Diel variation of formaldehyde levels and other VOCs in homes driven by temperature dependent infiltration and emission rates

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ABSTRACT

High time resolution monitoring of formaldehyde and other volatile organic compounds in the air of four homes in winter and summer revealed diel variation of VOC levels driven by infiltration and temperature dependent whole house emission rates. In unoccupied homes, these pollutants displayed a large diel concentration variation, with an afternoon maxima and early morning minima. VOC abundance lagged about 2 h behind changes in infiltration rates measured by a tracer release method, resulting in poor correlations between VOC concentration and air change rate. The data demonstrate that VOC abundance was not in steady state with respect to whole house emission rates. Formaldehyde and other VOCs displayed a positive correlation with indoor temperature in both winter and summer. Formaldehyde sensitivity to temperature ranged from 3.0 to 4.5 ppbv per °C, a useful metric for predicting the impact of heat waves and changing regional climate on indoor air quality. Gypsum wallboard used as radiant ceiling heating product in one home was identified as source of formaldehyde and potentially mercury.

1. Introduction

Poor air quality, both indoors and outdoors, is noted as a leading cause globally of non-communicable disease and mortality [1]. Exposure to chemicals in the indoor environment is important for understanding the impact of air quality on non-communicable diseases, such as asthma [2,3] and cardiovascular diseases [4]. Interestingly, exposure to airborne chemicals including elevated carbon dioxide has been implicated in reduced cognitive function and neurological health [5–9]. Inhalation exposure to chemicals in residential dwellings arises from off gassing of chemicals from building materials and furnishings [10–17], from the use of household chemical products [18–23], and from human activities such as cooking [24–29]. Chemical reactions indoors, such as the reaction of ozone with carpets [30] and human skin lipids [31,32], and radical gas phase reactions [33], are also potential sources of volatile organic compounds such as formaldehyde. Adequate ventilation of buildings should reduce concentrations of airborne chemicals that have indoor sources, but in the case of formaldehyde, a clear relationship between concentration and air change rate has not been established. Residential indoor concentrations of formaldehyde have been reported to display a strong negative correlation with air change rates [34–36] or no apparent correlation with air change rates [37,38]. This lack of clarity is in part attributable to the typical temporal resolution of indoor air studies where passive samplers and integrated sampling over long time periods obfuscates connections between pollutant levels and physical and chemical processes, including human activities that can be both a source of chemicals and variation in building air change rates.

Indoor air quality, ventilation, and building energy consumption are linked issues. Increased ventilation rates come at the cost of a greater energy requirement for heating and cooling needs. Residential buildings in the US are estimated to account for 20% of total national energy consumption and related CO_2 emissions [39]. In the US, ventilation requirements for residential dwellings are covered by ASHRAE standard 62.2 that recommends 0.35 air changes per hour (ACH), but it is not clear if this leads to acceptable levels of air quality for homes. A conundrum exists between the goal of reducing building energy needs that often involve making buildings more airtight and the public health goal of reducing our exposure to chemicals that make us unhealthy and less mentally fit. Efforts to mitigate climate change by reducing building energy needs through the construction of more energy efficient buildings, such as net-zero energy homes, must consider the potential public

health cost of increased chemical exposure. In the construction of net-zero energy homes where infiltration rates may be as low as 0.02 hr^{-1}, it is important to choose building materials with low outgassing rates paired with a reliable and energy efficient mechanical ventilation system to control indoor air pollutant concentrations [40]. Outgassing rates of volatile organic compounds (VOCs) from building materials are known to be temperature dependent [41–47], but there are few reports describing how indoor air VOC concentrations vary with temperature. For Chinese dwellings it has been reported that formaldehyde concentrations displayed a strong temperature dependence [48–51], while no dependence was found in a large study of US homes [37]. The difficulty in understanding the role of indoor temperature comes from the fact that indoor temperature can affect both infiltration rates [52,53] and VOC emission rates from materials, producing opposing effects on indoor air concentration.

The objective of this study was to better understand how climate change may impact indoor air quality of the US residential housing stock through meteorological influence on building infiltration rates and temperature dependent off-gassing rates from building materials. This paper describes the results of high temporal resolution monitoring of air change rates and concentrations of several air toxic compounds, including formaldehyde, acetaldehyde, benzene, and methanol, from four homes in eastern Washington, USA. The high time resolution VOC measurements allowed us to study the dependence of pollutant concentration on indoor temperature and infiltration rates.

2. Materials and methods

2.1. Description of homes

Houses were chosen in this study to provide a range of ages and design characteristics that represent typical detached home types listed in the NIST database of 209 representative residential building types in the US [54], with the idea in mind that older houses may off-gas at a lower rate or may off-gas different types of VOCs and other pollutants, and that newer homes may off-gas at a higher rate and may have lower air change rates (ACH). In this paper we focus on the results from homes H2, H6, H7, and H10 where clear patterns were evident in the data and provided opportunities to examine relationships between VOC abundance, infiltration rates, and indoor air temperature. General characteristics of the homes are listed in Table 1. These homes were located in the town of Pullman, WA and nearby communities. These communities would be considered rural, and outdoor air concentrations of pollutants were typically low except during wildfire events when $PM_{2.5}$ can be very high [55]. For all homes, measurements were performed twice, for a 1-week period in summer (August or September) and a 1-week period in winter (January or February). All the homes had attached garages and had a range of ventilation types and heating appliances. H2 and H7 had a central forced air system (CFA) that could provide heating and air conditioning. H7 also had an auxiliary fresh air intake (FAI) vent that periodically provided fresh air flow. H6 and H10 did not have a central air system and relied on electrical baseboard heaters for heating (H6) or radiant ceiling heating (H10), common designs for homes built in the Pacific Northwest of the US during the period 1960–1980. In the US it is estimated that approximately 35% of

homes do not have a central forced air ventilation system [56].

2.2. Measurement methods

A suite of instruments was used to measure both indoor and outdoor air pollutant concentrations. These instruments were typically located in the garage. Indoor and outdoor measurements were alternated by using a three-way solenoid valve (Galtek, USA) that connected the instruments to either a 1/2″ OD PFA sample line from inside the house or a 3/8″ PFA sample line that went to inlet tripod mounted on the house roof. The tripod also supported a mast for a surface weather station (Airmar 200WX). Indoor and outdoor sampling was switched every 15 min. For some houses, the garage air and attic air were also sampled on a 4-h measurement interval. The indoor inlet was run along the ceiling to a location chosen to be relatively open that connected several spaces within the house (i.e., kitchen-hallway-living room). The inlet was mounted so that it was several inches from the ceiling surface. From these indoor and outdoor inlets, continuous measurements of O_3, CO, NO_x (Teledyne Inc.), CO_2, H_2O (LI-840a, LI-COR), and selected VOCs (PTR-MS, Ionicon Analytik) were performed. Outdoor measurements of $PM_{2.5}$ mass concentration (DustTrak II 8530, TSI) were also made from a separate copper tubing inlet mounted on the roof tripod. Additional equipment was located indoors and mounted in a rack placed in the living room to measure temperature, $PM_{2.5}$ mass (DustTrak II 8530, TSI), PM number concentration (DC1100, Dylos), CO_2 and H_2O (LI-840a, LI-COR). Five wall mounted devices measured CO_2 (K30, Senseair), RH, and temperature and these were distributed throughout the home. Sensors for measuring window and door openings and movement of people were also installed [57].

A PTR-MS instrument measured selected VOCs, typically 30 ions were monitored. The drift tube was operated at 120 Td, with an O_2^+/H_3O^+ ratio less than 2% and a NO^+/H_3O^+ ratio less than 0.2%. The drift tube chamber was maintained at 60 °C and the drift tube pressure at 2 mbar. The PTR-MS response was calibrated with compressed gas standards with a stated accuracy of 5% (Scott Marrin, CA and Apel-Reimer Environmental, FL, USA) which were dynamically diluted to about 20 parts per billion (ppbv). A permeation source (KinTek, TX, USA) was used to calibrate PTR-MS formaldehyde response as a function of water vapor concentration [58]. Formaldehyde response factors used for indoor and outdoor data processing were calculated separately to account for the difference in indoor and outdoor water vapor levels. Formaldehyde has a small positive inference resulting from methanol and ethanol reactions with O_2^+. This interference was accounted for (typically < 5%) but could be significant at times (30%) in some homes when very high concentrations of methanol and ethanol occurred as a result of human activities. The instrument background signal was determined automatically by sampling zero air made by catalytically scrubbing ambient air [58]. Dissociative protonation reactions are common at 120 Td [59] and can be problematic for accurately determining the abundances of some compounds. Notably benzene suffers from a positive interference from ethylbenzene, cumene, and n-propyl benzene, compounds that are often present together due to their common source from automobile exhaust and gasoline vapor. We estimate that benzene levels in our measurements, in particular of garage air, may be overestimated by as much as 8% due to interferences

Table 1
Selected characteristics of homes investigated in this study.

Home	Year Built	Occupants (adults/children)	Living space (ft^2)	Ventilation type	Home Heating	Fireplace- Range[a]
H2	1963	2/0	1765	CFA	G	G-E
H6	1958	2/0	1804	no CFA	Baseboard	NA-E
H7	2010	1/0	1051	CFA -FAI	G	NA-E
H10	1972	2/1	2116	no CFA	Radiant ceiling	NA-E

[a] G = gas, E = electric, NA = none.

from compounds in gasoline vapor.

2.3. Air change rate measurement

The air change rate per hour (ACH) was determined from periodic release of CO_2 into the home via the central forced air system. Pure CO_2 was injected into the furnace supply duct, typically every 2 h, by flowing 20 SLPM of CO_2 gas for 2–3 min. CO_2 was continuously measured in the return duct (LiCor 840a) as a measure of the overall indoor level. The furnace fan continuously ran to move and mix the air throughout the home during tracer release experiments. The CO_2 increase after injection was typically between 250 and 300 ppmv. The ACH was determined from the rate of change of the CO_2 concentration with time due to the exchange of air from outdoors [60]. A box model analysis was applied to the CO_2 decay data to determine the ACH. Inputs to the model included the measured outdoor air concentration of CO_2, the assumption that CO_2 is conserved, and a human respiration source term. We used a human CO_2 respiration of 31.2 g h^{-1} during nonsleeping periods and 20.8 g h^{-1} during sleeping periods [61]. Periods, when the homes were not occupied, provided a clear picture of the ACH and were used to verify that the human respiration values used for occupied periods were appropriate. Given the uncertainty in the human respiration values, we estimate the uncertainty of the ACH during occupied periods was ∼20%.

2.4. Gypsum wallboard chamber test

Emission rates for the gypsum wallboard used in home H10 radiant ceiling heating system were measured in the lab using an emissions testing chamber. The size of the gypsum wallboard sample was 0.065 m^2. The sample was placed in a 150-L Teflon film chamber, as described in Toro et al. [62]. Zero grade air humidified to ∼45% RH flowed through the chamber at 10 L min^{-1} and fans inside the chamber provided constant mixing. VOCs, O_3, and SO_2 were measured from the chamber outlet by PTR-MS, an O_3 analyzer (Teledyne T400), and an SO_2 analyzer (Teledyne T100U). The gypsum wallboard was placed inside the chamber at room temperature of 25 °C and then heated to a 31 °C surface temperature using the native resistive wiring in the sample. Measured ceiling temperatures in the home using a non-contact IR thermometer were around 30–30.6 °C so the chamber test results should represent the actual condition in H10. This chamber test was repeated with the same setup except using zero air containing 100 ppbv NO to verify the measured O_3 signal.

3. Results and discussions

3.1. Diel variation of pollutant levels in unoccupied homes

Measurements made in unoccupied homes provided important data for understanding whole house emission rates and resulting air pollutant levels. Human activities in the home can be a source of these chemicals and can influence ventilation rates through window and door openings, obscuring relationships between indoor air concentrations and ACH. Fig. 1 displays the temporal variation of indoor levels of formaldehyde, acetaldehyde, methanol, and benzene, the measured ACH, and indoor and outdoor temperatures for H2 in summer during an unoccupied period with windows and doors closed. The air pollutant mixing ratios clearly varied with time of day. Highest mixing ratios occurred in the early afternoons and the levels decreased by about 40% to an early morning low. This diel trend in indoor pollutant levels appears to be driven by the factor of 2 diel variation in infiltration rates, ranging from an afternoon low of ∼0.2 hr^{-1} to early morning high values of 0.4 hr^{-1}. This diel variation in infiltration is driven principally by the variation of the indoor-to-outdoor temperature difference and is consistent with our understanding of how infiltration rates are influenced by temperature gradients across the building envelope.
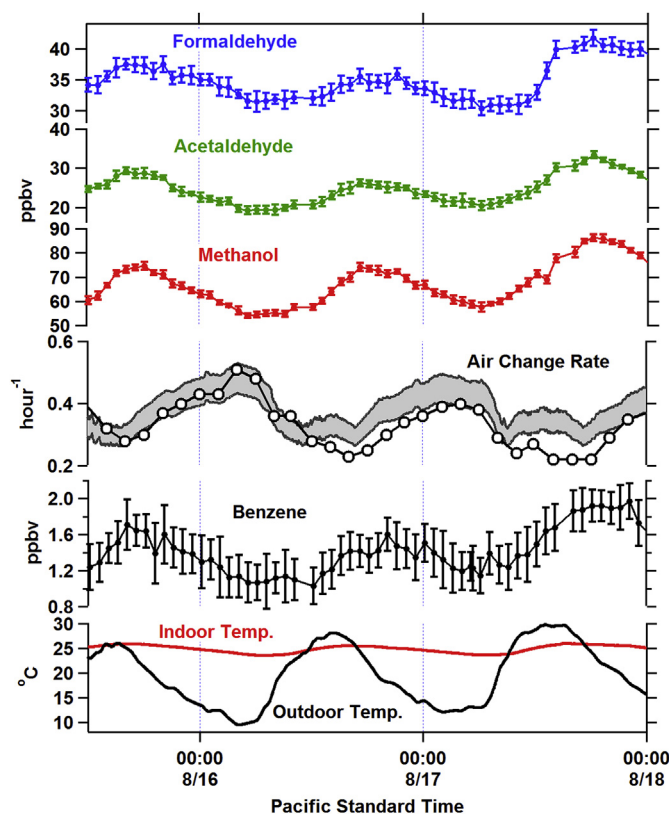

Fig. 1. Time series showing pollutant mixing ratio, indoor and outdoor temperature and measured ACH during an unoccupied summer period in home H2. VOC data are shown as 15 min averages with standard deviation. Expected infiltration rate due to the difference in indoor and outdoor temperature and wind speed as parameterized by Lamb et al. [53] is shown by black shading and is a reasonable match to observed values and their diel variation.

Lamb et al. [53] measured air change rates for homes in Pullman, WA and parametrized ACH with an empirical model using temperature differences and wind speed as variables. This meteorological parameterization was used to infer ACH for H2, as illustrated by the black shading in Fig. 1. The shading represents the 10% uncertainty range of the infiltration rate parameterization. The predicted infiltration rate yields a diel variation and values consistent with our measurements. The diel variation was largely driven by the difference in temperature across the building envelope. Wind speeds were low and accounted for less than 10% of the overall ACH diel variation. The continuously changing infiltration rates over the day created a dynamic indoor environment, and indoor VOC mixing ratios were not constant with time.

Close inspection of the data in Fig. 1 reveals a time lag of ∼2 h between the daily minimum indoor pollutant levels and the daily maximum of ACH. The lag time is related to the characteristic timescale for a compound to reach a steady state concentration. This characteristic time is a function of the ACH and the compound's first order loss rate inside the home due to irreversible surface uptake or homogenous gas phase removal. Of the compounds shown, only formaldehyde is thought to have a significant first order loss rate, on the order of 0.4 hr^{-1} [63,64], similar to the loss rates from dilution caused by infiltration. For compounds like methanol and benzene that are not thought to have appreciable loss rates inside the home, the characteristic timescale to reach steady state is determined by the infiltration rate. The lag between VOC abundance and ACH results in a relatively poor correlation between VOC mixing ratios and ACH as shown in Fig. 2. The left hand side panels of Fig. 2 show the average VOC mixing ratio over the 2 h ACH determination period plotted against measured ACH. Averaging the VOC data over a 2 h period following the ACH
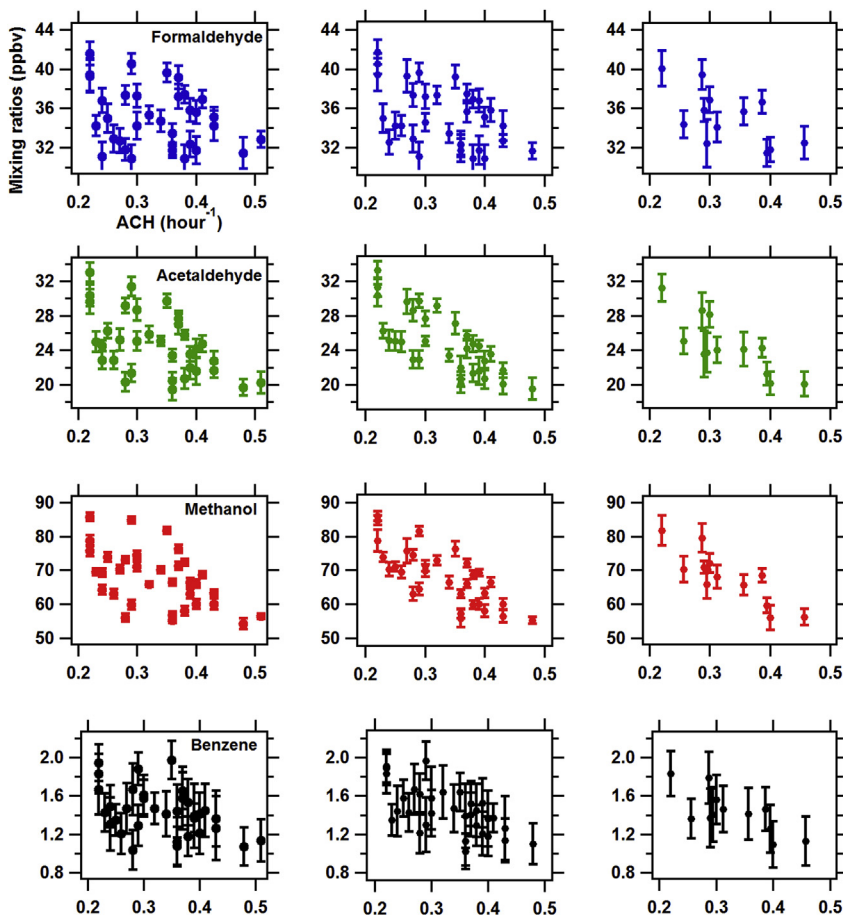
Fig. 2. The correlation between VOC abundance and measured ACH for the unoccupied period in home H2 as illustrated in Fig. 1. Left most panels show the resulting correlation when VOC data are averaged over the 2-hr ACH determination. Center panels illustrate the correlation if the VOC data are averaged over a 2-hr period following the ACH determination. Right most panels illustrate correlations for 6-hr averages that smooth out the lag time difference.

determination, to account for the apparent lag time, yields an improved correlation, as shown in the center panels of Fig. 2. A clear negative correlation can be observed for acetaldehyde, methanol, and benzene but the formaldehyde data are still quite scattered. One reason for this might be that the formaldehyde physical loss rate in the home is sufficiently large that it is the dominant factor in determining concentration-time trends. The right hand panels of Fig. 2 illustrate that a 6-h averaging period improves the correlation between VOCs and ACH. This averaging period was long enough to smooth out the time-lag differences. Longer time averaging hides the lag time inherent in the data and the fact the system was never in steady state. It is a common assumption in the indoor air quality literature to assume steady state conditions for measured pollutants so that concentrations can be related to emission rates through a simple analytical box model [36,65–69]. For homes where ACH is determined by meteorologically modulated infiltration rates, steady-state pollutant concentrations are unlikely to be achieved and thus the relationship between indoor pollutant levels and emission rates is more nuanced. Assuming steady state conditions is likely a poor assumption for most homes.

Fig. 3 illustrates another unoccupied period in a different house, H7, where the diel variation of formaldehyde and other pollutants was observed as a result of diel variation infiltration rates. In this house, the CFA had a fresh air intake (FAI) vent. This FAI was left continuously open for the first part of the monitoring period then closed for a 24-h period as a deliberate manipulation of air change rates to examine the impact on pollutant levels. During the unoccupied period with the FAI open, the measured ACH values were about 0.3–0.4 hr^{-1} greater than infiltration rates predicted from the meteorological parameterization. Even with the FAI vent open, a diel variation in ACH was observed due to the contribution of infiltration. The diel variation in ACH was qualitatively consistent with the time of day variation in infiltration rates

due to the changing temperature gradient across the building envelope. Normally the FAI vent is programmed to periodically open for a few minutes to allow fresh air flow into the home, so its typical impact would be much less pronounced than shown here. Measured ACH values for the period when the vent was open were between 0.54 and 0.95 hr^{-1}. With the FAI vent sealed shut, the measured ACH and model infiltration values were in fair agreement except at night when the homeowner opened a bedroom window. During the unoccupied period, the lowest formaldehyde values occurred around sunrise (~05:40 PST) and maximized shortly after 16:00 PST, similar to the pattern observed in H2. When the FAI vent was closed on Sept 27th, the average afternoon (14:00–16:00) formaldehyde levels increased by a factor of 1.7 compared to the previous afternoon, whereas methanol and benzene tripled in abundance, and acetaldehyde doubled. The much greater change in the abundance of methanol, benzene, and acetaldehyde to changes in ACH illustrates that formaldehyde levels are less sensitive to the variations in ACH than these other compounds.

3.2. Formaldehyde dependence on indoor air temperature

Diel variation in formaldehyde and a consistent day-to-day increase in abundance with warming temperatures were noted in home H10 in summer. Measurements made in this home again illustrated the impact of meteorological variation of infiltration rates on indoor air quality. Fig. 4 illustrates the variation of formaldehyde, infiltration rates, and temperature. H10 did not have a CFA system so infiltration rates were estimated from the meteorological parametrization of Lamb. et al. [53] as was done for H2 and H7. In those homes, the measured infiltration rates matched reasonably well with the estimated infiltration rates, giving us confidence that the meteorological parametrization would reasonably estimate infiltration rates for H10. During the week, the
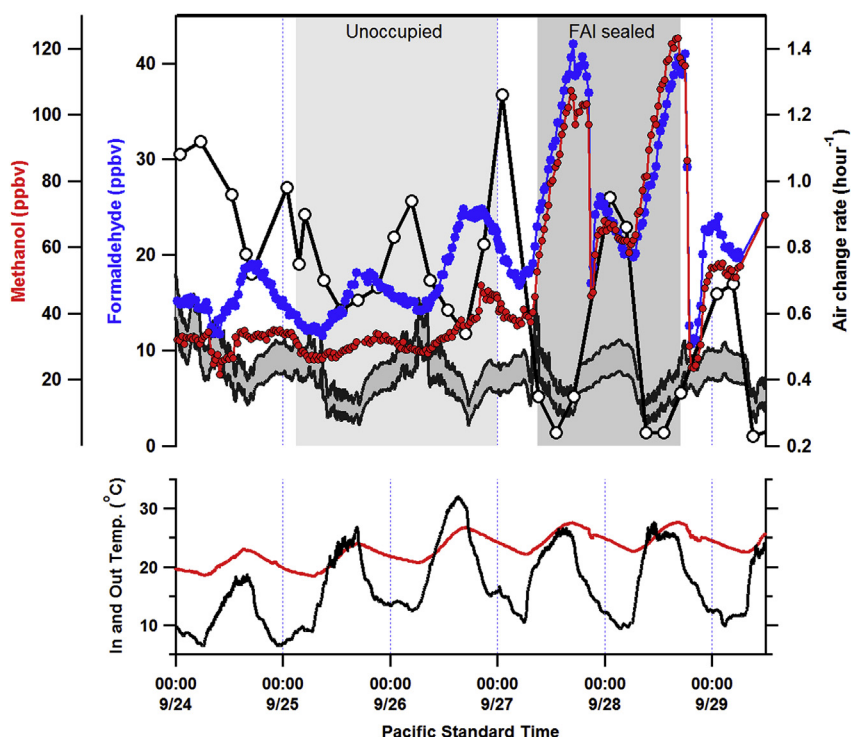
Fig. 3. Time series showing methanol and formaldehyde mixing ratio, indoor and outdoor temperature and measured ACH from home H7 in summer. Light grey shading indicates a period when the house was unoccupied. Dark grey shading indicates when the fresh air intake (FAI) valve was closed. Black shading indicates the range of home infiltration rates predicted by the meteorological model and open circles are the measured ACH. 15-minute averages every ½-hr of indoor formaldehyde (blue circles) and methanol (red circles) are shown. Indoor (red) and outdoor (black) temperatures are shown in the lower plot. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

outdoor afternoon air temperatures climbed steadily from an average of 11.7 °C on Sept 21st to 24.8 °C on Sept 26th. Indoor air temperatures increased correspondingly. Formaldehyde levels in the house tracked indoor temperature and formaldehyde levels steadily built up in the home as it got warmer. Formaldehyde levels were at their highest and reasonably constant from about 18:00 to 24:00 PST corresponding in time to when it was warmest in the house. From Sept 22nd, onward there was a consistent pattern of low air change rates in the afternoon

(0.30–0.35 hr^{-1}) and higher air change rates at night (0.40–0.45 hr^{-1}). For the first 1.5 days formaldehyde levels inside the house were relatively constant at 22 ppbv, attributable to little day-to-night variation in infiltration rates. Indoor formaldehyde levels were about a factor of 20 or more greater than outdoor levels. Outdoor levels displayed a diel variation, with afternoon abundance of ~1.9 ppbv on Sept 26th compared to over 40 ppbv inside the home on that day. The large decrease in formaldehyde occurring on Sept 24th was attributed to the door
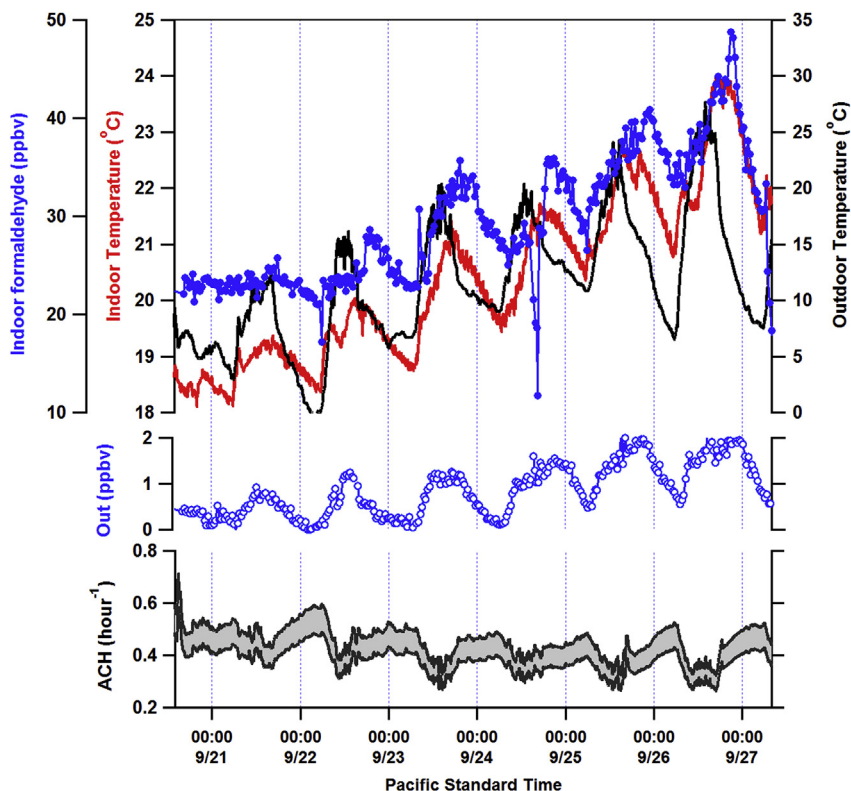

Fig. 4. Formaldehyde time series for H10 in summer showing a steady increase in formaldehyde with increasing indoor air temperature. Indoor formaldehyde (blue circles, upper panel) and outdoor formaldehyde (middle panel) mixing ratios are shown as 15 min averages every ½-hr. Indoor air temperature (red trace) and outdoor (black trace) air temperature are shown in the upper panel. Infiltration rates calculated from the meteorological data are shown in the lower panel. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)
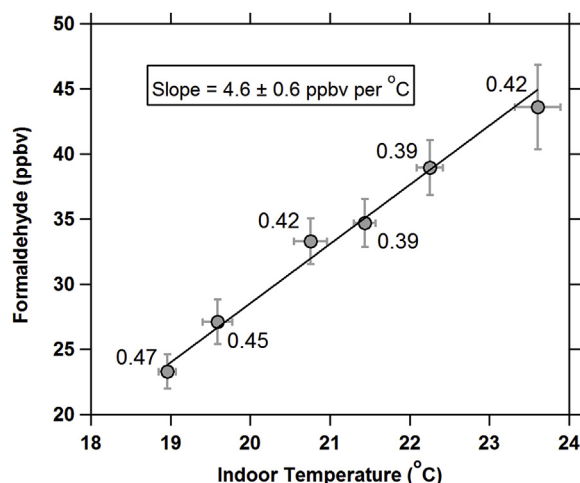
Fig. 5. H10 formaldehyde data showing a correlation between indoor and outdoor difference versus indoor air temperature averaged between the hours 18:00 to 24:00 PST when indoor air temperature and formaldehyde levels were highest. Error bars indicate the standard deviation of the averages. Numeric values adjacent to data indicate corresponding ACH average in units of hr^{-1}.

between the house and the garage being left open as the home owner worked in the garage that Saturday.

The steady increase in indoor formaldehyde was well correlated with temperature and illustrates the temperature dependence of whole house formaldehyde emission rates. In Fig. 5, the difference between indoor and outdoor formaldehyde mixing ratios averaged from 18:00 to 24:00 PST are plotted against the corresponding average indoor air temperature for the six evenings illustrated in Fig. 4. The corresponding estimated infiltration rates are indicated in the plot besides the symbols. The estimated infiltration rates for the six evenings were similar and essentially identical for the last four evenings of the week (Sept 23rd to Sept 26th). The change in formaldehyde levels was attributed to an increasing whole house formaldehyde emission rates as the indoor air temperatures increased. The slope of the plot is 4.6 ± 0.6 ppbv formaldehyde per °C. The 4.5 °C temperature change in the house over the week resulted in a doubling of formaldehyde levels inside the home. This sensitivity to temperature likely reflects whole house emission rate sensitivity from materials in this home including composite wood flooring and furnishings.

In general the relationship between VOC levels and indoor temperature is complex due to temperature's influence on infiltration rates and surface emission rates from materials inside the home. These rates can vary throughout the day, display seasonal differences, and can impact indoor air concentrations in opposite ways. Fig. 6 illustrates the 1-min indoor formaldehyde data as a function of indoor temperature for three of the study homes (H2, H6, and H10) to contrast winter and summer behavior for conditions where infiltration dominated air change rates. We removed data when natural ventilation through window or door openings impacted concentrations. The lowest 10% of

the data associated with a particular temperature range are highlighted to better reveal trends with temperature by avoiding obfuscation from human activity emission events. These highlighted data thus represent conditions with the highest infiltration rates and/or lowest emission rates. For these homes summer time formaldehyde levels increased with indoor air temperature. Lowest temperatures were associated with early morning periods when ACH values were typically greatest. Higher temperatures were associated with afternoon data when infiltration rates were typically lowest, and whole house emission rates would be greatest. The overall temperature trend thus represents the combined influence of these two factors and potentially other factors such as secondary source influence from O_3 infiltration into the home and indoor formaldehyde loss rates. A linear regression through the highlighted summer data display similar slopes: 4.1 ± 0.2 ppbv per °C for H2; 3.0 ± 0.1 ppbv per °C for H6; and 4.5 ± 0.07 ppbv per °C for H10. The slope through the H10 summer data is very close to the 4.6 ± 0.6 ppbv per °C determined from the H10 data in Fig. 5 that reflects whole house emission rate sensitivity. These trends provide a useful simple predictor of the influence of indoor air temperature on formaldehyde levels. A similar temperature dependence has been measured from large surveys of residential dwellings in China [48,49]. In those studies the formaldehyde mixing ratio sensitivity to temperature, assuming standard temperature and pressure conditions, was 5.1 ppbv per °C [49] and 3.5 ppbv per °C [48], very similar to our results.

The winter trend for house H6 is similar to summer with formaldehyde levels typically lower in winter, consistent with expected seasonal difference in infiltration rates. The winter time infiltration rates estimated from the meteorological parametrization ranged from 0.44 to 0.74 hr^{-1} compared to summer's 0.29 to 0.48 hr^{-1} range. In house H10 the winter data curiously lay atop the summer data with a slightly steeper slope (5.9 ± 0.2 ppbv per °C), likely as a result of our sample inlet being close to an important wintertime formaldehyde source in the home, the radiant ceiling, discussed in section 3.3. The steeper slope may reflect the influence of the radiant ceiling as an extra source in winter compared to summer. The winter data for house H2 display a much weaker dependence on temperature (0.58 ppbv per °C) with overall much lower home temperature and lower formaldehyde than in summer. During the winter measurement period, the home was largely unoccupied, and the thermostat was set back to save energy. As a result, the home was quite cool. The home had relatively high measured ACH that could not be explained by infiltration driven by meteorology. High ACH may have been driven by unintended natural ventilation as a result of a fireplace damper being left open or a "doggy door" being ajar. Low whole house formaldehyde emission rates as a result of lower indoor temperatures coupled with relatively high ACH resulted in much lower concentrations of formaldehyde and VOCs for this home compared to other homes in winter. The slope through the winter data is a factor of 7 less than the summer trend. The lower sensitivity to temperature in winter is likely a consequence of the home ventilation being driven by natural ventilation rather than infiltration driven by temperature gradients across the building envelope. We postulate that at some point indoor temperatures might become cool
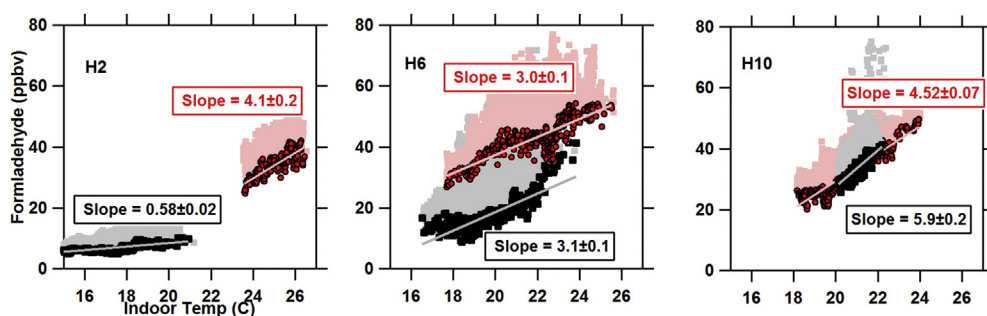

Fig. 6. Indoor extra formaldehyde levels (indoor-outdoor) as a function of indoor air temperature for homes H2, H6, and H10. Shown are 1-min data for summer (red circles) and winter (black squares). Some H10 winter data are off-scale. Data are selected for periods where infiltration dominated air exchange (natural ventilation periods excluded). Lowest 10% of the data points are highlighted for trend analysis. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

enough that surface emission rates would be low enough so that the primary formaldehyde sources become less important than secondary sources such as the reaction of O_3 with interior surfaces. This would allow for better qualification of indoor secondary sources caused by ozone surface reactions in homes.

The correlations with temperature for the three other VOCs are illustrated in Figures S-1, S-2, and S-3. The lower 10% of the data were highlighted to discern baseline trends. The summer time VOC data from the unoccupied period of house H2 displayed a clear temperature dependence: 13.4 ± 0.5 ppbv per °C for methanol, 9.6 ± 0.2 ppbv per °C for acetaldehyde, and 0.93 ± 0.04 ppbv per °C for benzene. Again, the temperature dependence for the H2 winter measurements were weaker; the slopes were a factor 10 less for all three VOCs, similar to what was observed for formaldehyde. For home H6 and H10 temperature trends were less clear because of large variability in mixing ratios attributed to home occupancy and human activity emission events such as cooking and household chemical use. For methanol in houses H6 and H10 in summer, the lower 10% of the data displayed a trend with temperature: 6.5 ± 0.3 ppbv per °C for H2 and 19.8 ± 0.4 ppbv per °C for H10, values within $\pm 50\%$ of the H2 trend. For benzene no trend was observed in house H6 in summer but a trend was discernible in the winter data. For house H10 a similar temperature trend was observed for the summer and winter data, ~0.4 ppbv per °C, about half that observed in H2. For acetaldehyde the lowest 10% of the data defined a trend with temperature in house H6 in winter and in H10 in summer. Acetaldehyde in house H6 was strongly and regularly influenced by events associated with cooking, with values as high as 1.5 ppmv as a 10-min average, off scale in Figure S-2. The acetaldehyde temperature sensitivity displayed for houses H2 summer, H6 winter, and H10 summer were similar, but clearly for some homes human activity emissions can be much more important source of this compound than emissions from building materials. The building material emissions act as a "baseline" upon which the human activity emission events play out. These human activity sources may overwhelm the more continuous building emission sources, obscuring trends with temperature that are otherwise expected from material emission rates and infiltration rates.

Other factors may influence VOC emission rates from materials such as relative humidity, which has been shown to influence VOC emissions in chamber testing of materials. Similar to what was found by Hun et al. [37], no trends between VOC abundance and indoor relative humidity was discernible in our data. Variation in VOC concentrations was much larger than variations in indoor relative humidity for unoccupied homes. For example, for house H2 depicted in Fig. 1, the RH varied from 44% to 40%.

3.3. Formaldehyde emissions from heated gypsum wallboard

It has been suggested in the literature that homes with baseboard heating can have higher formaldehyde levels as a result of strong local heating of wall surfaces [35]. H10 had a radiant ceiling heating system. The H10 winter data for formaldehyde and acetaldehyde are shown in Fig. 7 and levels were much higher than expected. During this period, the home was heated with the thermostat set at 20 °C (68 °F). The estimated winter infiltration rates were larger than summer due to larger indoor-outdoor temperature differences in winter and much higher winter wind speeds (up to 12 m/s) that drove much of the hour-to-hour and day-to-day variability in infiltration rates. Estimated winter infiltration rates ranged between 0.5 and 1.5 hr^{-1} compared to summer's 0.3 to 0.5 hr^{-1} range. Despite the greater infiltration rates in winter, formaldehyde levels in H10 were typically higher than in summer, ranging from 30 to 35 ppbv with events to 159 ppbv, compared to summers 20–45 ppbv range. Winter VOC data did not display a diel pattern as there was no consistent diel variation in ACH. However, indoor formaldehyde and acetaldehyde tracked small variations in indoor air temperature. Increases in indoor air temperature (up to 1 °C) occurred at about 18:00 during week days when home owners returned

home from work. Associated with these temperature increases were increases in formaldehyde, acetaldehyde, methanol, and benzene. In other homes, the spikes in acetaldehyde were associated with cooking, but these events in H10 must have an additional source component to explain the covariation with formaldehyde and other VOCs. Interestingly, when these spikes occurred, there was a large response from the ozone analyzer yielding indoor ozone values as high as 143 ppbv for a 1-min average.

We determined these higher formaldehyde and acetaldehyde levels and O_3 spikes were due to the enhanced sources in the home from the ceiling radiant heating system. This commercial heating system consisted of resistive heating wires inside the gypsum wallboard. This type of heating system was commonly installed in Pacific Northwest homes. The homeowner permitted the removal of a sample of the radiant ceiling product from a room within the home, and this was tested in the lab. When heated, the sample emitted more formaldehyde, acetaldehyde, and benzene as shown in Fig. 8. It is known that temperature can impact the emission rates from building materials by affecting the diffusion coefficient, material/air partition coefficient, and initial emittable concentration [42,43,46,47]. The emission rate in µg hr^{-1} increased from 1.7 to 4.0 for formaldehyde, from 3.4 to 5.8 for acetaldehyde, and from 0.08 to 1.3 for benzene. SO_2 increased as well, but we did not measure SO_2 in the homes. A response on the O_3 monitor was also observed. The observed increases of the VOCs and O_3 response were qualitatively consistent with what was observed in the home. There was no evidence of elevated aromatic compounds which at high concentrations can produce a response in UV absorbance based O_3 monitors [70]. The O_3 response was also observed when we repeated the test using zero air flow containing a 100 ppbv NO to titrate O_3 in the chamber. The addition of NO made no difference to the measured O_3 abundance, and we conclude the O_3 monitor was not responding to O_3 but to some other contaminant. The O_3 monitor uses a low-pressure Hg lamp as the light source, and it is known that such UV absorption based instruments will respond to Hg vapor [71]. Tests done with similar instruments revealed that 0.04 ppbv Hg test gas mixtures yielded a 20 ppbv O_3 response [71]. We conclude that this particular gypsum product is a strong source of formaldehyde, acetaldehyde, and potentially Hg when heated. Since our inlet was positioned about 8 cm from the ceiling surface, our measurements were strongly impacted by this source. Other reports have stated that gypsum wallboard products are not strong sources of mercury in the home [72]. However, those tests did not heat the samples as might occur with resistive heating ceiling products or with baseboard heaters that increase the temperature of gypsum wallboard surfaces. Wallboard emissions as a function of surface temperature should be investigated in the future to provide more information on the role of these materials as sources of mercury, acetaldehyde, formaldehyde, and SO_2 in the home.

4. Conclusions

A strong diel variation in the indoor air concentrations of formaldehyde, acetaldehyde, benzene, and methanol, was observed in unoccupied homes. The variation in VOC abundance in the home was caused by time of day variation in infiltration rates. In this study, the infiltration rate variation was principally caused by the time of day variation of the temperature gradient across the building envelope. Maximum VOC concentrations typically occurred in the afternoon when infiltration rates were lowest and minimized during the early morning when they were highest. VOC abundance appears to lag behind changes in infiltration rates so that the correlation between VOC abundance and air change rate (ACH) was poor. The results demonstrate that for homes where infiltration dominates air change rates, the VOC abundance is not in steady state with respect to whole house emissions and ACH. The assumption of steady state is common practice to calculate whole house emission rates from measured indoor air concentrations; this is probably a poor assumption in many cases. We
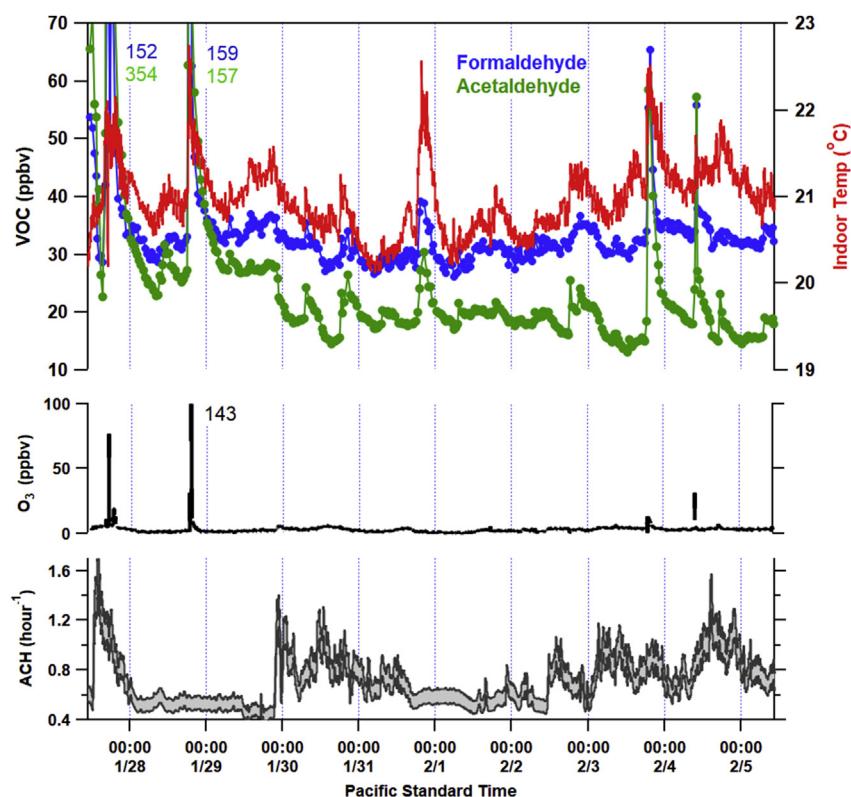
Fig. 7. Time series showing winter indoor formaldehyde, acetaldehyde, and ozone mixing ratios in home H10. Middle plot shows 1-min averages of indoor O_3, top plot shows 15-min averages every 30 min of formaldehyde (blue circles) and acetaldehyde (green circles), and 1-min average indoor air temperature (red trace). Bottom plot shows estimated ACH range due to infiltration (black shading). Highest values of formaldehyde, acetaldehyde, and O_3 are shown as numeric labels for events that are off the scale. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)
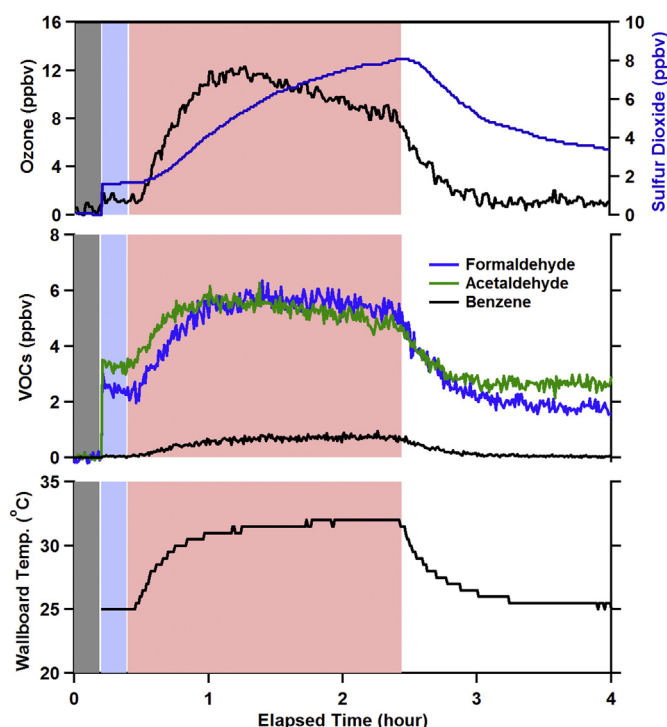
conclude that whole house emission rates need to be determined for homes where constant mechanical ventilation provides the air flow to avoid the complication of the diurnally varying infiltrations rates and resulting non-steady state VOC concentrations.

Formaldehyde and other VOCs were positively correlated with indoor temperature. For one home where ACH was relatively constant and determined by infiltration, a warming weather trend over the week resulted in increasing indoor levels of formaldehyde that tracked increasing indoor air temperature. For this home a whole house formaldehyde emission rate sensitivity of 4.6 ppbv per °C was determined. In general, indoor summertime formaldehyde levels displayed a positive correlation with indoor temperature that ranged from 3.0 ppbv per °C to 4.5 ppbv per °C, accounting for building materials emissions sensitivity to temperature and the impact of indoor temperature on infiltration rates. This is a useful metric for predicting changes in the indoor abundance of formaldehyde to changes in home temperature as a result of thermostat set points, heat wave impacts, and potential impacts of regional climate change on indoor air quality.

Declarations of interest

None.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this chapter can be found online at https://

Fig. 8. Testing of the gypsum wallboard radiant ceiling product. Shading indicates the experimental conditions: empty chamber (grey), unheated wallboard (light blue), and heated wallboard (light red). Heating the wallboard causes a response on the O_3 monitor and increases in SO_2, formaldehyde, acetaldehyde, and benzene. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

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