INVESTIGATION OF VOLATILE ORGANIC COMPOUNDS EMISSIONS IN DIFFERENT STAGES OF HVAC SYSTEM

by
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Abstract

Efficient heating, ventilating, and air-conditioning (HVAC) systems are essential for improving indoor air quality and human health. However, HVAC systems those days typically rely on mechanical ventilation and filters to reduce air pollution. Volatile organic compound (VOC), a common type of gas phase air pollution, is usually neglected but is considerably higher indoors than outdoors and can cause severe health problems. This research explored how the VOC concentrations changed in different steps of the HVAC system and investigated the relation between VOCs and the HVAC system, building, and occupants. This research aimed to 1) identify and quantify health-related VOC concentrations within the building and compare them with current guidelines; 2) compare VOC concentrations in separate steps of the HVAC system; 3) explore temporal tendencies and source apportionment of VOCs. VOCs were monitored and quantified using a proton transfer reaction time-of-flight mass spectrometer (PTR-TOF-MS) with additional measurements of CO, CO_2, CH_4, NH_3, and CH_2O using cavity ringdown spectroscopy instrumentation from Picarro through a four-valve switching chamber. This process allowed us to sample multipoint pollutants in mixed air, post-filter air, supply air, and return air in a frequent switching sequence. Our observations show that long-term exposure to some health-related VOCs in the sampled building, such as benzene, formaldehyde, and trimethylbenzene, can potentially cause health problems. The average VOC concentrations were highest in the return air and lowest in the mixed air for most indoor source VOCs. The unexpected VOC concentration increase in supply air suggested a leak in the HVAC system. This study will fill fundamental knowledge gaps in the HVAC system and offer quantitative evidence of how HVAC systems influence chemistry in indoor environments by heating, cooling, and filtering the air. With an improved understanding of the association among indoor VOCs, HVAC systems,

buildings, and occupants, engineers can design more effective ventilation systems for buildings that minimize indoor VOCs and reduce the risk of health concerns for occupants.

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Table of Contents

List of Tables

List of Figures

1. Introduction

On average, Americans spend 87% of their time indoors, including 68.7% of their time in residences, 5.4% in offices and factories, 1.8% in bars and restaurants, and 11% in other indoor locations (Klepeis et al., 2001). As the buildings become more energy-intensive and highly occupied, people begin to pay more attention to the health impacts of indoor environmental pollutants. Volatile organic compound (VOC), a group of gas-phase chemicals with high vapor pressure, is one of the major indoor air pollutants. Some VOCs' inhalation and dermal exposure can be related to many acute and chronic health effects. Some common acute symptoms of VOC exposure are eye, nose, and throat irritation (Ernstgård et al., 2007), headaches, nausea/vomiting, dizziness, and worsening asthma symptoms (US EPA, 2014). There is also evidence indicating linkages between mixtures of multiple VOC exposures from indoor sources and the “Sick Building Syndrome (SBS)” (Brinke et al., 1998). Many VOCs have been shown to be animal carcinogens and potential human carcinogens in a high concentration environment (Saalberg and Wolff, 2016).

Formaldehyde is one of the most concerning VOCs. Formaldehyde is classified as a human carcinogen by the IARC (IARC, 1987). Acute, 8-hour, and chronic exposure to formaldehyde can associate with adverse health effects from sensorial irritation to myeloid leukemia and nasopharynx cancers, even with low concentrations (EPA, n.d.). Benzene, another known human carcinogen, is another health-threatening VOC widely present in manufacturing products (Arnold et al., 2013; Edwards and Jantunen, 2001; Harrison et al., 2010; IARC, 1987). In many epidemiological and laboratory studies, the inhalation of benzene has been linked to cancer development (ATSDR, 2007; Health Effects Institute, 2007). There are significant

associations between benzene exposure and acute and chronic myeloid leukemia, nonlymphocytic leukemia, non-Hodgkin lymphoma, multiple myeloma, and other cancers.

Although the toxicity and risk assessment of VOCs have been published for years in the academic field, government regulations are limited. The Clean Air Act requires the EPA to regulate industrial facilities' hazardous air pollutants (HAPs). However, there are no enforceable VOC standards at the federal level for the non-industrial indoor environment (US EPA, 2014). At the state level, California has taken the lead in establishing VOC limits for various products and their associated emissions. California's Air Resources Board (CARB) has conducted the consumer products regulatory program and mandated limits on VOCs, toxic air contaminants (TACs), and greenhouse gases (GHGs) for a variety of consumer products, including adhesives, personal care products, paint, and insecticides, to reduce air pollution and public exposure to the hazards (California Air Resources Board, n.d.).

Indoor VOCs sources are mainly dependent on buildings and occupants. Building materials typically come in composite forms and could be wet or dry. Dry materials with homogeneous diffusivities, such as wood, vinyl, and cloth, have relatively low internal VOC concentrations. Wet material (such as glue, paint, and adhesive) VOC emissions have a high initial emission rate and quick decline, followed by a low emission rate and gradual decay (Haghighat and Huang, 2003; Han et al., 2012). Off-gassing from those construction materials in confined spaces with high emissions and perhaps limited air exchange, VOCs were frequently observed at higher levels, and the emissions are usually consistent and long-lasting (Yu and Crump, 1998). Common VOCs emitted from building materials include but are not limited to formaldehyde, benzene, toluene, and xylene. Like formaldehyde and benzene mentioned earlier, toluene and xylene also have their own health concerns. Long-term exposure to toluene may

cause fatigue, delayed responsiveness, difficulty sleeping, limb numbness, miscarriage, and the loss of female fertility. Exposure to xylene can cause breathing difficulty and impaired lung function and the nervous system (Dehghani et al., 2018).

VOCs can be emitted from body effluents and human skin oil oxidation, including isoprene, methanol, acetone, acetic acid, 4-oxopentanal (4-OPA), and 6-methyl-5-hepten-2-one (6-MHO). Human emissions can account for more than half of the total VOCs, even in a well-ventilated indoor setting (Tang et al., 2016). Another human-related VOC source is personal care products, such as antiperspirants, cosmetics, and hair care products. Cyclic volatile methylsiloxanes (cVMS) are manufactured chemicals that are commonly used in those products. A study at the University of California observed sharp peaks in cVMS concentrations at the beginning of each class and then decayed toward the background level over time (Tang et al., 2015). Housekeeping/cleaning products also have an essential role in indoor VOC emissions. When cleaning agents and air fresheners are used in buildings, residents are exposed to a wide range of airborne chemicals, such as carbonyls, hydrocarbons, and glycol ethers. Cleaning product ingredients' interactions with indoor surfaces significantly affect time-dependent concentration and hence exposures (Nazaroff and Weschler, 2004).

In general, the indoor VOC level was usually considerably higher than those observed outdoors (Pekey and Arslanbaş, 2008), but VOCs can be transported from outdoor to indoors through ventilation or human. Because the physical nature of the outdoor environment is distinct from the indoors, the outdoor VOCs are also markedly different from the indoors. Although the building structure functions as a transport barrier that limits airflow to and from the outdoors, the buildings' ventilation system and air infiltration through leaky walls and windows allow air to exchange between outdoor and indoors. Trimethylbenzene is commonly found outdoors because

it naturally exists in coal and petroleum (Liu et al., 2008). Health effects of trimethylbenzene exposure include skin, eye, and lung irritation, dizziness, headaches, and impaired blood clotting abilities (Gralewicz and Wiaderna, 2001). VOCs can also be carried by humans and transported indoors through activities, such as third-hand smoke (THS). THS is the smoke that can stay on indoor surfaces, such as carpets, clothing, and hair, and continue to smell smoke long after a cigarette is put out. Styrene is a synthetic chemical widely utilized in tobacco, polymers, and rubber production and is classified as "possibly carcinogenic to humans" by the IARC. The neurological system and kidneys are most commonly affected by styrene exposure (IARC, 2019).

Regarding the health concerns related to VOC emissions, there are four principles for reaching good indoor air quality, “minimize indoor emission,” “keep it dry,” “ventilate well,” and “protect against outdoor pollution (Nazaroff, 2013).” The last three principles all can be accomplished via a sound HVAC system, which can maintain a stable temperature and humidity, timely remove the VOC inside the building and reduce the risk of indoor health concerns for occupants. A sound HVAC system introduces purified outdoor air into the indoor environment, diluting indoor VOC concentrations. Specific adsorbents, such as activated carbon filters and desiccant wheels, can also sorb VOCs to remove them effectively (Zhang et al., 2011).

However, high-efficiency particulate air (HEPA) filters, which can capture a wide range of particles by physical separation, are most common in buildings but have some to no ability to reduce VOCs indoors (Batterman et al., 2005; Polidori et al., 2013). Even for the buildings that use activated carbon filters, the process of VOC Sorption can also generate some by-products, such as particles, and are very sensitive to humidity (Fang et al., 2008; Haghighat et al., 2008). Moreover, sometimes HVAC systems can be emission sources because particles or VOCs

captured on the filter surface may be re-emitted or undergo chemical reactions, leading to downstream emissions (Metts and Batterman, 2007; Schleibinger and Rüden, 1999).

This research explored how the VOC concentrations changed in different steps of the HVAC system and investigated the relation between VOCs and the HVAC system, building, and occupants. This research aimed to 1) identify and quantify health-related VOC concentrations inside the building and compare them with existing guidelines; 2) compare VOC concentrations in different stages of the HVAC system; 3) explore temporal trends and source apportionment of VOCs. By exploring the VOC characteristics distinctions among the HVAC system, more effective techniques and strategies can be developed to reduce VOC exposures.

2. Materials and Methods

Measurements were taken in one of the air handling units (AHU-3) located on the penthouse of Malone Hall from November 5th to December 1st, 2021. The building was a normally functioning academic building at the Homewood Campus of Johns Hopkins University in Baltimore, MD. The AHU-3 supplies air for the ground floor (15,884 sqft) which has labs, large offices with computers and printers, small restrooms, a seminar room, classrooms, janitor storeroom and a food corner with a sink, a fridge, three coffee machines, and one microwave. All windows of the offices were closed 24 hours a day. Due to the relatively new age of the building, none of the rooms had any current refurbishing or renovation projects during the air sampling period. No smoking in the buildings occurred due to campus ban on smoking and electronic cigarettes.

There are four major parts of the AHU-3 (Fig. 1), return, mixed, post-filter, and supply air. First, the outdoor air fan draws the air in, and the outdoor air mixes with return air. The

minimum outdoor to indoor airflow rate is 39.8%. Then the mixture goes through a minimum efficiency rating value (MERV) 10 pre-filter and a MERV 15 final filter to filter airborne particulates, including pollen, fungal spores, ground dust, etc. No activated carbon filter is used in this HVAC system, so VOCs are only removed from the indoor atmosphere by deposition or dilution with outdoor air. The system operates the heating and/or cooling coil once the outdoor temperature reaches a certain point (season-dependent). After heating/cooling, two pumps in the supply air channel pump the air out and distribute to rooms via ductwork. Part of the room air exhausts outdoors, and the other part is recycled and used as return air to save energy.

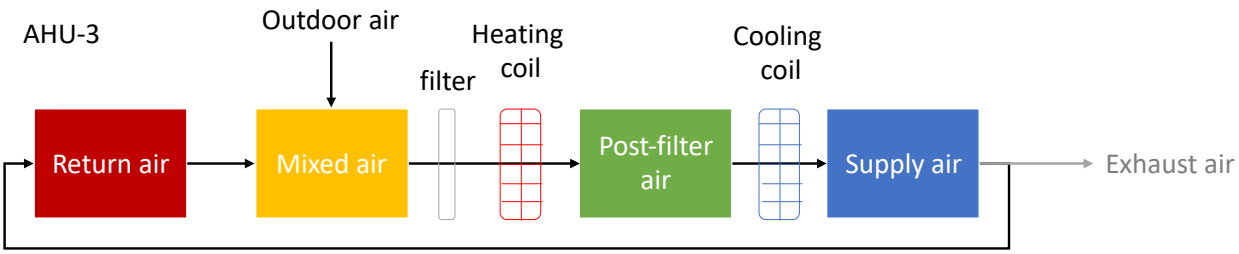

Figure 1. Simplified schematic of the AHU-3 in Malone Hall

The sampling conductive PFA tubes were placed in the middle of each the HVAC stages using speaker stands. The other end of the sampling tubes were attached to a custom four channel valve switcher accordingly (Fig. 2). Channel 1 was return air, channel 2 was mixed air, channel 3 is post filter air, channel 4 is supply air. The four-channel valve switcher is a selector valve that selects one of four air sample lines and delivers that line to analytical instruments while maintaining full flow, using mass flow controllers (MFCs) through the remaining three unselected lines. Each channel was sampled for 7 minutes and then switched to the next channel. This research focused on monitoring VOCs, but it belongs to a larger project where particles were also monitored.

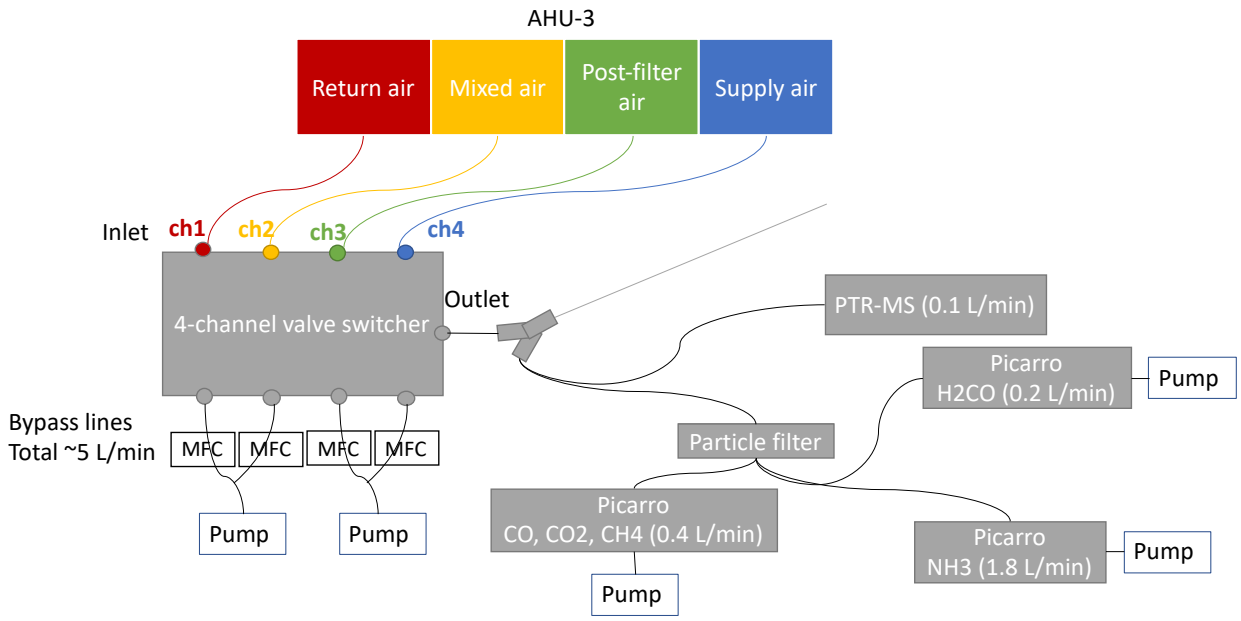

Figure 2. Simplified experiment layout

A PTR-TOF-MS (Vocus PTR-ToF Elf, Tofwerk Switzerland) was used to monitor concentrations of different VOCs. The PTR-TOF-MS uses H_3O^+ as the primary reagent ion with a mass range of 30-500 mass-to-charge ratio (m/z) to track the VOCs. The sensitivity of this Vocus PTR-TOF Elf is 500 cps/ppb, LOD is less than 20 ppt per minute and the maximum resolving power is 750 M/ΔM (TOFWERK, 2019). The detailed description and application of PTR-TOF-MS as an analytical method can be found elsewhere (Gouw and Warneke, 2007). PTRMS took samples every 5 seconds. Calibration of the instrument used the 5 ppb compounds (Table 1) in a gas cylinder. The process of the calibration follows Krechmer's study on PTRMS (Krechmer et al., 2018). The Vocus ZeroAir (Serial No. VZA-030) was used to generate zero air by catalyzed combustion of VOCs into CO_2. To ensure the instrument's sensitivity is stable and the data are reliable, calibration gas was introduced every six hours and zero air was sampled every hour. Intensity (ions/s) was converted to concentration by dividing by the slope of the linear regression.

Table 1. 13-component calibration mixture in nitrogen

Standard Compound	CAS#	Concentration (ppb)
Methanol	67-56-1	5
Acetonitrile	75-05-8	5
Acetaldehyde	75-07-0	5
Acrylonitrile	107-13-1	5
Acetone	67-64-1	5
Isoprene	78-79-5	5
Methyl Vinyl Ketone	78-94-4	5
Methyl Ethyl Ketone	78-93-3	5
Benzene	71-43-2	5
Toluene	108-88-3	5
m-Xylenes	108-38-3	5
1,3,5-Trimethylbenzene	108-67-8	5
a-Pinene	80-56-8	5

The Picarro G2401 gas concentration analyzer (Picarro Inc., USA) was used to measure carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4) and water (H_2O) vapor with a sensitivity of ppb. It also corrected for the humidity and provided dry gas mole fractions for CO_2, CH_4 and CO. The CO_2 concentration are related to the number of occupants. The Picarro G2307 gas concentration analyzer (Picarro Inc., USA) was used to monitor the concentration of formaldehyde (H_2CO) in ppb along with CH_4 and H_2O. Both of the Picarro instruments took samples every second.

The PTR-MS data were analyzed using Tofware, and Picarro data were analyzed using Igor 8. Calibration and zero air data were taken out from the time series, and the rest were separated into different channels. Hourly averages were calculated for time series analysis to minimize the noise. High-resolution data (5-second data for PTRMS, 1-second data for Picarro) were employed for diurnal and weekly trend analysis to maximize the accuracy.

The observed VOC concentration were compared to existing official guidelines. The reference concentration (RfC) inhalation toxicity released from the Integrated Risk Information System (IRIS) of the US EPA indicates an estimate (with magnitude uncertainty) of continuous inhalation exposure to the human population over a lifetime that is unlikely to pose a

noticeable risk of adverse effects. The RfC is usually used in EPA's noncancer health assessments and can be calculated through no-observed-adverse-effect level (NOAEL), lowest-observed-adverse-effect level (LOAEL), or benchmark concentration divided by uncertainty factors (UF) (US EPA, 2002). Inhalation unit risk (IUR) refers to the upper-bound excess lifetime cancer risk from continuous exposure to an agent at a concentration of 1 μg/m3 in air (US EPA, 2005). The chronic reference exposure level (chREL) that was published by the California Office of Environmental Health Hazard Assessment (OEHHA) implies there will be no adverse noncancer health effects if someone exposed to that concentration or below continuously for up to a lifetime (OEHHA, 2015). We hourly averaged concentrations for VOCs in different channels during the whole experiment and compared 600-hr average concentrations in return air, which circulates from the ground floor, to the chronic RfC and chREL to evaluate the long-term health relevances of VOCs. After we hourly averaged each identified VOC in return air, mixed air, post-filter air, and supply air, we tested correlations on that hourly averaged VOC and CO_2 concentrations in the return air. The correlation coefficients were used to test for linear relationships between VOCs and CO_2, which was an indicator for occupancy.

3.Result and Discussion

3.1 HVAC system operation

The concentration changes among different stages of the HVAC system can inform the HVAC operation procedure and potential flaws. For CO_2 (Fig. 3) and most VOCs in a sound HVAC system, with a few exceptions of VOCs from outdoor sources, the return air was expected to have the highest average concentration, followed by mixed air, post-filter air, and supply air. However, based on the non-holiday measurements, supply air generally had the

second-highest average concentration for CO_2 and most VOCs. It was higher than the ones from post-filter air and mixed air. The pumps in the supply air channel draw the processed air into distribution duct and kept the pressure low. The elevated levels in supply air stages may have indicated a small leak in the supply air channel that draws return air or air from the penthouse (where the HVAC system is placed) into the supply air.

Thanksgiving break (5 pm, 11/24/2021 to12 pm, 11/26/2021) was an exception to this trend. During this time, the chemical concentrations from the HVAC system's four phases were identical and generally higher than average VOC concentrations on non-Thanksgiving-break period. This unique pattern implied the HVAC system was not fully operating during the Thanksgiving break. Although the specific operation time of this shutting down was not recorded by the system, the mechanical engineer of the building confirmed they switched off the HVAC system as their holiday protocol. For the universality of the result, Thanksgiving week (11/21/2021 to 11/27/2021) was not included in all the daily and weekly trend analyses.

Monitoring CO_2 concentrations and their changes over time in indoor environments appear to be a reliable way of calculating the relative changes in occupants (Stönner et al., 2018). However, the observed CO_2 concentration during the sampling time (Fig. 3), along with most indoor source VOCs, gathered continually throughout the day and commonly peaked at night. This trend suggested that the CO_2 concentration measurement was not an accurate indication for determining indoor spaces' occupancy in this experiment. It was also largely affected by the

economizer mode of the HVAC system operation, which limits the outdoor ambient air to the system at night for energy efficiency purposes.

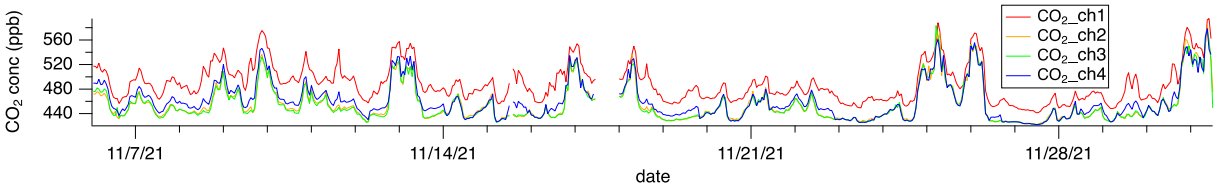

Figure 3. The hourly average CO_2 concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) from Nov 6th to Dec 1st, 2021

3.2 Health-related VOCs

Assessing VOC exposure and related health outcomes for the general population in non-occupational conditions is difficult because individual activities are extremely varied, ambient weather conditions are unpredictable, and building settings are highly diversified.

Although the observed maximum concentration of benzene was 3.9 ppb, which was lower than the EPA IRIS reference concentration (RfC), the average concentration was 1.0 ppb, which was higher than the California OEHHA chREL. This suggested the benzene level in this building may lead to potential health effects for occupants over time. Although the maximum concentrations of trimethylbenzene, formaldehyde, styrene, toluene, and xylene were below the EPA IRIS RfC and California OEHHA chREL, the average concentration of trimethylbenzene, formaldehyde were within one or two magnitudes of the CA chREL, and the long-term exposure of those levels was still concerning to the occupants.

The four-channel average formaldehyde concentration in the HVAC system was 1.5 ppb, but the return air had the highest average of 2.0 ppb, and the mixed air had the lowest average of 1.2 ppb. This confirmed the emission source of formaldehyde was mostly indoors and by introducing the outdoor air, the concentration of formaldehyde decreased. The highest concentration (excluding Thanksgiving break) was detected in the post-filter and post-heating air, and it was close to the OEHHA chronic REL. This suggested that filter is a potential source that emit

formaldehyde, or the heating process accelerate the formaldehyde off-gassing. The carcinogenic IUR for formaldehyde is 1.3E-5 per μg/m^3, which means the lifetime cancer risk estimation is 1.3E-5 based on inhalation exposure to a concentration of 1 μg/m^3. In here, that means 1.5 excess cancer cases (upper bound estimate) are expected to develop per 100,000 people if exposed daily for a lifetime to 1 μg of formaldehyde per m^3 of air.

Table 2. Summary of observed health-related VOC concentrations and existing guideline values

Chemical Name	California OEHHA chRELs* (ppb)	EPA IRIS RfC** (ppb)	Observed Avg. Conc ± SD (ppb)	Observed Max Conc (ppb)
Benzene	0.9	9.4	1.0 ± 0.4	3.9
Trimethylbenzene		12.2	0.5 ± 0.4	8.7
Formaldehyde	7.3	Carcinogenic IUR*** 1.3E-5 per (μg/m^3)	1.5 ± 0.8	5.3
Styrene	211.3	234.8	0.5 ± 0.2	6.1
Toluene	111.4	1326.8	0.5 ± 0.3	8.6
Xylenes	161.2	23.0	0.5 ± 0.3	3.0

** California Office of Environmental Health Hazard Assessment (OEHHA) chronic reference exposure level*
*** United States Environmental Protection Agency (EPA) Integrated Risk Information System (IRIS) reference concentration*
**** Carcinogenic inhalation unit risk (IUR)*

3.3 Source apportionment

The quality of indoor air varies greatly depending on the sources. In this experiment, in addition to the outdoor source, there were typically three categories of indoor sources, building material, human metabolism emission, and occupant-related activities.

3.3.1 Building material emissions

For common building material VOCs, including formaldehyde (Fig. 4A), benzene (Fig. 5A), toluene, and xylene, the return air had the highest average concentration (excluding Thanksgiving break) during the campaign, while mixed air and post-filter air had the lowest. This indicates an indoor source is important for all of these chemicals. Because benzene, toluene, and xylene have comparable chemical structures, their chemical behaviors are very similar. A

similar daily and weekly tendency (Fig. 4B, AC, Fig. 5B, 5C) were also found in all building material-related VOCs, which suggested homogeneous diffusivity of chemicals. However, it remained unclear why the weekday concentration was higher than the weekends.

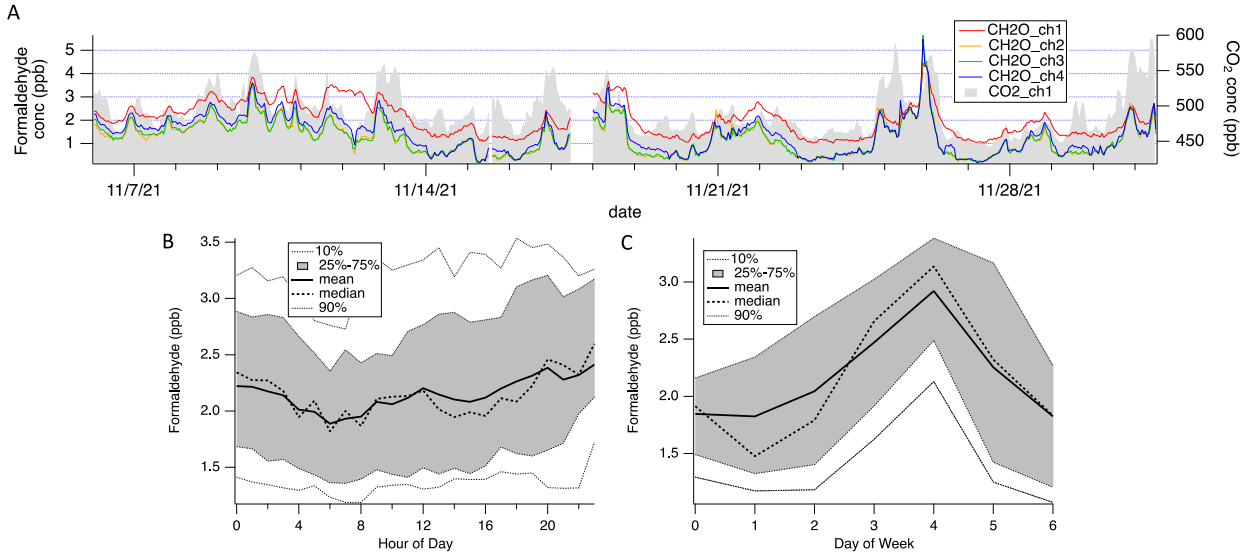

Figure 4. A. The hourly average formaldehyde concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021.

B. Diurnal trend of formaldehyde concentration (ppb) in return air. The analysis excluded Thanksgiving week.

C. weekly trend of formaldehyde concentration (ppb) in return air. The analysis excluded Thanksgiving week.

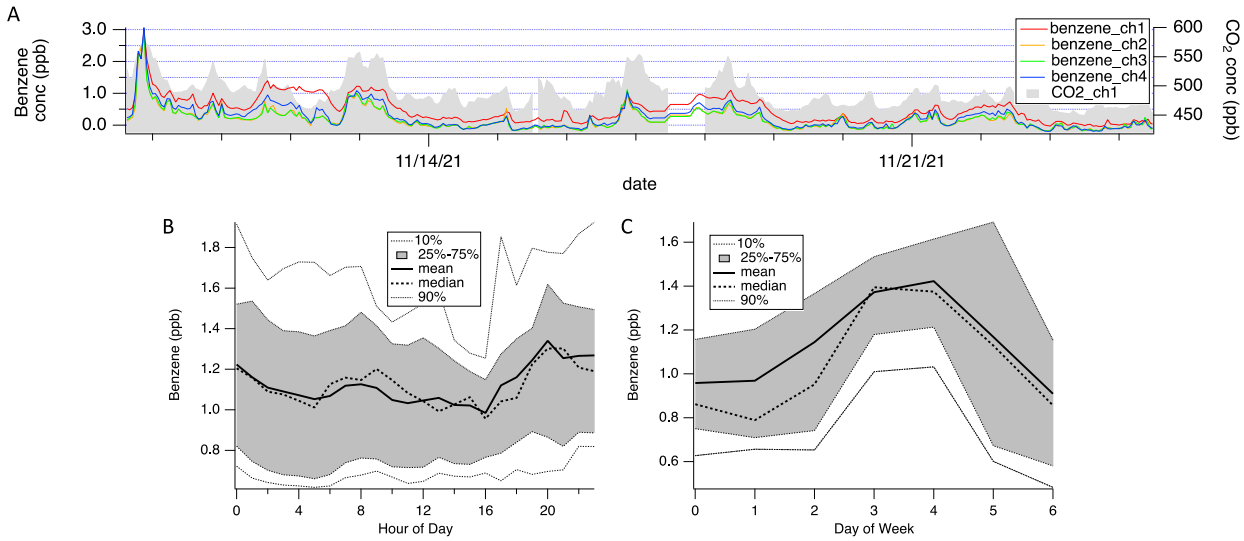

Figure 5. A. The hourly average benzene concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021

B. Diurnal trend of benzene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

C. Weekly trend of benzene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

3.3.2 Human metabolism emissions

Many studies have demonstrated that human metabolism emissions can be a major indoor VOC source. Humans are the source of several VOCs along with CO_2 via exhaled breath, saliva, sweat, skin secretions, and other biological excretions (Drabińska et al., 2021).

Isoprene is one of the most recognized VOCs in breath since it is a by-product of the mevalonate pathway. Body movement and exertion can substantially enhance isoprene concentration in exhaled breath (Mitova et al., 2020). Isoprene and CO_2, another prominent component of exhaled breath, have a correlation coefficient of 0.72, which indicate a significant and positive relationship between the two. Isoprene (Fig. 6A) was highest in the return air and lowest in the mixed air. The concentration had started to rise at 6 am and reached its peak around 8 pm (Fig. 6B). This suggested isoprene concentration was correlated with occupants and continuously accumulated through the day. The weekday concentration (Fig. 6C) was higher than the weekend, which further indicated the positive relationship between isoprene and occupancy.

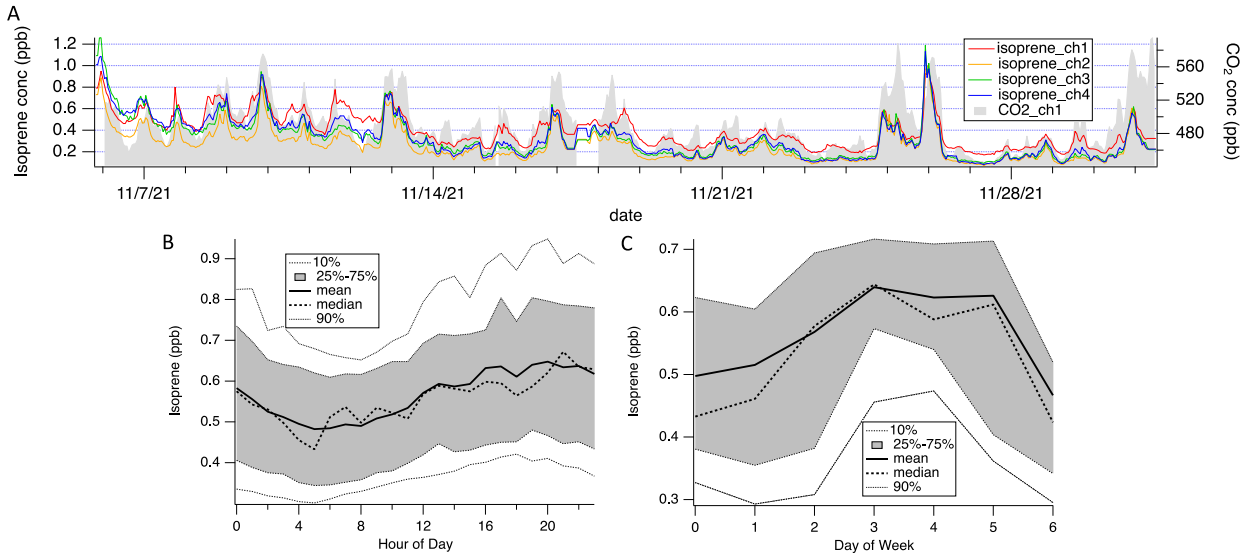

Figure 6. A. The hourly average isoprene concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021
B. Diurnal trend of isoprene concentration (ppb) in return air. The analysis excluded Thanksgiving week.
C. Weekly trend of isoprene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

Another paradigmatic example of human metabolic VOCs is acetaldehyde. It is endogenous product that can be initiated in ethanol metabolism (Drabińska et al., 2021). Like isoprene, acetaldehyde also had the highest concentration in the return air and lowest in the mixed air (Fig. 7A). It had multiple spikes from 8 am to 4 pm (Fig. 7B), and the weekday concentration was more elevated than on weekends (Fig. 7C). This weekly trend insinuated a good agreement between acetaldehyde and human emission, and the diurnal trend indicated a discontinuous human source, which can be generated from activities such as students attending classes.

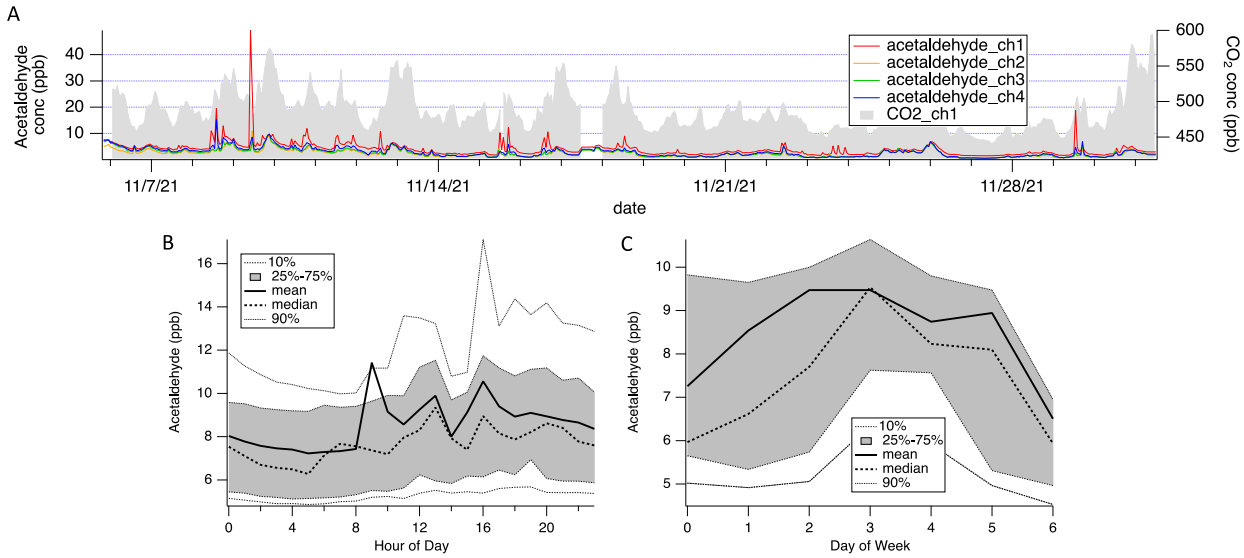

Figure 7. A. The hourly average acetaldehyde concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021

B. Diurnal trend of acetaldehyde concentration (ppb) in return air. The analysis excluded Thanksgiving week.

C. Weekly trend of acetaldehyde concentration (ppb) in return air. The analysis excluded Thanksgiving week.

3.3.3 Occupant-related activities

Occupant-related activity is another distinct bracket of indoor VOC sources with considerable variability. This might be attributed to numerous resident activities, from work (lab work, office supplies, etc.), personal activities (application of cosmetic products, heating food, dry-clean clothes, etc.) to cleaning activities (Ongwandee et al., 2011; Steinemann, 2015).

Mainly two different activities stood out in this experiment. Based on the chemical identification and behavior, we categorized them as "short term spikes" that may affiliate with air freshener spray (Ibrahim ALshaer et al., 2019) and "long lasting peaks" associated with cleaning.

The phenol group chemicals (including phenol, methylphenol, 4-ethylphenol) as well as 2-hydroxy-3-methyl-2-cyclopenten-1-one and benzyl alcohol had a very unique tend. Phenol (Fig. 8A), 4-ethylphenol, 2-hydroxy-3-methyl-2-cyclopenten-1-one had three outstanding peaks at Monday 2 pm (11/8/2021), 5 pm (11/15/2021), 3 pm (11/22/2021). Methylphenol (Fig. 9) and benzyl alcohol had an additional distinguished peak at Sunday (11/7/2021) 6 pm. The concentration in return air was not always the highest for phenol group chemicals. For example, the phenol concentration on the first two Monday spikes was highest in return air, but the last Monday spikes on 11/22/2021 were highest in post-filter air. While the spike appear in different locations of the HVAC this may be a sampling artifact due to the short duration of the source and the timing in relation to where in the HVAC the instruments were sampling at the time of the emission since there is 7 minutes per sampling site. The sharp increase of the phenol concentration hinted that sources were likely sudden surges, such as air freshener sprays. Monday and Fridays had the highest phenol group concentration (Fig. 8C) suggested personal habits of occupants.

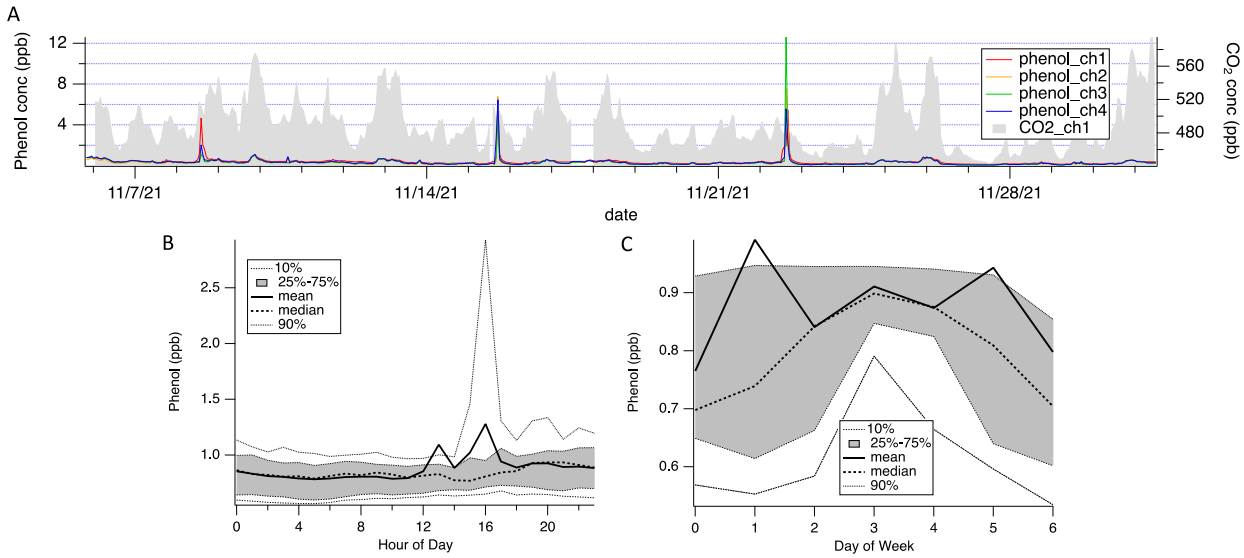

Figure 8. A. The hourly average phenol concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021
B. Diurnal trend of phenol concentration (ppb) in return air. The analysis excluded Thanksgiving week.
C. Weekly trend of phenol concentration (ppb) in return air. The analysis excluded Thanksgiving week.

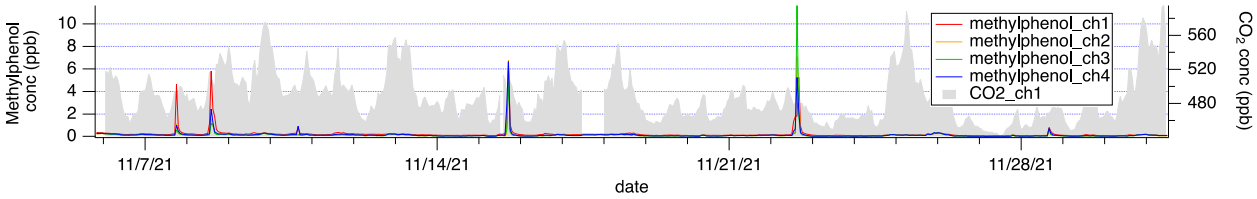

Figure 9. The hourly average methylphenol concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021

Monoterpenes, like alpha-pinene and limonene, are among the most abundant VOCs reported in indoor environments (Gokhale et al., 2008; Król et al., 2014). Although they naturally occur in plant-based materials, such as wooden furniture, they are frequently added as fragrances to various commercial products, such as cleaning agents and hand-wash alcohol gel (Ongwandee et al., 2011). Moreover, monoterpenes can easily react with oxidative agents and form hazardous secondary pollutants such as formaldehyde, formic acid, and acetic acid (Król et al., 2014). Unlike the sharp peaks that were observed in phenols, monoterpenes had dull peaks that lasted hours (Fig. 10A), which suggests chemical release periods were much longer and could be categorized as cleaning events such as cleaning the bathroom. The concentration of alpha-pinene and limonene was generally highest before 6 am, gradually declined over the

daytime, and accumulated again after 6 pm (Fig. 10B). This might be because monoterpenes undergo oxidation reactions. However, there was no noticeable variation in the average concentrations of monoterpenes measured on weekends versus weekdays (Fig. 10C).

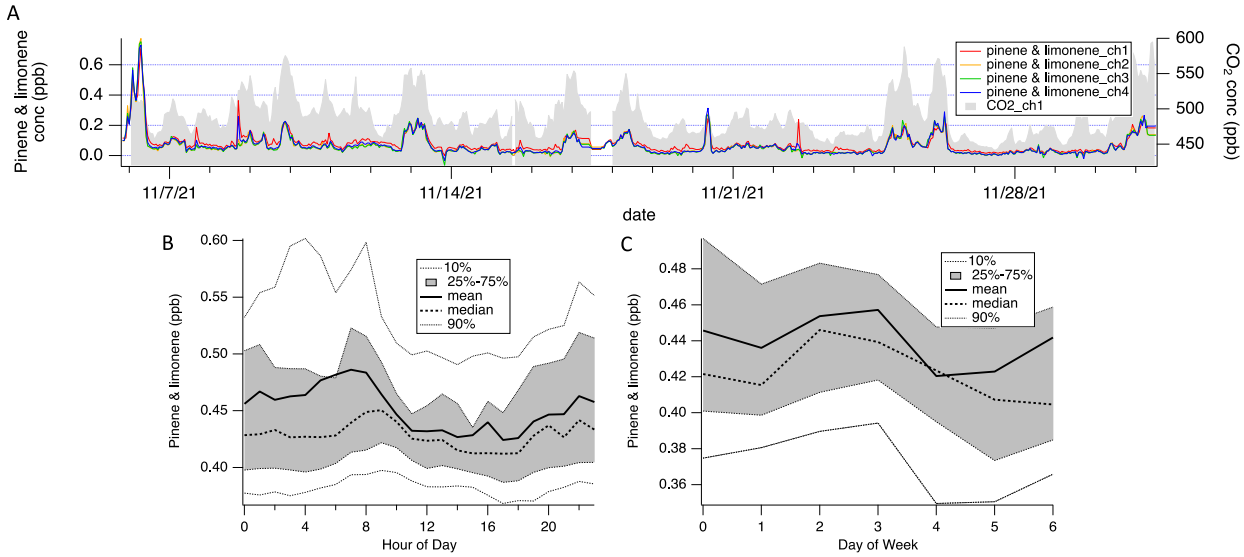

Figure 10. A. The hourly average pinene & limonene concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021

B. Diurnal trend of pinene & limonene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

C. Weekly trend of pinene & limonene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

3.3.4 Outdoor sources

Fossil fuel-related aromatic compounds, such as trimethylbenzene and styrene, are often associated with vehicle emissions and should have much higher concentrations in the outdoor environment than indoors (Jia et al., 2008). However, styrene (Fig. 11A) and trimethylbenzene (Fig. 12A) were observed to have the highest average concentration in post-filter and post-heating air and the lowest average concentration in the mixed air channel. The concentration difference among channels was substantial during the first week of our experiment and continues to decline gradually over time. Both styrene (Fig. 11B) and trimethylbenzene (Fig. 12B) had a relatively steady concentration over the day, with a slight increase in the afternoon around 4 pm.

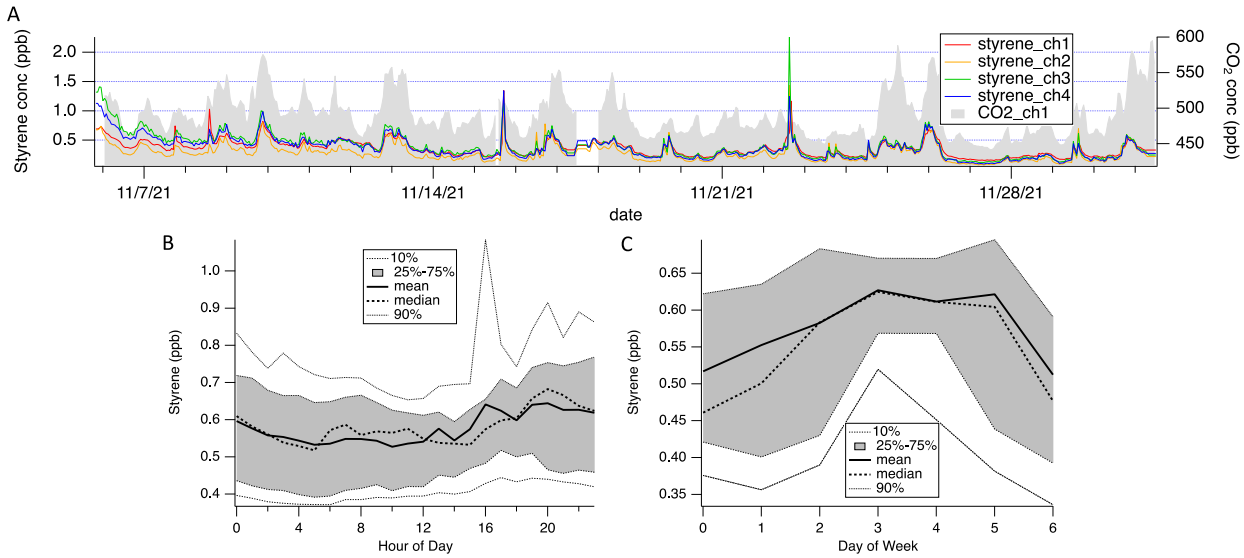

Figure 11. A. The hourly average styrene concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021
B. Diurnal trend of styrene concentration (ppb) in return air. The analysis excluded Thanksgiving week.
C. Weekly trend of styrene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

Trimethylbenzene had more distinctive spikes in the mixed air, particularly on Tuesday and Friday (Fig. 12C), which may have implied outdoor emissions potentially from vehicles or machinery such as lawnmowers or chainsaws since Malone Hall is located beside a campus lawn and the traffic node of campus south gate.

One potential hypothesis is that the pre- and main filters that the HVAC system used contain polystyrene, which can release styrene through the heating process. Another possible explanation is that the filters were stored in in a maintenance area that also contained fuel or gasoline-powered equipment was placed and adsorbed fossil fuel-related VOCs before being used. Once the filters had been put into the HVAC system, those VOCs were slowly off-gassed. Further experiments and investigation of these filters is needed to identify the sources of these aromatic chemicals. Additional experiments with filter material directly are needed to verify the

potential of styrene emission.

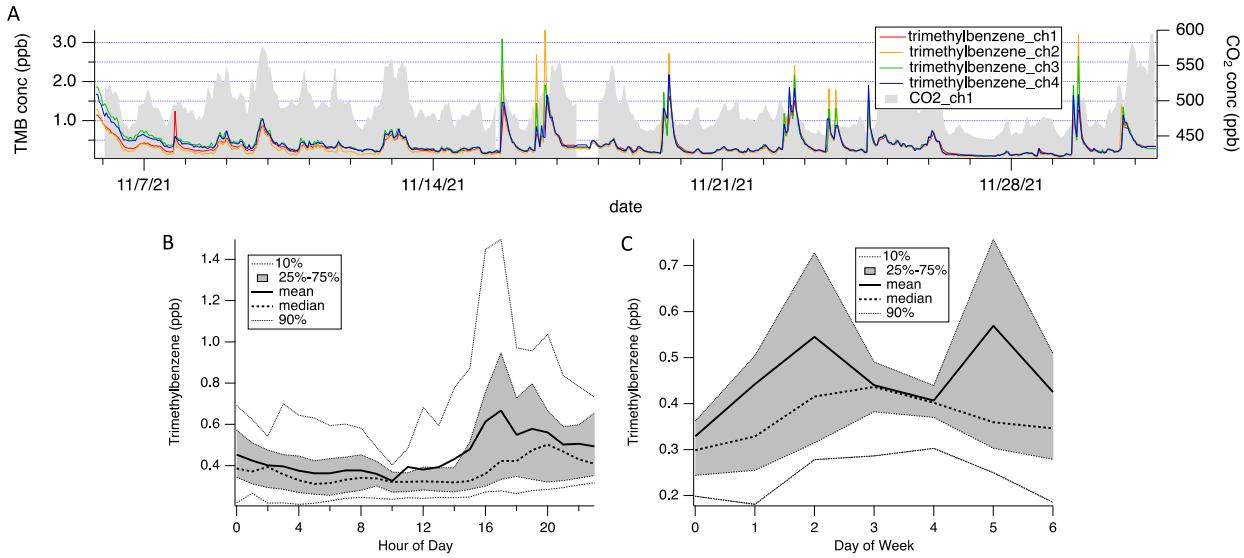

Figure 12. A. The hourly average trimethylbenzene (TMB) concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021

B. Diurnal trend of trimethylbenzene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

C. Weekly trend of trimethylbenzene concentration (ppb) in return air. The analysis excluded Thanksgiving week.

3.3.5 Combination sources

However, many chemicals are not from single sources but rather a combination of them. Acetone, for instance, is frequently detected in human breath due to biological processes (Stönner et al., 2018). Still, it is widely used as a solvent for paints, plastics, adhesives, nail polish, and cleaning agents (Weisel et al., 2008). Although the acetone concentration didn't follow the same trend as CO_2 or other singular categorized sources, it started to creep up after 8 am and declined around 8 pm (Fig. 13B). In addition, it typically peaked on Mondays, Thursdays, and Fridays (Fig. 13C). This suggested acetone was highly related to occupancy-related sources. However, acetone had multiple noteworthy peaks at different stages of the HVAC system, which signifies that it was hard to distinguish the sole source of acetone.

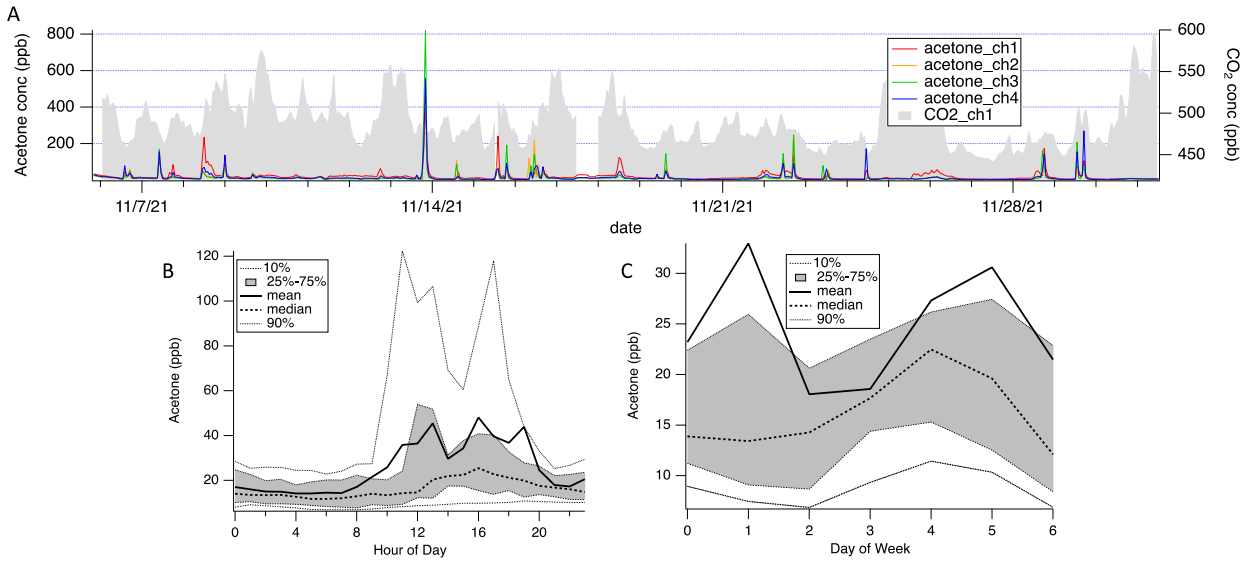

Figure 13. A. The hourly average acetone concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021
B. Diurnal trend of acetone concentration (ppb) in return air. The analysis excluded Thanksgiving week.
C. Weekly trend of acetone concentration (ppb) in return air. The analysis excluded Thanksgiving week.

Methyl ethyl ketone (MEK) is another example of chemicals from a mix of origins, but unlike acetone, the source combination for MEK was more clear. MEK is a highly volatile chemical that is frequently used as a commercial cleaner and solvent for glues, paints, coatings, and printing inks (Gad, 2005). MEK followed the CO_2 concentration for the most part throughout the experiment, with flat diurnal and weekly variations (Fig. 14B, 14C) like a typical construction material source. On the other hand, it had a unique peak (Fig. 14A) at 6 pm on Nov 7, 2021, which was identified as an air freshener spray event that was also associated with both methylphenol and benzyl alcohol.

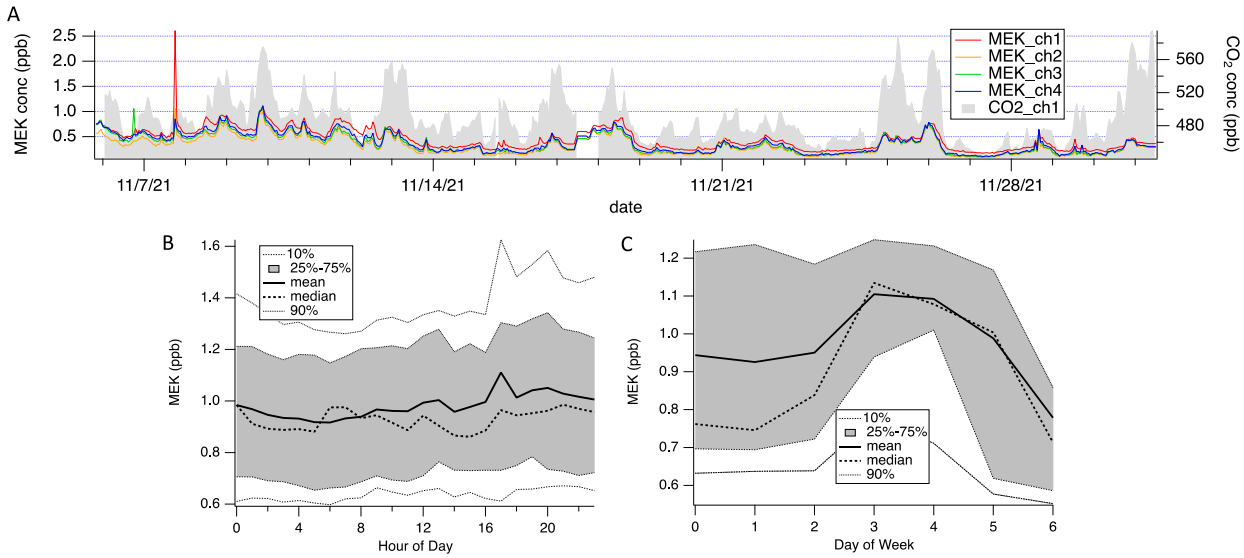

Figure 14. A. The hourly average methyl ethyl ketone (MEK) concentration (ppb) in return air (ch1), mixed air (ch2), post-filter air (ch3), and supply air (ch4) with a background of CO_2 concentration in ch1 from Nov 6th to Dec 1st, 2021

B. Diurnal trend of MEK concentration (ppb) in return air. The analysis excluded Thanksgiving week.

C. Weekly trend of MEK concentration (ppb) in return air. The analysis excluded Thanksgiving week.

3.4 Limitations

One major limitation of this study is the lack of HVAC sequence of operation data. The operation sequence is the series of steps for how the HVAC system responds to the outside temperature and its settings of operation for Malone Hall. It can provide useful information that can be used to improve VOC source apportionment, such as the HVAC economizer operation schedule, indoor/outdoor air ratio, recycle rate of the return air, temperature, humidity, etc.

Another limitation is data loss due to instrument downtime. PTR-MS and three different Picarros continuously monitored VOC data throughout the campaign. However, PTR-MS was lost data from 11 am Nov 17 to 12 am Nov 18, 2021. This loss of data is a common occurrence, but if happens frequently can affect VOCs' diurnal and weekly analysis or even potential spike events that may appear.

In addition, our PTR-MS is not a higher resolution instrument so chemicals with similar mass-to-charge ratios cannot be completely resolved in the mass spectrum leading to some ambiguity. For example, ethanol (m/z 47.049) and formic acid (m/z 47.013) cannot be

distinguished in the mass spectrum. Similarly, the reagent ion that binds to the VOC of interest for the PTR-MS is typically H^+, but can also be H_3O^+. This can lead to overlapping signals which can be accounted for, but require additional post-processing. For example, acetic acid with H^+ will be observed at (m/z 61.028) and with H_3O^+ observed at m/z of 79.043 in the mass spectrum. The latter overlaps with the benzene + H^+ signal at (m/z 79.054). Although a softer ionization method, some chemicals may fragment into smaller pieces and generate signals that can overlap with signals of smaller VOCs. For instance, pinene has a parent peak at m/z of 137.132 and a secondary peak at 81.070, which will overlap with other chemicals around m/z 81. To solve this problem, GC can be utilized and attached to the PTR-MS to improve the separation and identification of the analytes.

Moreover, a single VOC substance can be emitted through multiple sources, and one emission origin can contribute to numerous VOCs. Without source apportionment analysis, it is hard to separate or determine the types of emission sources and their fingerprints (Batterman et al., 2014). In the future study, PMF model can be used to calculates source contributions.

4. **Conclusion**

Americans spend most of their time indoors, and indoor VOC levels are usually higher than those observed outdoors with distinct chemical patterns. A well-designed HVAC system is considered to be the answer to improving indoor air quality. However, the indoor and outdoor air exchange is mainly through mechanical ventilation with filters designed to remove particles, not gas-phase pollution. In this study, we used PTR-MS and Picarros to explore the VOC concentrations in different stages of the HVAC system and investigate the trend and sources of VOCs. The maximum concentrations of trimethylbenzene, formaldehyde, styrene, toluene, and

xylene were below the EPA IRIS reference concentration and California OEHHA chronic reference exposure level. Although the observed maximum concentration of benzene was lower than the EPA IRIS reference concentration, the average concentration was higher than the California OEHHA chronic reference exposure level, which may lead to potential health effects. The average VOC concentrations were highest in the return air and lowest in the mixed air for most indoor source VOCs from building material, human biological emissions, and occupant-related activities. VOCs from diverse sources responded differently and displayed distinct temporal patterns in four phases of the HVAC system. The unexpected VOC concentration increase in supply air stages suggested the presence of a leak in the HVAC system that pulls return air into the supply air. This study offered quantitative evidence of how HVAC systems influence chemistry in indoor environments by mixing, heating, cooling, and filtering the air. With a deeper understanding of the relationship between indoor VOCs and HVAC systems, engineers can design more effective ventilation systems for buildings that minimize indoor VOCs and reduce the risk of health concerns for occupants.

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