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Formaldehyde Concentrations in a Net-Zero Energy House: Real-time Monitoring and Simulation

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SUMMARY

Measured real-time formaldehyde concentrations in a net-zero energy house were compared to simulated concentrations from a recently-developed, coupled building energy and airflow/indoor air quality model. Measured and simulated formaldehyde concentrations in living spaces ranged from 4 ppb_v to 10 ppb_v (5 $\mu g/m^3$ to 12 $\mu g/m^3$) while concentrations in the conditioned attic ranged from 13 ppb_v to 28 ppb_v (16 $\mu g/m^3$ to 34 $\mu g/m^3$). During the 15 minutes the heat recovery ventilator was off each hour, the measured concentration in a bedroom increased by 1 ppb_v (1.2 $\mu g/m^3$). In addition, year-long simulations suggest the formaldehyde concentration in the attic may reach almost 50 ppb_v (62 $\mu g/m^3$) during the summer. These results highlight the need for source control and effective ventilation (both outdoor air and air distribution) to reduce the concentration of indoor pollutants, particularly in tighter buildings. This research reaffirms the need to consider buildings as multizone systems and provide adequate ventilation to all building zones, particularly those with low outdoor air change rates.

PRACTICAL IMPLICATIONS

Proper mechanical ventilation is increasingly important as new buildings are constructed with or retrofitted to have lower infiltration rates to conserve energy. Without adequate ventilation, indoor pollutant concentrations can exceed levels of concern even in buildings built with low emitting construction materials.

KEYWORDS

Formaldehyde, Net-zero, Model, CONTAM, TRNSYS

1 INTRODUCTION

Formaldehyde is a known human carcinogen (IARC 2012) and exposure to elevated levels of formaldehyde has been linked with a higher incidence of certain types of cancer in cohort studies. Formaldehyde and formaldehyde-based resins are used in the manufacture of particleboard, plywood, paper products, insulation foam, flame resistant fabric, as well as to mold plastic parts for home appliances and consumer products (WHO 2010). Formaldehyde can be emitted from hydrolysis of formaldehyde-based resins (Salthammer et al. 2010). Its widespread use in man-made products is a major source of formaldehyde indoors. Formaldehyde can also be a by-product of combustion. Secondary formation of formaldehyde can occur through the oxidation of alkenes (especially terpenes) as demonstrated in chamber studies (Singer et al. 2006, Waring et al. 2008).

The World Health Organization (WHO) has set a short-term (30 minute) guideline for formaldehyde exposure of 100 $\mu g/m^3$ (81 ppb_v) to prevent sensory irritation as well as long-term health effects, including nasopharyngeal cancer and myeloid leukemia (WHO 2010). In

comparison, the California Office of Environmental Health Hazard Assessment has set a chronic reference exposure limit (REL) for formaldehyde of 9 $\mu g/m^3$ (7 ppb_v,) (OEHHA 2014). This REL is the concentration at which adverse noncancer health effects are not expected for continuous chronic exposures.

Whether a formaldehyde health standard is exceeded in an indoor environment is primarily dependent upon source strength (emission, reaction, combustion) and outdoor air change rate. Formaldehyde concentrations will be lower in environments with low source strengths and high air change rates. High performance buildings are typically constructed with low source strength materials but may be designed to operate at low air change rates.

Typically, time-averaged methods (e.g., ASTM D5197-09e1) have been used to measure formaldehyde concentrations using derivatizing agent-filled sorbent tubes that are sampled over periods lasting several hours or even days. These methods do not capture variations in concentration at the time scales (seconds to hours) required to understand transient effects such as indoor chemistry that results in formaldehyde formation or the impacts of space conditioning equipment operation. To date, limited knowledge exists of the dynamic nature of indoor formaldehyde concentrations. Most of the research on the influence of environmental conditions (such as temperature and relative humidity) on formaldehyde concentrations has been limited to chamber studies. Measuring real-time, high resolution formaldehyde concentrations is vital to understanding the emission of formaldehyde from building materials, its formation from indoor chemical reactions and the impacts of equipment operation. Measuring real-time indoor concentrations of formaldehyde also allows for the verification of simulation models of indoor formaldehyde concentrations.

Real-time monitoring of indoor ambient formaldehyde concentrations has been limited to Fourier Transform Infrared (FTIR) spectrometry and sensor-spectrophotometric devices. Typically, FTIR spectrometers are limited to a method detection limit (MDL) of about 8 ppb_v (9.8 $\mu g/m^3$) for formaldehyde (Wei et al. 2013). A sensor-spectrophotometric device has been shown to have a 2 ppb_v (2.5 $\mu g/m^3$) MDL and a sampling cycle of one minute (Carter et al. 2014). Recent advancements in laser spectrometry have led to the development of a real-time formaldehyde monitor that has an MDL of 0.1 ppb_v (0.12 $\mu g/m^3$). This instrument was used in this study to measure real-time formaldehyde concentrations in a net-zero energy house and to demonstrate the short term, dynamic impacts of a heat recovery ventilator (HRV) on indoor concentrations. The measured concentrations were also compared to simulation results from a coupled building energy and airflow/contaminant transport model of the house.

2 MATERIALS/METHODS
A net-zero energy test house was monitored in real-time to examine variations in indoor formaldehyde concentrations and to determine if these variations can be captured in models.

Test Facility
The National Institute of Standards and Technology (NIST) constructed the Net-Zero Energy Residential Test Facility (NZERTF) in Maryland, USA in 2012. The facility functions as a laboratory to support the development and adoption of cost-effective net-zero energy (NZE) designs and technologies, construction methods, and building codes. The two-story house with a basement and attic is similar in size (242 m^2 for occupied floors, 485 m^2 inside the building envelope including the attic and basement) and aesthetics to homes in the surrounding communities. The house is not furnished other than permanently installed cabinetry. One key design objective was to provide for occupant health and comfort through

adequate ventilation and reduced indoor contaminant sources. For source control, guidelines were implemented to minimize use of products with urea-formaldehyde resin and elevated emissions of volatile organic compounds (VOC). More information on the NZERTF design and long term monitoring of VOC concentrations can be found in Poppendieck, et. al. (2015).

Several technologies are employed in the house to achieve the net-zero energy goals including a 10.2 kW photovoltaic system, a high efficiency air-to-air heat pump, a solar hot water system, and a heat recovery ventilator (HRV). All floors of the house, including the attic, are within the conditioned space. A central heat pump system provides supply air to all floors except the attic. Passive air transfer grilles connect the basement to the first floor and attic to the second floor of the house. Air is returned to the heat pump via two return air grilles located on the first and second floor. A separate HRV system provides air to the first floor kitchen and second floor bedrooms and draws air for heat recovery from three bathrooms located on the first and second floors. To comply with the outdoor air requirements in ASHRAE Standard 62.2-2010 (ASHRAE 2010), the HRV was sized to deliver 137 m^3 h^{-1} of outdoor air, which is equivalent to an air change rate of 0.11 h^{-1}. Tracer tests performed in the summer and winter showed the infiltration through the building envelope to vary between 0.02 h^{-1} to 0.06 h^{-1} when the HRV was off and the total outdoor air change rate (mechanical ventilation plus infiltration) to vary between 0.17 h^{-1} to 0.19 h^{-1} when the HRV was on. (Ng et al. 2015)

Model
Modelling of the NZERTF was performed using the whole-building multizone airflow and indoor air quality software CONTAM (Dols et al. 2015) coupled with the TRaNsient Systems Simulation Tool (TRNSYS, (Duffy et al. 2009)) building energy analysis software. CONTAM accounts for the interaction between external driving forces (ambient temperature and wind) and internal mechanisms (building mechanical system airflows) to determine resultant pressures and airflows across internal and external building partitions, i.e., interzone and infiltration/exfiltration airflows. It can then account for external and internal contaminant sources and removal mechanisms to calculate contaminant transport associated with the previously determined airflows. TRNSYS has a modular structure that enables multiple energy-related systems to be considered together within a single simulation environment. Modules are referred to as Types. Type 56 implements a whole-building multizone heat transfer model that can account for conductive, convective and radiant heat transfer associated with building materials (e.g., walls, floors, ceilings and windows); interzone and infiltration airflows; and heating and air-conditioning systems that can be simulated using a wide range of existing and/or user-defined modules.

Both CONTAM and TRNSYS Type 56 have limitations. CONTAM relies on user-defined values for internal temperatures to calculate airflows, while Type 56 requires the input of infiltration and inter-zone airflow rates to calculate temperatures. By coupling these two models, the limitations of each can be overcome. During the simulation, data is exchanged between the two models to form the coupled simulation as described in Dols et al. (2015).

The NZERTF was modelled as a four zone building consisting of one zone for each floor including the basement and attic. Model inputs were determined based on building design and measurements. Ventilation system airflow rates including heat pump supply and return; HRV supply and return; and bathroom, range hood and dryer vent exhaust were measured with a balometer or a duct traverse using a hot wire anemometer. A blower door test was performed to measure the building envelope leakage rate. This envelope leakage was distributed over the entire above-grade building envelope in the CONTAM representation. These measurements

and other house properties are provided in Ng et al. (2015) and the TRNSYS representation including the mechanical systems was developed by Leyde (2014) and calibrated with measured data by Balke (2016). Poppendieck, et. al. (2015) previously used a CONTAM-only model to predict infiltration rates by defining all system flows and zone temperatures. In contrast, this coupled model simulated thermostat control of the space conditioning system to establish heat pump operation and calculate zone temperatures.

The average occupied floor area (1st floor and 2nd floor) formaldehyde emission rate over one year (6.7 µg h^{-1} m^{-2}) was measured using one hour 2,4-dinitrophenylhydrazine (DNPH) cartridge sampling according to ASTM D5197 (ASTM 2009) and reported in Poppendieck et. al. (2015). Preliminary investigations indicated that there was likely no significant source of formaldehyde in the basement, but there are potential sources on other levels. The source was modeled as being present in the 1st floor, 2nd floor and attic. Hence, the floor area formaldehyde emission rate (5.1 µg h^{-1} m^{-2}) was normalized to include the attic floor area.

Measurements
A real-time spectrophotometric formaldehyde monitor was placed in the NZERTF for three weeks. The sensitivity of the monitor is 0.1 ppb$_v$ (0.12 µg/m^3) with a one second sampling time. Sampling lines were run from each room to an automatic seven port sampling valve, which fed into the monitor. The monitor recorded the formaldehyde concentration at each location in series for two three-day periods (Session 1 and Session 2). Each location was sampled for two minutes every 15 minutes. The monitor was zeroed every 15 minutes.

Tracer gas decay tests were conducted concurrently with the formaldehyde monitoring. During Session 1 a fan was placed at the top of each stairwell (between basement and 1st floor, and between 1st and 2nd floor) to enhance the mixing of the tracer. During Session 2 a single fan was placed between the 1st and 2nd floor, while the door to the basement was closed. Estimated mixing fan flows were included in the model. Formaldehyde concentrations were similar for floors connected with a mixing fan.

The formaldehyde monitor was also used to individually monitor each of seven locations continuously over day long periods. The monitor recorded data from each location for 12 out of every 15 minutes. The remaining time was used to zero the instrument and record outside concentrations. Relative humidity, temperature and ozone concentrations were separately monitored.

3 RESULTS AND DISCUSSION
The measured formaldehyde concentrations in the 1st floor, 2nd floor, and basement were similar during both measurement sessions (Figure 1). The measured concentrations in the zones ranged from 4 ppb$_v$ to 10 ppb$_v$ (5 µg/m^3 to 12 µg/m^3). These concentrations bracket the OEHHA chronic REL for formaldehyde of 7 ppb$_v$ (9 µg/m^3) (OEHHA, 2015).

The measured formaldehyde concentrations in the attic were two to four times higher than the concentrations in the other zones. The attic in the NZERTF is within the conditioned space, but is only connected to the occupied zone via two passive transfer grills. The tracer decay rate in the attic was two to three times lower than other measured locations in the NZERTF during Session 1. This lower air change rate is consistent with the attic having higher formaldehyde concentrations assuming the equivalent emission rate for all the spaces. In the CONTAM model the formaldehyde emission rate was defined to be evenly distributed throughout the 1st floor, 2nd floor and attic based on floor area. The model data (solid lines in Figure 1) follow the same trends as the measured data, with the attic concentration being two

to three times higher than the other zones. Since the model was assigned the same formaldehyde emission value throughout the 1st floor, 2nd floor and attic, this result indicates that the elevated concentrations in the attic are largely due to the reduced ventilation in the attic compared to the other zones in the house.
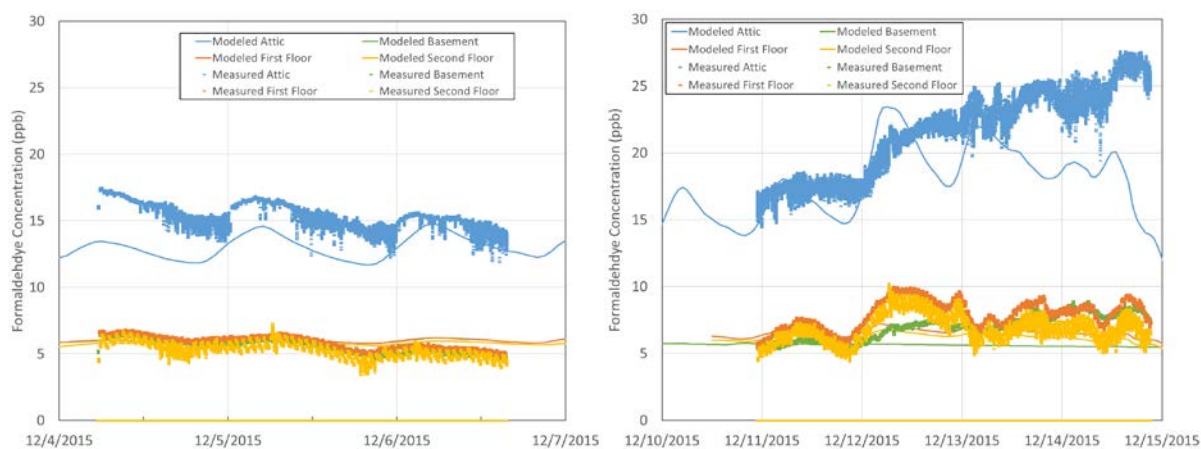
Figure 1. Real time formaldehyde concentration (ppb$_v$) data from two three-day sampling sessions. During Session 1 (left) the average outdoor temperature was 3.5 °C (standard deviation SD=4.4 °C) and the average wind speed was 0.9 m/s (SD = 1.3 m/s). During Session 2 (right), the average outdoor temperature was 12.7 °C (SD = 4.9 °C) and the average wind speed was 0.9 m/s (SD=1.1 m/s)..

The higher average outdoor temperature of 12.7 °C during Session 2 (Figure 1, right) likely resulted in higher formaldehyde concentrations as compared to Session 1 (3.5 °C). The average wind speed was the same for both sessions. As the outdoor temperatures approach indoor temperatures there is a lower driving force for infiltration. Lower infiltration rates should have greater influence on chemical concentrations in zones without direct mechanical ventilation, such as the attic in this house. The modeled emission rates were constant and not adjusted for indoor temperature changes. This indicates that the greater increase in formaldehyde concentrations during Session 2 in the attic compared to the rest of the zones is likely the result of reduced outdoor air change in the attic.

Air mixing within the NZERTF also varied between Session 1 (more mixing) and Session 2 (less mixing). Due to the lower temperatures during Session 1, the heat pump system operated for a longer period of time and resulting in more mixing during Session 1 than Session 2. In addition, during Session 1 a fan was placed at the top of the stairwell between the basement and 1st floor to enhance the mixing of the tracer. During Session 2 the door to the basement was closed. The decrease in mixing in Session 2 led to a greater difference between the measured 1st and 2nd floor concentrations and the basement concentrations (especially on 12/12/15) (Figure 1, right) compared with Session 1 (Figure 1, left).

The measured formaldehyde concentration on the 2nd floor varied to a greater extent than the measured concentration on the 1st floor and basement (Figure 1). To investigate this variation, the formaldehyde concentration was measured in the middle of the 2nd floor master bedroom continuously for one day (Figure 2). The master bedroom was supplied with outside air via the HRV for 40 minutes of every hour (black line Figure 2). Every time the HRV was off the formaldehyde concentration in the room increased 1 ppb$_v$ (1.2 μg/m^3) in roughly 15 minutes. The formaldehyde concentration decreased by a similar amount during the 45 minutes the HRV was supplying outdoor air to the room.

Formaldehyde emissions rates in laboratory settings have been shown to be temperature and relative humidity dependent (Liang et al. 2015). Preliminary observations show that the master bedroom formaldehyde concentration correlated more strongly with the outdoor relatively humidity values (Figure 2), rather than the indoor relatively humidity. No trends were observed between the formaldehyde concentration, ozone concentration, indoor temperature and outdoor temperature. These dependencies will be the subject of future studies at the NZERTF. The collected master bedroom data does show that during the winter, the impact of a 12 % change in outdoor relatively humidity at the NZERTF had a lesser effect on formaldehyde concentrations than turning off the HRV.

The importance of ventilation in high performance buildings is underscored by the fact that (i) a lower air change rate in the NZERTF attic led to elevated formaldehyde concentrations in the attic and (ii) the formaldehyde concentration and the HRV operation in the master bedroom were correlated.
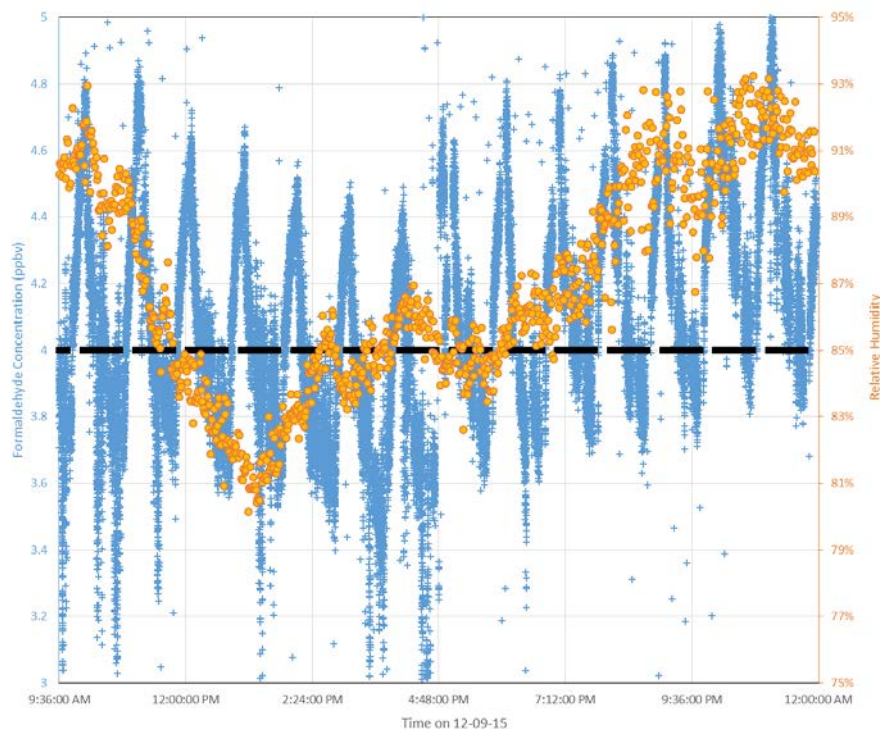
Figure 2. Real-time formaldehyde concentration (ppb_v) data from master bedroom. The HRV was on and providing outdoor air to the master bedroom for 40 minutes of every hour, as indicated by solid portion of the black line. The gaps in the black line show when the HRV was off. Blue data points are the formaldehyde concentration (ppb_v) and orange data points are the outdoor relative humidity (%).

The model achieved a reasonable, although not exact, agreement with the measured data (Figure 1) taken at the NZERTF. While measurements were made in winter, the model was run for a full year, using 2015 weather data, to assess peak formaldehyde concentrations in the NZERTF (Table 1). During the modelled year, the maximum simulated formaldehyde concentration in the attic was 47 ppb_v (56 $\mu g/m^3$), while the maximum simulated formaldehyde concentration in the living space was 9.5 ppb_v (11.7 $\mu g/m^3$). The average predicted formaldehyde concentrations in the summer months (June, July and August) were 29 ppb_v (± 5.8 ppb_v, 36 $\mu g/m^3$ ± 7.1 $\mu g/m^3$) in the attic and 9.5 ppb_v (± 0.6 ppb_v, 12 $\mu g/m^3$ ± 0.7 $\mu g/m^3$) on the 1st floor.

Table 1. Predicted Formaldehyde Concentrations in the NZERTF over a year of operation (ppb$_v$). Average summer values are from June, July and August.

Zone	Maximum	Summer Average	Yearly Average	Yearly Standard Deviation
Attic	47.1	28.0	18.8	6.2
Second Floor	8.7	6.8	5.9	0.8
First Floor	9.5	7.8	7.3	1.0
Basement	8.8	7.4	6.3	1.0

4 CONCLUSIONS

The purpose of the NZERTF is to demonstrate that a typical home can achieve net-zero energy operation while maintaining acceptable indoor environmental conditions. Key design elements of the NZERTF include thermal envelope construction with minimal air leakage and the provision of controlled mechanical ventilation. This research shows that even though the NZERTF meets the ventilation requirements in ASHRAE Standard 62.2 and uses building products with low emission rates, formaldehyde concentrations were elevated during times when the ventilation (HRV) is off and in zones with minimal ventilation air distribution (attic). As new construction seeks to employ these same design principles, specifically low envelope infiltration rates as well as effective and reliable mechanical ventilation, including both adequate outdoor air intake rates and good air distribution are critical for controlling indoor pollutants.

While time-averaged sampling techniques are appropriate for evaluating potential chronic health impacts and have a cost advantage, real-time measurements of formaldehyde concentrations provide new insights to the indoor environment. Real time monitoring of formaldehyde proved to be beneficial for investigating coupling among zones, short term variations in concentrations attributable to mechanical system operation, and associations of concentrations with physical parameters. In addition, the frequent and high-accuracy measurements throughout the NZERTF allowed for verification of a coupled CONTAM and TRNSYS model.

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