-hr reference exposure levels of 9 µg m^{-3} (OEHHA 2013) in 98% of homes and exceeded the World Health Organization 30-minute exposure guideline of 100 µg m^{-3} (WHO 2010) in 5% of the homes. Acetaldehyde is classified as 'reasonably anticipated to be a human carcinogen' (NTP 2011). Offermann (2009) reported that 82% of new California homes had acetaldehyde concentrations exceeding the US EPA reference concentration for chronic inhalation exposure of 9 µg m^{-3} (US EPA 1999). Studies suggest the range of formaldehyde concentrations in European residences is similar: although measurements were taken under a range of conditions and sampling strategies, results summarized by Sarigiannis et al. (2011) indicate concentrations ranged from 6 to 171 µg m^{-3} with mean values in a given study between 12 and 41 µg m^{-3}. Studies of indoor acetaldehyde are limited (e.g., Lovreglio et al., 2009), and concentrations are typically much lower than formaldehyde.

The largest source of formaldehyde in homes is thought to be building materials such as composite wood, coatings, fiberglass insulation and paper products (Baumann et al. 2000, Salthammer et al. 2010). Additional sources of formaldehyde include permanent press fabrics and personal care products (Kelly et al. 1999), natural gas combustion, tobacco smoke, and chemical reactions between ozone and terpenes (Wolkoff et al. 2000). Acetaldehyde can be emitted by building materials such as wood, cork and linoleum as well as in episodic events such as cooking, combustion, human exhalation and the use of household products.

Two main strategies to reduce indoor exposures to these volatile aldehydes are increasing ventilation and reducing emissions from indoor sources, i.e. source control. These strategies are theoretically solid but there are limited empirical data quantifying their effectiveness in homes.

Formaldehyde emission from materials varies with seasonal factors (temperature, relative humidity, insolation); the quantities and types of materials; and material age. Variations in these factors can obscure the relationship between ventilation rate and formaldehyde in occupied homes. For example, Hun et al. (2010) analysed cross-sectional data on formaldehyde and air exchange rate and found no dependence of indoor formaldehyde concentration on air exchange rate. In contrast, Offermann (2009) presented data showing indoor formaldehyde concentration decreasing as air exchange rate increased in a cross-sectional study of new California homes. Recent findings suggest supply ventilation may be substantially more effective than exhaust ventilation at lowering residential formaldehyde concentrations (Hun et al. 2013).

If the emission rate from indoor sources is independent of ventilation rate and there are negligible outdoor sources, then doubling the ventilation rate should halve the time-averaged indoor concentration. Laboratory chamber experiments indicate that the VOC emission rate from building materials depends on the air exchange rate as reviewed by Myers (1984) and Salthammer et al. (2010). Increasing ventilation lowers the bulk indoor air concentration through dilution; this causes a steeper concentration gradient in the boundary layer resulting in a faster rate of mass transfer from the surface to the bulk air. Whether the apparent increase in emission rate is durable depends on the availability of the chemical at the material surface and the rate of diffusion within the material.

Mass transport from building materials has been modelled by assuming a thin layer of air near the emitting material is constantly in equilibrium with the contaminant concentration in the surface layer of the storage medium (Dunn 1987; Sparks et al. 1996). Transport between this equilibrium layer and the bulk air volume is governed by the time constant *kL* [h^{-1}], which is the product of the transport coefficient *k* [m/h] and the loading factor *L* [1/m]. *L* is the ratio of storage medium surface area to bulk air volume. The effective storage medium surface area includes any sink materials that could absorb and reemit the contaminant (Matthews et al. 1987, Gunschera et al. 2013). This concentration-dependent emission model has been validated against laboratory emission data for formaldehyde and other VOCs (Myers 1984; Dunn 1987; Sparks et al. 1996; Won et al. 2001), and the implications of this model for indoor formaldehyde exposure are discussed by Sherman and Hult (2013). According to this model, the steady-state indoor concentration in excess of the outdoor concentration, *C*, decreases as the air exchange rate *A* [h^{-1}] is increased:

$$C = C_{eq}\frac{kL}{A + kL} \tag{1}$$

where C_{eq} is the indoor concentration when $A=0$. In contrast, in developing ventilation guidelines, it is often assumed that the emission rate is constant:

$$C = \frac{E}{A} \tag{2}$$

where *E* is the emission rate per unit volume. Here both models assume a well-mixed indoor air volume and no contaminant in the outdoor air. Although not included here, diffusion within the source material can impact VOC emissions (e.g., Cox et al. 2002). In the limit where kL is small relative to A, the concentration-dependent emission model reduces to the constant emission rate model and the assumption that the emission rate is independent of air exchange rate may yield reasonably accurate predictions.

If increasing the ventilation rate leads to a higher emission rate, then in the short term, ventilation will be less effective at reducing indoor concentrations than it is in the constant-emission case. However, elevated emission rates also deplete contaminant sources more quickly, yielding a steeper decline in concentrations over time. Sherman and Hult (2013) show that if occupants are exposed to a source material from its introduction until the source is depleted, the total exposure varies inversely with the air exchange rate, whether emissions occur at a constant rate or at a rate that depends on indoor concentration. Long-term health effects, however, may depend on the concentration profile rather than simply the total, integrated exposure.

Limiting VOC emissions in the indoor environment, known as ‘source control’, is another strategy to decrease indoor exposure. Source control can be pursued by limiting the quantity of VOC-emitting materials in the space, sealing materials to reduce emissions, and changing use patterns of household products. While laboratory emission data exists for a range of specific materials and conditions (Myers 1984; Kelly et al. 1999; Baumann et al. 2000; Kim et al. 2007; Zhang et al. 2007; Willem and Singer 2010), emission rates from installed materials in occupied buildings have been observed to vary significantly from the same materials tested in the laboratory (Kang et al. 2012). At the whole-residence level, few studies have examined the effectiveness of source control, specifically how VOC concentrations in homes built with low-emitting materials differ from those in homes built with conventional materials. Järnström et al. (2006) reported VOC concentrations in 14 new residential units, built with materials classified in Finland as ‘low-emitting’. Levels were comparable to new California homes built with conventional materials (Offermann 2009).

In this study we investigated the extent to which formaldehyde concentrations in inhabited homes depend on air exchange rates and the use of certified low-VOC materials. The key hypotheses were 1) increasing the ventilation rate in homes lowers indoor formaldehyde concentrations, but less than would occur if sources were to emit at a constant rate, and 2) homes built with low-emitting materials have lower formaldehyde concentrations. In-home measurements are essential to validating and quantifying the formaldehyde reduction benefits of ventilation and of building with low-emitting materials. The relative importance of construction and finishing materials vs. other formaldehyde sources is unknown and expected to vary across homes.

This paper presents results for formaldehyde as well as acetaldehyde, which can be emitted both from building materials and through specific processes such as cooking. To examine the relationship between ventilation rate and indoor concentration, measurements were taken at three controlled ventilation rates in nine residential units, while controlling the indoor temperature and attempting to minimize the opening of doors and windows; we refer to this as the ventilation and indoor air quality (VIAQ) study. To assess the impact of source control, measurements were made in 13 homes constructed with certified low-emitting materials and compared to data from homes built with conventional building materials; we refer to this as the low-emitting materials (LOEM) study.

Methods

VIAQ study protocol

The VIAQ study was designed to assess the impact of ventilation on VOC concentrations in furnished homes. A complete description of this study can be found in Willem et al. (2013) and a summary is provided here. The intent was to control other environmental variables to study the effect of air exchange rate on formaldehyde and acetaldehyde concentrations.

Target criteria for study homes were as follows: 0.5 to 5 years old; 80-300 m^2; airtight (preferably ≤5 air changes per hour at 50 Pa); furnished; featuring mechanical ventilation equipment (including kitchen and bath exhaust fans) capable of providing 0.8 air changes per hour (h^{-1}) or potential to install a temporary balanced ventilation system in a window; thermostatically controlled with heating and/or cooling as needed for local climate; at least 400 m from major outdoor sources such as highways and industrial sites; and sealed to be isolated from an attached garage if present.

We selected six detached houses meeting all criteria: H1, H4, and H6–H9, and two identical, single-room guesthouse units (R2 and R3) that met all criteria other than size. Though it did not meet the age criterion, H5 was included as a home with high formaldehyde concentrations based on measurements conducted as part of the California New Home Study (Offermann 2009).

The air exchange rate was determined by measuring concentrations of perfluorocarbon tracer gases (PFTs) released from passive emitters deployed throughout the homes. PFT emission rates were determined from the change in mass of the emitters. Bag samples of room air were collected during the PFT releases. The PFTs perfluorodimethylcyclobutane (PDCB, CAS 2994-71-0) and perfluoromethylcyclohexane (PMCH, CAS 355-02-2) were used, with one released on each level in 2 floor homes and one or both used in single floor homes.

Three air exchange rates were established in each home. Target rates were 0.2, 0.4 and 0.8 h^{-1}. In homes with an existing energy or heat recovery ventilator (ERV or HRV) system, the flow rate was adjusted to target the medium and high ventilation rates, and the system was turned off to target the low rate. A temporary balanced ventilation system was installed in residences without a dedicated mechanical ventilation system. Variations in infiltration-induced air exchange were reduced by studying the three ventilation conditions for each home under similar ambient environmental conditions and with thermostatic control set at 22±1°C. R2 and R3 were studied in December 2010 and H1 and H4-H9 between May and September of 2011. Residents in occupied homes (H6-H9) agreed not to open their windows starting from the night before each sampling event. In one case (H8), though, windows were observed to be open when we arrived for sampling. Furnishings and the number of residents (if any) were the same throughout the measurements in each home. Each ventilation setting was maintained for at least two days, and the indoor air quality sampling was carried out on the final afternoon. Low, medium and high ventilation settings were maintained over periods of 3-8 days in R2, R3 and H5 to allow for potential transient aldehyde transfer to sink materials in the space, and it was established that two days was sufficient to establish a pseudo-steady-state condition.

During site visits, researchers checked windows and exterior doors to note status upon arrival, confirmed that the mechanical ventilation system was operating at the intended setting, then conducted air sampling for VOCs, aldehydes, and PFTs. After sampling, the ventilation system was adjusted to the next setting to be tested. All PFT emitters were deployed at least two days before the sampling events. An emitter was placed every 30-50 m^2 of floor area, or at least one emitter in each zone or area of the house. Area-specific emission rates were 19.4 - 37.3 μg h^{-1} m^{-2} for the single-family homes, and somewhat higher in R2 and R3 due to smaller floor area. Tracer gas concentrations were between 0.1 and 19.8 ppb, with a median value of 1.5 ppb.

LOEM study protocol

The LOEM study sought to assess whether homes built with low-emitting materials have lower concentrations of formaldehyde and other VOCs when compared to homes built with conventional materials. Homes meeting the Indoor airPLUS qualification were identified and recruited with assistance from the U.S. Green Building Council (USGBC). The Indoor airPLUS qualification (US EPA 2009) was introduced by U.S. EPA to assist homeowners looking to improve the indoor air quality of their homes, and is now part of the ENERGY STAR home labelling program and the LEED for Homes certification by USGBC (http://www.usgbc.org/leed/homes). Use of low-emitting materials is a key requirement to obtain the Indoor airPLUS qualification. The study was conducted in 13 homes (G1-G13) within two clusters of qualified homes in New Mexico, in collaboration with the contractor for the developments. As controls, two new homes in the same area built with conventional building materials (C1 and C2) were also recruited, and homes from previous studies served as additional controls. Each home had a central air conditioning unit and an air distribution system connected to an HRV.

The Indoor airPLUS construction specifications (US EPA 2009) refer to material emissions guidelines in the California Department of Public Health specification document, Section 01350 (CDPH 2010). These specifications required that 1) wood products for the home structure, finishing, and cabinetry were certified compliant with Section 01350 requirements or other equivalent low- or no-formaldehyde standards for wood-based materials; 2) surface finishing (wet) products such as interior paints were third-party-certified low-emitting products tested according to Section 01350, or other equivalent low- or no-VOC paints; and 3) carpet materials and backing systems were third-party-certified low-emitting products based on Section 01350 requirements, or other equivalent certification systems.

The LOEM study required only one site visit for air sampling. Homeowners set the ventilation rate and assisted in the installation of PFT emitters, which were mailed to them at least two days prior to the site visit. The package contained PFT emitters, installation materials and instructions, and at least one data-logging temperature and RH sensor. On the day of package delivery, a researcher contacted each homeowner to verify delivery and understanding of instructions. Emitters were placed in each room, with up to two emitters in larger rooms or areas. The air temperature was maintained at the homeowner’s preferred set point. A temperature/RH sensor was installed on each floor of the home and the calculated PFT emission rate was adjusted for any significant temperature variations observed. Homeowners were asked to keep their windows and doors closed starting at least one day before the site visit.

During the home visit, PFT emitter installation was verified and the following factors were determined: temperature set point, HRV ventilation setting, home size, number of floors, and the presence of attached garage. In a single air sampling session at each site, active air sampling methods were used to measure formaldehyde and PFT concentrations. During the aldehyde sampling, at minimum, one indoor sample and one travel blank were collected at each of the two indoor sampling locations. One location at each site was randomly selected for duplicate sampling. Because homes were located in two small clusters, outdoor samples were collected at five sites that were considered representative of the clusters. Air samples for aldehyde quantification were collected over 40 min periods. Home visits took on average 2 hours to complete.

LOEM PFT sampling and analysis

The LOEM study used either PMCH or PDCB at each site: PDCB was used in 9 homes and PMCH in 6 homes. The PFT emitters were 2-dram glass vials with a PVC cap and Teflon-lined septum. Area-specific emission rates were in the range of 15.1–47.3 μg h^{-1} m^{-2}. Air samples were collected in a 7-bag set of 0.3L Tedlar bags, conditioned and checked for contamination prior to the site visits. Six bags were evacuated and filled in each home. The remaining bag was pre-filled with PFT-free air and carried as a travel blank to confirm that no PFT contamination occurred during transit. Bags were filled using a hand syringe pump in G1-G6 and a manually operated pump in G7-G13 and C1-C2.

Each 7-bag set was analysed for PFTs using a dual-column, dual-detector gas chromatograph equipped with electron capture detectors (GC-ECD); details of the system are provided in Section 2.4 of Willem et al. (2013). The GC was calibrated using PFT standards in 12 concentrations, ranging from 0.79 to 15.1 ppb for PDCB and from 0.61 to 11.4 ppb for PMCH. Standards were prepared in clean Tedlar bags filled with diluted gas mixture from a calibrated gas cylinder. A calibration curve was developed for each PFT to determine the PFT concentration. A new calibration took place each time the GC-ECD system was restarted. The quantitation limit was 0.05 ppb, based on past analysis. Samples were extracted from the bags using a peristaltic pump (Cole-Parmer 7553-80 with size 17, Norprene tubing, www.coleparmer.com) and an actuator setup for 16 positions (Valco Instruments EMTMA-CE, www.vici.com) equipped with a 0.25 mL sample loop.

Aldehyde sampling and analysis

Aldehyde sampling followed the procedure described in Section 2.6 of Willem et al. (2013). DNPH-coated cartridges were analysed for target aldehydes following ASTM Method D5197-09e (ASTM 2009); details are provided in Section 2.6 of Willem et al. (2013).

Quality assurance measures for the VIAQ study are described in Section 2.7 of Willem et al. (2013). In the LOEM study, 10% additional samples were collected as duplicates with at least one duplicate for each study visit.

Data analysis

Total PFT emission rates and the average measured PFT concentrations were used to determine the whole-house air exchange rate. Single level VIAQ homes as well as all LOEM homes were each treated as a single well-mixed zone and only one PFT was used. Using a single zone, pseudo-steady state, mass-balance model to determine the air exchange rate:

$$A = \frac{E_{PFT}}{C_{PFT}V} \tag{3}$$

where E_{PFT} is the total PFT emission rate (μg h^{-1}), C_{PFT} is the volume-average PFT concentration in the home (μg m^{-3}), and V is the effective house volume (m^3). For two floor VIAQ homes, two PFTs were emitted (see Willem et al. 2013 for calculation of the air exchange rate).

Aldehyde emission rates were calculated assuming well-mixed zones at pseudo-steady state:

$$E = \frac{VA}{A_f}(C - C_{out}) \tag{4}$$

where E is the emission rate of target compound (µg h^{-1} m^{-2}); C is the average concentration measured at indoor locations (µg m^{-3}); C_{out} is the measured outdoor concentration (µg m^{-3}); and A_f is the effective floor area of the house (m^2).

The mean relative uncertainty in the PFT emission rate is approximately 15%, with 10% bias error and 5% random error assumed, given that emission rates were corrected for temperature (Lunden et al. 2012, Sherman et al. 2014). The percent uncertainty in the PFT concentration is approximately 10% based on the accuracy of the GC-ECD calibration. The calibration error is not assumed to have the same sign for all samples and thus is not treated as a bias error. Uncertainty in the air exchange rate is estimated to be 27%, including 20% uncertainty in the residence volume. The AER uncertainty is 18% if uncertainty in the volume is neglected, which it may be when comparing varying ventilation rate conditions in each VIAQ home. The mean uncertainty in aldehyde concentration measurements is 4% based on duplicate samples.

Bayesian modelling of ventilation control

If there were no uncertainty in the measurements, and if the model closely matched real-world behaviour, then the parameter values kL and C_{eq} that yield the best fit to the data would be close to the true values of the parameters. But with rather large measurement uncertainties in air exchange rate and to a lesser extent concentration, the parameter values that best fit the data can differ substantially from the true values. To obtain the best possible estimates, a hierarchical Bayesian model (see the SAT example in Gelman et al., 2004 for an introduction to this concept) was used to fit the concentration-dependent emission model to the VIAQ measurements. The analysis was run using Stan (Stan development team 2013). The Bayesian model, defined below and in the Supporting Information, partially shares or “pools” information from various houses. To see why this is reasonable, consider the following example. Suppose we knew the true value of C_{eq} in 8 randomly selected houses, and that in all of these cases the value was between 40 and 150 μg m^{-3}; it would be surprising if the value in a 9th randomly selected house turned out to be 500,000 μg m^{-3}, or 0.0001 μg m^{-3}. By assuming a common statistical distribution for the underlying parameter values, measurement uncertainty can be taken into account when predicting the underlying true value from the measured value: the estimated parameter value for a specific house is a compromise between the value that gives the best fit to the data from that house, and the value that is closest to the mean of the other houses.

The model assumes that 1) given the true values of C_{eq}, kL, and A, Equation (1) would predict the value of C with normal error proportional to C, within an unknown proportionality constant that is one of the parameters to be estimated; 2) the measured value of C is drawn from a normal distribution with mean equal to the true value of C and standard deviation proportional to C, with an unknown proportionality constant that is to be estimated; 3) the measured value of A is drawn from a normal distribution with a mean equal to the true value of A and a standard deviation proportional to A, with the proportionality constant to be estimated; 4) the true values of C_{eq} are drawn from a lognormal distribution with unknown geometric mean and geometric standard deviation; 5) the true values of kL are drawn from a lognormal distribution with unknown geometric mean and geometric standard deviation. Weakly informative prior distributions were provided for all of the distributional parameters, as described in the Supporting Information.

The output of the Bayesian estimator is a large set of simulations, each representing a possible set of true parameter values, including possible true values of A and C. These values are consistent with the measured values as well as the assumptions above. Some areas of parameter space are more likely than others, and those areas are more heavily sampled. In addition to the model described above, we experimented with different distribution types (such as Cauchy and t-distributions, rather than normal distributions) to assess the sensitivity to these choices. The selection of the prior distribution impacted the results slightly at the tail ends of the distributions of C_{eq} and kL, but did not substantially affect the central portions of the distributions. The model did not include formaldehyde removal or production by chemical reaction as these processes were assessed as not likely to have impacted indoor concentrations under the conditions of the study, and this assessment is discussed in detail in Section 1 of the Supporting Information.

Analysis of source control datasets

Table 1 Summary of differences between source control datasets.

Factor	Low-emitting materials		Conventional construction			Impact on concentration
	LOEM* N=11	VIAQ^ N=4; H1,4,6,7	CNHS N=54	LOEM N=2	VIAQ N=3; H5,8,9	
Median temp [°C] (std dev)	23 (1.4)	22.2$^{\#}$	24.4 (1.5)	22.2, 22.2	22.2$^{\#}$	Approx. 11% increase per °C (Eq. 3) (Myers 1985)
Median RH [%] (std dev)	46 (8.5)	NA	41.5 (10.4)	60, 64	NA	Approx. 2% increase per percentage point (Eq. 3) (Myers 1985)
Median house age at sampling [yr] (std dev)	0.8 (0.3)	0.9 (0.7)	3.3 (0.7)	3, 6	2.5, 2.5, 7.5	Decrease in first 2 years; (Eq. 4) (Park & Ikeda 2006)
Sampling date	Jul-Sep 2011	May-Aug 2011	Aug-Sep 2006	Sep 2011	Jul-Sep 2011	Emissions higher in summer months
Year built	2010-2011	2009-2011	2001-2004	2008, 2005	2009, 2009, 2004	New standards may lower baseline

* LOEM homes 7 & 8 were omitted because air exchange rate measurements were not available. ^VIAQ residences 2 and 3 were omitted because measurements were taken in December. $^{\#}$VIAQ temperatures were verified to be within 1.1°C of the 22.2°C set point.

To analyse the impact of source control, we sought data on additional control homes built with conventional building materials beyond the two sampled in the LOEM study. The best existing comparison dataset was the California New Homes Study (Offermann 2009), which measured indoor formaldehyde concentrations during the summer season in 58 homes. The CNHS sampled single-family, detached homes less than 5.5 years old at sampling, with most only 3-4 years old. Although CNHS homes may have contained some low-emitting building materials, the study was designed to select a cross-section of new California homes, and is considered to represent homes built with conventional building materials.

Measured concentrations in the LOEM study were combined with data from the CNHS and the VIAQ homes with low-emitting and conventional materials for analysis. Both the CNHS and the measurements in this paper were limited to homes built within 5.5 years of sampling, and are in similar climates. Data from the VIAQ study homes that requested or had certified low-emitting materials were included in the 'low-emitting' category. There are some important differences between the datasets, however. The median home age in the CNHS was 3.3 years, whereas the LOEM study homes were much newer, with a median age of 0.8 years (Table 1). The temperature in the VIAQ homes was controlled to a set point of approximately 22.2±1.1°C (72±2°F), whereas LOEM and CNHS homes were operated at thermostat set points selected by homeowners. Both temperature and relative humidity can increase emission rates of formaldehyde from building materials, so differences in HVAC control and climate zone can impact concentrations.

To compare the datasets, we report both the measured data and data adjusted to a temperature of 23.2°C and relative humidity of 43%. The standard temperature T_S [K] and relative humidity RH_S [%] are the median values for the combined data. The adjusted concentration C_S was calculated from the measured concentration C at conditions RH [%] and T [K]:

$$C_S = C[1 + \alpha(RH_S - RH)]exp\left[\beta\left(\frac{1}{T_S} - \frac{1}{T}\right)\right] \quad . \tag{5}$$

Constants $\alpha = 0.0195\%^{-1}$ and $\beta = -8930K$, were assumed based on the compilation of results from experimental testing of building materials containing formaldehyde (Myers 1985). VIAQ homes were assumed to be at the specified set point temperature and were not adjusted for RH because measurements were not available.

In combination with the temperature and relative humidity adjustment, indoor formaldehyde concentrations were adjusted for house age based on the results of Park and Ikeda (2006) who measured concentrations in homes initially less than 6 months old, and in homes older than 6 months. The same homes were sampled during three subsequent summers. They found no decrease after 2 years in older homes, but a decrease of 36% in the new homes. Brown (2002) found a similar dependence in Australian homes. Interpolating from the results of Park and Ikeda, concentrations in homes less than 2.25 years were adjusted to an age of 2.25 years, where *C* is the measured concentration at age y in years:

$$C_A = C[1 + 0.279(2.25 - y)] \qquad (6)$$

Concentrations in homes older than 2.25 years were not adjusted for age.

Results

Ventilation control

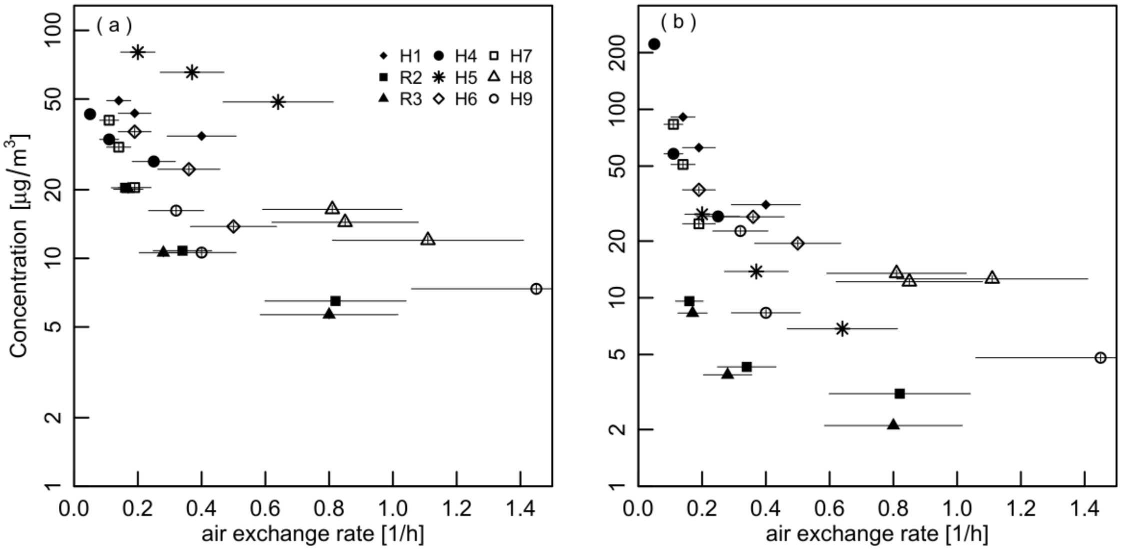

Figure 1 Concentration of (a) formaldehyde and (b) acetaldehyde for three air exchange rates at each site. Error bars indicate uncertainty in measured quantities, noting 4% uncertainty in C is small relative to the symbol size.

The range of aldehyde concentrations measured across the VIAQ residences was quite large, even at comparable ventilation rates, as shown in Figure 1. These data suggest overall emission rates varying by roughly a factor of 5 across the sample of homes of similar age. Figure 1 shows that in all residences, higher air exchange rates yielded lower indoor concentrations of formaldehyde and acetaldehyde, as expected.

To assess whether the emission rate of formaldehyde in the indoor environment is independent of the ventilation rate, we compared observations with the concentration-dependent emission (CDE) model of Equation (1) and with the constant emission model of Equation (2), as shown in Figure 2. The two models were fit to concentration measurements at three air exchange rates in each of the 9 residences of the VIAQ study. Previously, these observations were used to validate a portion of the CDE model presented in Sherman and Hult (2013).

Overall, increasing the air exchange rate leads to a smaller decrease in indoor concentration than the constant emission rate model predicts. When kL is large relative to A, the emission rate depends strongly on the bulk air concentration, and the emission process is not well approximated by the constant emission rate model. Of greatest interest are results from homes H1-H6, which had air exchange rates bracketing the common benchmark value of 0.35 h^{-1}. For H7, only low air exchange rates were achieved. For H8 and H9, low air exchange rates were not achieved and the parameters kL and C_{eq} could not be as well determined, as shown by the spread in the Bayesian fitted curves.

From the Bayesian model results for formaldehyde emission, median values of C_{eq} were between 45 and 118 μg m^{-3} and kL were between 0.10 and 0.46 h^{-1}. Even though the mean and standard deviation of the distribution of C_{eq} were not specified, just specifying in the Bayesian model that this variable has a lognormal distribution tends to cluster the fitted values of C_{eq} (listed median values), compared with the least squares fitted curves (solid black curves).

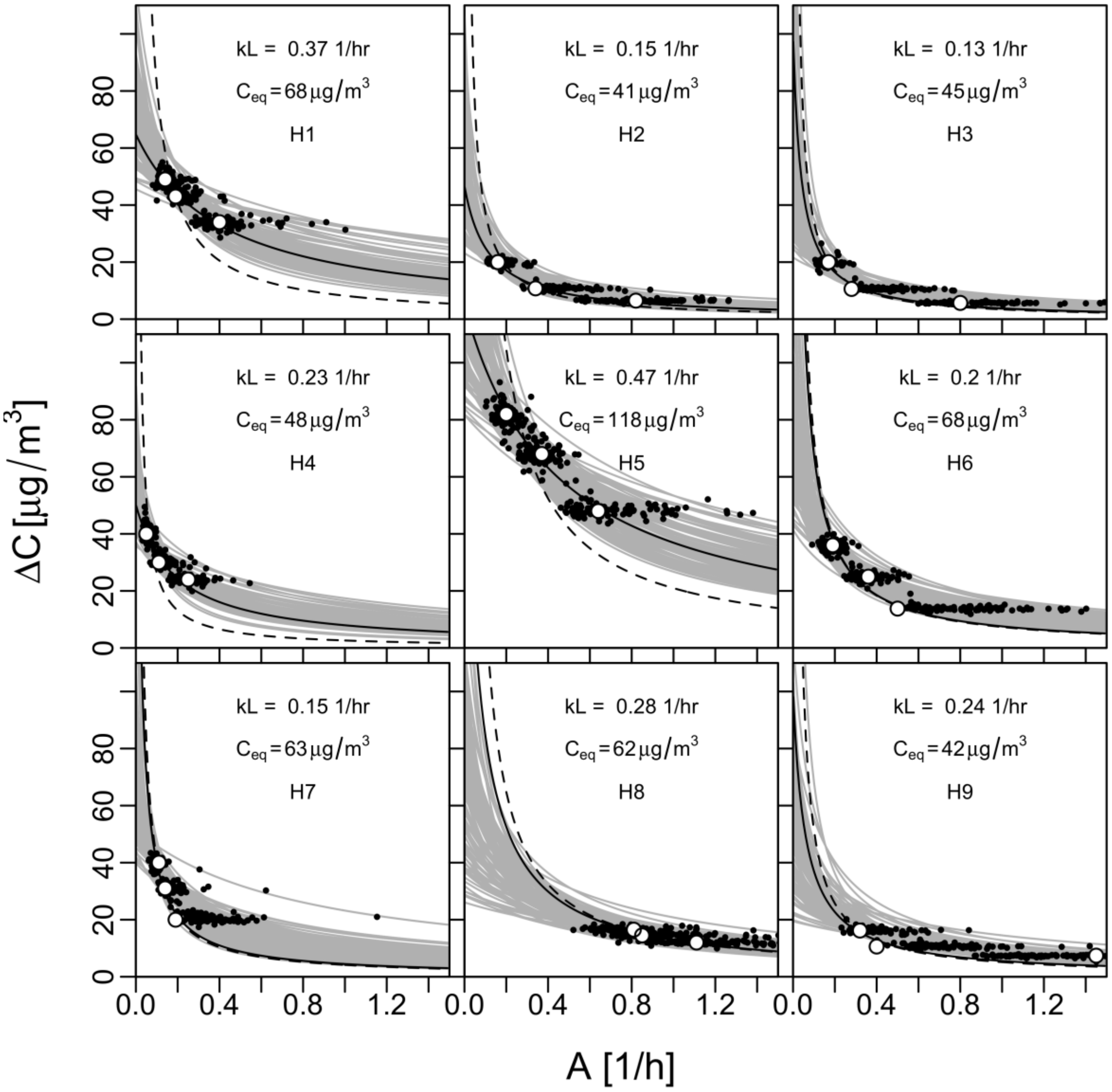

Figure 2 Indoor formaldehyde concentration (above outdoor level) as a function of the air exchange rate *A* by residence, including with model curves and Bayesian estimator results. Solid lines show the best-fit concentration-dependent emission model of Equation (1). Dashed lines show the best-fit model with a constant emission rate, as in Equation (2). In general, the concentration-dependent model matches the measured data (white circles) more consistently. Solid points show 100 samples from the posterior distribution of the Bayesian model and the light grey curves show Equation (1) fit to these values. Parameter values listed are the median values from the Bayesian model.

To determine the effect of measurement uncertainty, the following procedure was employed: 1) for each residence a possible ‘true’ value was sampled from the posterior distribution associated with each measurement; i.e., each solid black point in Figure 2 represents a possible ‘true’ value of the associated white circle, if there were no measurement error. 2) A curve, displayed in grey, is fit to the resulting 3 ‘true’ values, under Equation (1). 3) Steps 1 and 2 are repeated for 32,000 samples, of which 100 are shown in Figure (1). The spread in the grey curves illustrates the impact of measurement uncertainty. Analogous to conventional error bars, the extent to which the simulated ‘true’ values of A and C (solid points) differ from the measured values (open symbols) provides an indication of the uncertainty in the measured values. The uncertainty in air exchange measurements is relatively large (estimated as 18% based on error propagation), thus some simulated ‘true’ values of A deviate noticeably from the measured values of A.

When the CDE model was fit to acetaldehyde, values of kL were low (0.0003 to 0.0009 h^{-1}) relative to A, as shown in Figure SI.1 of the Supporting Information. Thus, for acetaldehyde, the CDE model is not a substantial improvement over the constant emission rate assumption in modelling the dependence of indoor concentration on air exchange rate (see Figure SI.2 of the Supporting Information). Acetaldehyde has a greater diversity of indoor sources, including event-specific emissions like cooking and the use of consumer products in addition to building material emissions. Results suggest the overall household acetaldehyde emission rate was not strongly affected by boundary layer dynamics of storage materials in the study homes. Other VOCs sampled generally did not appear to be affected by boundary layer dynamics in their response to ventilation (Willem et al. 2013).

The time constant kL is the product of the transport coefficient k and the loading factor L. Assuming a loading factor L of 0.5 to 1 m^2/m^3 (Hodgson et al. 2005), the range of kL found for formaldehyde corresponds to a transport coefficient k between 0.05 and 0.46 m/h: within the range of 0.011 to 3.6 m/h reported for individual materials (Myers 1984).. The lowest lab values were for wallboard sealed to minimize formaldehyde emissions. For measurements in single family and mobile homes, Myers reported *k* between 0.19 and 2.7 m/h. Homes likely contain a range of formaldehyde-containing materials, but the fastest timescales (higher *kL*) will tend to dominate the effective value for a home (Sherman and Hult 2013). The values of *kL* here are comparable to the values of 0.15-0.18 h^{-1} from in-home measurements reported by Hun et al (2013).

The relative humidity in the space can also impact formaldehyde emission rates (e.g., Myers 1985, Parthasarathy et al., 2011). Unfortunately measurements of relative humidity were not made in all VIAQ homes. Based on limited RH information, a relatively low variation in RH was observed with no systematic dependence on air exchange rate. In house H6, average RH levels were 52.3%, 55.1%, and 54.3%, for low, medium, and high ventilation settings. In house H7, the average levels were 55.1%, 53.3%, and 57.2% for the same settings as H6. At all sites, measured indoor temperatures were consistent and the three periods of measurement (three air exchange rates) occurred over a relatively short period of time in each home. Thus it is expected that humidity levels did not vary substantially across the three air exchange rates for each home.

One factor not included when calculating *kL* and C_{eq} is the re-entrainment of contaminant-loaded exhaust air within mechanical ventilation systems. For the ERV used in H5, up to 30% of formaldehyde in the exhaust stream can re-enter the conditioned space through air leakage and adsorption/desorption from the rotary wheel (Hult et al., 2014), lowering the effective ventilation rate through the ERV. If this process were included in Equation (1), the fitted value of *kL* would be lower proportionally to the reduction in the effective *A*.

Does it make a difference whether the concentration-dependent or the constant emission rate model is used to predict how concentration depends on air exchange rate? A constant emission rate is often assumed in the development of ventilation guidelines, but this may bias estimates of how ventilation impacts concentrations of chemicals emitted from building materials. To quantify the impact of using the

CDE model versus assuming a constant emission rate, we used the ratio of the slope of the two curves at the common benchmark of A=0.35 h^{-1} as a metric of air change effectiveness. Taking the reduction in concentration predicted by the constant emission rate as a baseline, the air change effectiveness quantifies what fraction of that reduction is achieved according to the CDE model. Figure 3 illustrates that the CDE model estimates that increasing the air exchange rate is 40-100% as effective at lowering formaldehyde concentrations as the constant emission rate assumption predicts. Although the metric depends on the air exchange rate at which it is evaluated, we are most interested in the behaviour at lower air exchange rates where concentrations are most sensitive to ventilation.

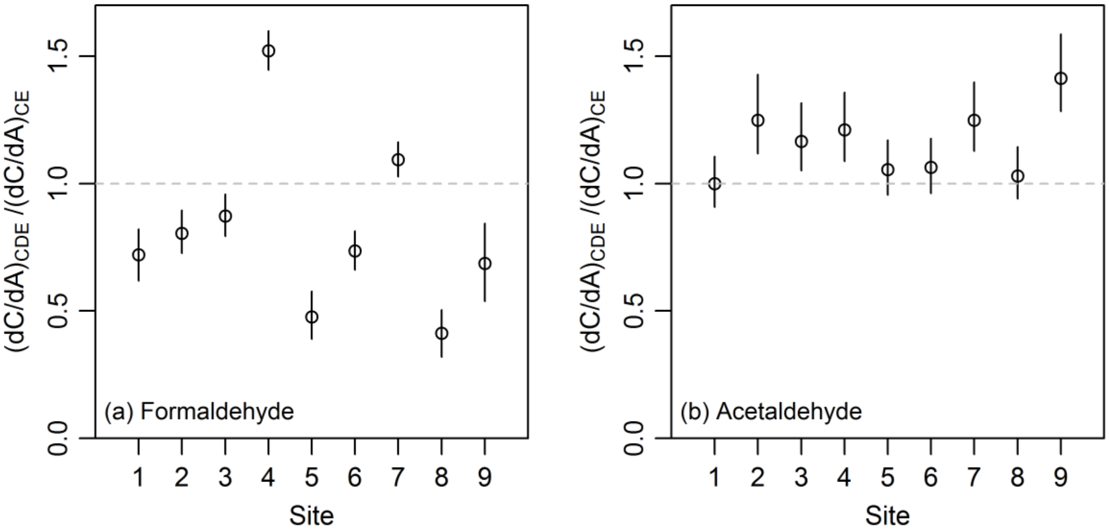

Figure 3 Slope of concentration-dependent emission (CDE) model at A=0.35 h^{-1} as a fraction of the slope of the constant emission rate (CE) model at A=0.35 h^{-1}. Error bars show the 25th and 75th percentile of the results from the Bayesian model simulations.

For acetaldehyde, on the other hand, this metric tends to be closer to 1 (typically slightly above), as the CDE model predicts similar dependence on air exchange rate as the constant emission rate model. This difference suggests that regular, episodic emissions of acetaldehyde may be more important than emissions from building materials. While the boundary-layer buffering process is likely to occur for acetaldehyde, the response of airborne concentrations to ventilation may be dominated by episodic emissions. Episodic emissions can generate the same signal as a constant emission source, as viewed by daily concentration measurements. It is not clear why the air exchange effectiveness is consistently slightly greater than one for acetaldehyde, but generally the difference from 1 is not statistically significant. In H4, a recent application of furniture polish may have temporarily increased the acetaldehyde level; this may have obscured the response to ventilation control. Overall, day-to-day variation in acetaldehyde emission did not dominate the signal, because the concentration dependence on air change rate was generally well approximated by the constant emission rate model.

Source control

Table 2 ventilation rate, concentration and emission rate (LOEM homes)

ID	Ventilation rate (h^{-1})	Formaldehyde average indoor concentration ($\mu g\ m^{-3}$)	Formaldehyde emission rate ($\mu g\ h^{-1}\ m^{-2}$)	Acetaldehyde average indoor concentration ($\mu g\ m^{-3}$)	Acetaldehyde emission rate ($\mu g\ h^{-1}\ m^{-2}$)
G1	0.15	46.1	18.5	48.6	19.4
G2	0.08	38.5	8.1	46.0	9.7
G3	0.19	44.1	22.9	44.4	23.1
G4	0.74	22.8	46.5	16.1	32.8
G5	0.31	38.0	32.2	37.7	32.0
G6	0.14	67.4	25.2	55.4	20.7
G7	-	73.0	-	29.2	-
G8	-	45.4	-	36.3	-
G9	0.09	38.4	9.8	37.5	9.5
G10	0.03	44.8	3.6	52.0	4.2
G11	0.27	50.0	36.4	36.2	26.4
G12	0.39	28.4	30.6	13.9	15.0
G13	0.48	33.4	44.0	18.6	24.5
C1	0.11	60.3	19.0	38.7	12.2
C2	0.07	100	20.6	46.9	9.6

The mean air exchange rate for Indoor airPLUS homes was 0.26 h^{-1} (SD 0.24 h^{-1}) and three homes had ventilation rates greater than the common benchmark of 0.35 h^{-1} (G4, G12, and G13). An open external door during sampling contributed to the high ventilation rate in G4. G13 was the only home to have the mechanical ventilation system set to run continuously on minimum rate without recirculation. No ventilation data were collected in two homes (G7 and G8) because of events that prevented homeowners from setting up the PFT vials.

To compare the impact of building materials on indoor formaldehyde concentrations, a curve of the form of Equation (1) was fit to the unadjusted data from homes built with low-emitting materials, and then to homes built with conventional materials, primarily from the CNHS (see Table 1). Figure 4(a) shows the measured formaldehyde concentration as a function of air exchange rate for the low-emitting and conventional building material groups, while Figure 4(b) shows the concentrations adjusted to a common temperature, humidity and house age. For as-measured concentrations at an air exchange rate of 0.35 h^{-1}, fitted curves give indoor concentrations of 46 $\mu g\ m^{-3}$ for conventional homes and 34 $\mu g\ m^{-3}$ for homes built with low-emitting materials. This corresponds to a 27% reduction (±11 percentage points) in concentration for the low-emitting materials below conventional construction. The uncertainty in the percentage difference was calculated using bootstrapping, selecting concentration values from the dataset with replacement and refitting curves for 10,000 resampling iterations. Results suggest that the benefit of using low-emitting materials is even greater at lower air exchange rates: below 0.75 h^{-1}, the two curves diverge as the air exchange rate decreases. At 0.1 h^{-1} there is a 55% reduction (±8 percentage points) for low-emitting materials below the level of conventional materials.

Figure 4(b) shows indoor formaldehyde concentrations adjusted to consistent conditions of 23.2°C , 43% relative humidity, and an age of 2.25 years (or older) using Equations (5) and (6). Least squares fits to the adjusted data for low emitting and conventional groups indicate that the concentration in low-emitting homes was 42% lower than in homes built with conventional materials at an air exchange rate of

0.35 h^{-1}, with an uncertainty of 10 percentage points from bootstrapping. As in the unadjusted case, the impact of low emitting materials is even greater at lower air exchange rates, with a 60% reduction at 0.1 h^{-1} (±7 percentage points).

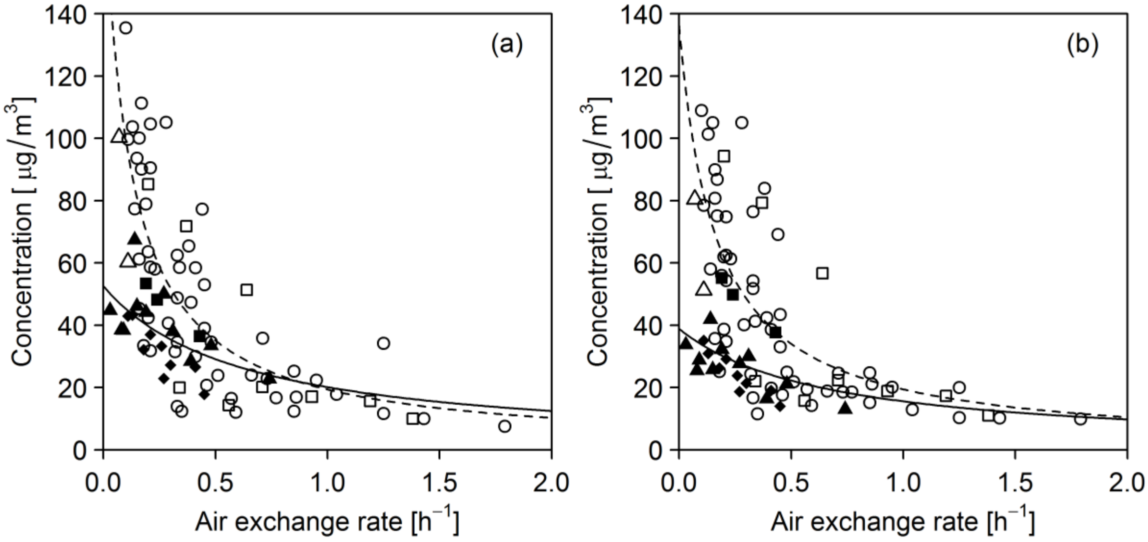

Figure 4 Formaldehyde concentration (above outdoors) measured in homes built with conventional materials (open symbols) and low-emitting materials (solid symbols). In (a), data are not adjusted, and in (b), data are adjusted to consistent conditions: 23.3°C, 43% RH and 2.25 years old. Conventional California New Homes Study data (o), LOEM control homes (Δ) and conventional homes from the VIAQ study (□). Low-emitting category includes LOEM Indoor airPLUS homes (▲) VIAQ homes with certified low-emitting materials (■) and VIAQ low-emitting materials were requested but not verified (♦). The dashed line is the least squares fit of Equation (1) fitted to the conventional home data, and the solid line is fit to the low emitting home data.

While temperatures were higher on average in conventional building material homes, those homes were also older at sampling. The curve fit to low-emitting homes would benefit from additional measurements at higher air exchange rates. But at lower air exchange rates typical of occupied home conditions, on average low-emitting homes clearly have lower indoor formaldehyde concentrations. There is significant variability about the fitted curves, particularly in the conventional building materials group.

One difference not accounted for here is that the CNHS used 24-hr active sampling, whereas the VIAQ and LOEM studies collected samples over 40-min periods. Although formaldehyde concentrations tend to vary diurnally, the variation is in large part due to changes in temperature and relative humidity, which we have attempted to control for in Figure 4(b). One outlier in the low-emitting home grouping is represented by three filled squares near the curve fitted to the conventional construction group. This is H1 from the VIAQ study, which had certified low-emitting materials including wet surface finishing but the carpet materials and backing were not certified as low emitting.

Results suggest that building with low-emitting materials is much more effective at reducing formaldehyde than acetaldehyde in new homes. From bootstrapping analysis, acetaldehyde concentrations in low-emitting homes were on average 38% higher than in conventional homes, but with uncertainty of 23 percentage points (see Figure SI.3 in the Supporting Information). This result is consistent with the hypothesis that acetaldehyde emissions in the study homes were dominated by episodic events rather than emissions from building materials. Acetaldehyde data was not adjusted for temperature, RH or house age since these adjustments are not appropriate for episodic emissions.

Conclusions

Results provide insight on the extent to which increasing ventilation rates and controlling the type of building materials used in home construction can decrease indoor formaldehyde concentrations in new homes. As the ventilation rate is increased the observed reduction in contemporary formaldehyde concentration was less than would be predicted by a constant emission rate model. The reduction based on data from this study is estimated to be up to 60% at a reference air exchange rate of 0.35 h^{-1}. This observation is consistent with material storage theory and controlled laboratory studies: that the formaldehyde emission rate increases as room air concentrations decrease, offsetting some of the reduction that would result if emissions were constant. A higher emission rate would tend to accelerate source depletion that can result in longer-term reductions in indoor formaldehyde concentrations. Considering the relatively small number of homes sampled, inconsistencies in achieving the target range of air exchange rates, and the potential impacts of activity related sources in the few homes that were inhabited during sampling, there remains some uncertainty in the typical magnitude of the effect of source material buffering on the response of indoor formaldehyde concentration to increased ventilation. This uncertainty is reflected by the range in the response curves resulting from the Bayesian Model simulations in Figure 2. To reduce the uncertainty in the response to ventilation, we recommend additional measurements in unoccupied, furnished homes over substantially varied ventilation rates (i.e., at target ventilation rates of 0.2, 0.4 and 0.8 h^{-1}) to further investigate this effect. There was no evidence of increased acetaldehyde emission rate in response to increased air exchange rate.

After adjusting for temperature, relative humidity and home age, formaldehyde concentrations in homes built with low-emitting materials were 42±10% lower than in homes with conventional building materials at a reference air exchange rate of 0.35 h^{-1}. Unadjusted results indicate a 27% reduction (±11 percentage points) in low-emitting homes below levels in conventional homes. Results varied by home; this may be due to the range of materials and furnishings used in conventional homes as well as the addition of formaldehyde sources, such as new furniture, in the low-VOC homes. The benefits of source control appeared to be larger at lower air exchange rates. Unadjusted acetaldehyde concentrations were higher, though not significantly so, in homes built with low-emitting materials. To refine the assessment of benefits of low-emitting construction and finishing materials, a study could examine homes of similar design and material loading, all of similar age and with similar environmental conditions, but with half constructed with conventional and half with low-emitting materials.

Neither method alone reduced formaldehyde concentrations below the California OEHHA chronic and 8-hr reference exposure levels of 9 µg m^{-3} (OEHHA 2013), so combining source control with ventilation is recommended. While we believe source control is the preferred option for robustness, increasing ventilation is a suitable response for any home found to have higher than tolerable formaldehyde levels. Increasing ventilation for at least the first several months and possibly for 1-2 years may be a suitable measure for all new homes built with conventional materials.

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