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Volatile organic compound emissions from a multi-unit residential building to ambient air†

Amirashkan Askari [ID] [a] and Arthur W. H. Chan [ID] *[b]

Emerging sources, such as volatile chemical products (VCPs) and other non-traditional emission categories, are becoming increasingly important in urban air pollution as the contributions of recognized sources such as traffic and industrial emissions decline. Indoor emissions constitute a large fraction of organic gaseous species from these sources, making buildings potential contributors to ambient air pollution. This study illustrates building emissions by presenting findings from a sampling campaign in downtown Toronto, analyzing volatile organic compounds (VOCs) from the mechanical ventilation inlet and exhaust air streams of a multi-unit residential building (110 units). Due to indoor emissions, VOCs were detected more frequently and at higher concentrations (median levels higher by about 22%) in the exhaust stream than in the inlet stream, indicating that the building serves as a net VOC source to the ambient air. VCP-related species were consistently more abundant in the exhaust air, confirming the influence of indoor sources. In particular, median concentrations of volatile methyl siloxanes and monoterpenoids associated with emissions from adhesives, personal care products, and cleaning agents ranged from about 2–5 $\mu g\ m^{-3}$ in the exhaust stream in comparison with 0.2–0.5 $\mu g\ m^{-3}$ within the inlet stream. Source apportionment analysis of VOC concentrations across the exhaust and inlet airstreams revealed indoor emissions of siloxanes, monoterpenoids, and oxygenated VOCs from coatings, cleaners, and personal care products as primary contributors to exhaust stream trends. Net building VOC emissions, defined as the rate of outflowing minus the inflowing VOCs, were calculated from the measured concentrations and ventilation rates. The resulting values aligned with indoor emissions predicted from a published VCP emission inventory for Canada, emphasizing the pivotal impact of VCP indoor sources on urban air quality. Exhaust and inlet stream concentrations of VCP-related species were found to be significantly ($p < 0.05$) correlated, suggesting the building emissions influencing outdoor VOC levels. These results highlight the crucial impact of indoor emissions, especially from VCPs, on ambient air quality and the need for further research into indoor-to-outdoor pollutant transfer mechanisms to address urban air pollution.

Environmental significance

Continuous air pollution mitigation measures during the last few decades have led to an increasing impact from alternative sources, such as volatile chemical products (VCPs). These emerging sources are characterized by a more even distribution of emissive activities across indoor and outdoor spaces. Since indoor emissions are prone to be transported outdoors through ventilation, buildings may play a critical role in ambient air pollution. This study provides experimental evidence that quantitatively aligns with previous insights concerning indoor VCP emissions. By elucidating the key indoor sources driving the observed trends, the results from the present work can inform future efforts aimed at improving urban air quality by controlling indoor-to-outdoor pollutant transport.

[a]*Department of Civil and Mineral Engineering, University of Toronto, Toronto, M5S 1A4, Canada*

[b]*Department of Chemical Engineering and Applied Chemistry, University of Toronto, Toronto, M5S 3E5, Canada. E-mail: arthurwh.chan@utoronto.ca; Tel: +1-416-978-2602*

† Electronic supplementary information (ESI) available: Details on the air sampling procedure, sample analysis, inlet, exhaust, and indoor concentration profiles, downscaling VCP indoor emissions to a single building, net building emission calculations, source apportionment analysis, and statistical significance hypothesis tests (PDF). See DOI: https://doi.org/10.1039/d4em00689e

1 Introduction

Indoor spaces exchange air pollutants with ambient air through ventilation.[1] Indoor air pollution from outdoor sources and its implications regarding residents' well-being have been the subject of prior studies.[2,3] Air pollutant emissions from traffic and industrial sources have decreased recently owing to technological advancements and emission controls.[4] These changes have led to the increasing relative

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importance of alternative sources such as cooking[5–9] and volatile chemical products (VCPs).[10–12] VCPs encompass a wide range of chemical products, including coatings, adhesives, cleaners, and personal care products. These products can be broadly categorized into industrial and consumer-use VCPs. Industrial VCPs are often associated with large-scale manufacturing and construction, making their corresponding emissions to mainly occur outdoors. In contrast, consumer VCPs—especially household, maintenance, and architectural products—are utilized within indoor spaces, contributing to indoor emissions in residential and commercial buildings. Many of these emerging sources feature chemical and volatility profiles that have distinctive impacts on air quality, especially through forming secondary air pollutants such as ozone and secondary organic aerosol (SOA).[13,14] For example, the reactivities and SOA yields from oxygenated species found in VCPs are different from those of alkanes and aromatic species dominating traffic emissions, leading to alternative atmospheric chemical pathways.[15] Another distinctive feature of emerging sources is that many of them are situated indoors, contrary to recognized sources such as traffic, which are essentially classified as outdoor emissions. For instance, a recent Canadian emission inventory estimated that approximately 60% of VCP emissions occur indoors, with coatings and cleaners identified as the dominant contributors.[16] Therefore, pollutant transfer from indoor spaces such as residences and offices to ambient air is expected to play an increasingly important role in urban air quality and ambient atmospheric chemistry.

Particulate matter has been the focus of most prior studies on indoor-to-outdoor transfer of air pollutants.[17,18] However, gaseous pollutants, including volatile organic compounds (VOCs), known as important precursors to secondary pollutants, have received less attention. Studies based on experimental measurements of indoor air quality impacts from various indoor activities, such as cooking and cleaning within a test evaluative space, have examined indoor VOC transport to outdoors by contrasting indoor and outdoor concentration profiles.[9,19–21] In addition to emissions associated with indoor activities, there are continuous indoor emissions from building materials and furniture, even from unoccupied spaces.[22] Xylenes, toluene, ethylbenzene, acetone, and formaldehyde, for example, are known to be emitted from wood products and construction materials.[23] Additionally, SVOCs and IVOCs, such as phthalates from plasticizers and brominated flame retardants, also contribute to indoor emissions from furniture and building materials.[24]

Although the controlled conditions of studies involving test indoor spaces are beneficial for a mechanistic understanding of emission dynamics and the related physicochemical interactions, real-world measurements in everyday spaces capture more realistic variations in emission intensity and timing. Stinson et al. quantified per-person and whole-building VOC emissions in a school by analyzing the exhaust airstream of its mechanical ventilation system.[25] Their results showed that monoterpenes (280 ± 80 µg per person per h) and isoprene (270 ± 60 µg person per h) were among the most significant occupant-derived emissions. Personal care product use contributed to siloxane emissions (D5: 5.3 ± 6.7 µg person per h), while eating and physical activity increased isoprene levels. Cleaning activities influenced ethanol and acetone concentrations. Their findings highlighted that 5.9 ± 1.7 g.h^{-1} of indoor-origin VOCs were emitted to the outdoor environment, with air scrubbing reducing total emissions by ~40%. These insights emphasize the role of human activity in shaping both indoor and outdoor air composition.[25] Residential buildings represent a larger fraction of the building stock in urban areas, serving as indoor spaces where the urban population spends most of their time.[26] Therefore, a study on residential buildings would provide new opportunities to advance knowledge about urban air quality. Multi-unit buildings, in particular, are expected to provide more representative insights, given the diversity of emissive activities.

One of the key factors in pollutant indoor–outdoor transfer is air exchange.[27] Air exchange rates vary across different spaces[28] and can change for the same space during different seasons depending on weather conditions.[29] Therefore, it is necessary to contextualize building net emissions in relation to the associated air exchange rates and examine indoor-to-outdoor pollutant transfer at locations and seasons different from previous studies (e.g., Texas during June[14] and Oregon during May[25]). Buildings located in regions with a colder climate often strive to maintain airtightness to ensure reasonable heating expenses, minimizing involuntary air exchange through building cracks. Under such conditions, a mechanical ventilation system offers a reliable means to characterize building air exchange rates provided that the system's performance remains within the design specifications. This condition addresses the challenges of monitoring the air exchange rate by tracer gas concentration evolution.[30] Energy efficiency considerations are expected to minimize the significance of alternative paths such as opening windows when the building is equipped with a mechanical ventilation system. Therefore, winter measurements involving buildings served by mechanical ventilation offer a robust characterization of air exchange rates and paths.

This study investigates indoor-to-outdoor transfer of VOCs from a residential space with 110 units in downtown Toronto, Canada, during February–March 2023. We analyzed the gas phase composition of air streams entering and leaving from the air handler of the building's mechanical ventilation system to inspect air pollutant occurrence and concentration. We identified the major sources driving the observed pollutant concentration trends via a source apportionment analysis. Building emissions were calculated from the measured concentrations and compared with predictions from our previous study[16] concerning indoor VCPs, as an important class of indoor sources. Finally, we illustrated the impact of indoor emissions on ambient air quality by examining the correlations and concentration ratios across the air handler's inlet and exhaust streams. The results of this work expand the current knowledge on the emerging role of buildings in urban air quality.

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2 Methods

2.1 Sampling period and location

Gas phase air samples were collected over 18 days from February to March 2023. The sampling location was a multi-storey residence in the Bathurst Quay neighborhood of downtown Toronto, Canada (Fig. 1). The building has 110 units with different floor areas, ranging from smaller one-bedroom ($\sim$240 m^2) to larger four-bedroom ($\sim$403 m^2) spaces. Based on information from the management, the building had approximately 200 residents during the sampling period. The Billy Bishop Toronto City Airport, a significant nearby point source, is located within less than one kilometer from the building's southern face.

2.2 Sampling procedure

To investigate the role of a multi-unit residential building as a net source of VOCs to the outdoor environment, we conducted sampling at the rooftop air handling unit rather than within individual units. The building's mechanical ventilation system includes two makeup air units (MAUs) that are installed on the building's rooftop. Based on the mechanical ventilation plans provided by the building's management, the MAUs were designed to deliver outdoor air to the common hallways and exhaust air from ducts connected to vents in living rooms, kitchens, and bathrooms. Fig. 1(c) shows a schematic diagram of air streams connected to the MAUs. The ventilation system connects exhaust emissions from the building's garbage room to one of the MAUs. We limited our samplings to the other MAU to avoid the potential complexities arising from waste emissions. Following the general protocol described in the U.S. Environmental Protection Agency Compendium Method TO-17,[31] we determined the gaseous pollutants by the gas chromatographic analysis of sorbent tube samples. The sampling setup and procedure are explained in Section S1.† Briefly, the volatile gaseous organic components of MAU's inlet and exhaust air streams were actively sampled onto adsorbent tubes filled with conditioned TENAX® TA. A quartz particle filter, impregnated with an ozone-scavenging reagent (Section S1.2†), was installed on the sampling line upstream of the adsorbent tube to avoid sampling airborne particulates and prevent the ozonolysis of sampled species.

Each building visit involved collecting four samples from the MAU's inlet stream and four from the exhaust stream, with each sampling interval lasting for two hours. The samples were collected sequentially, starting with an 8:00 to 10:00 am sample and finishing with a 2:00 to 4:00 pm (local time) sample. While short-lived emission events may influence individual samples, the concentration variations across adjacent sampling intervals
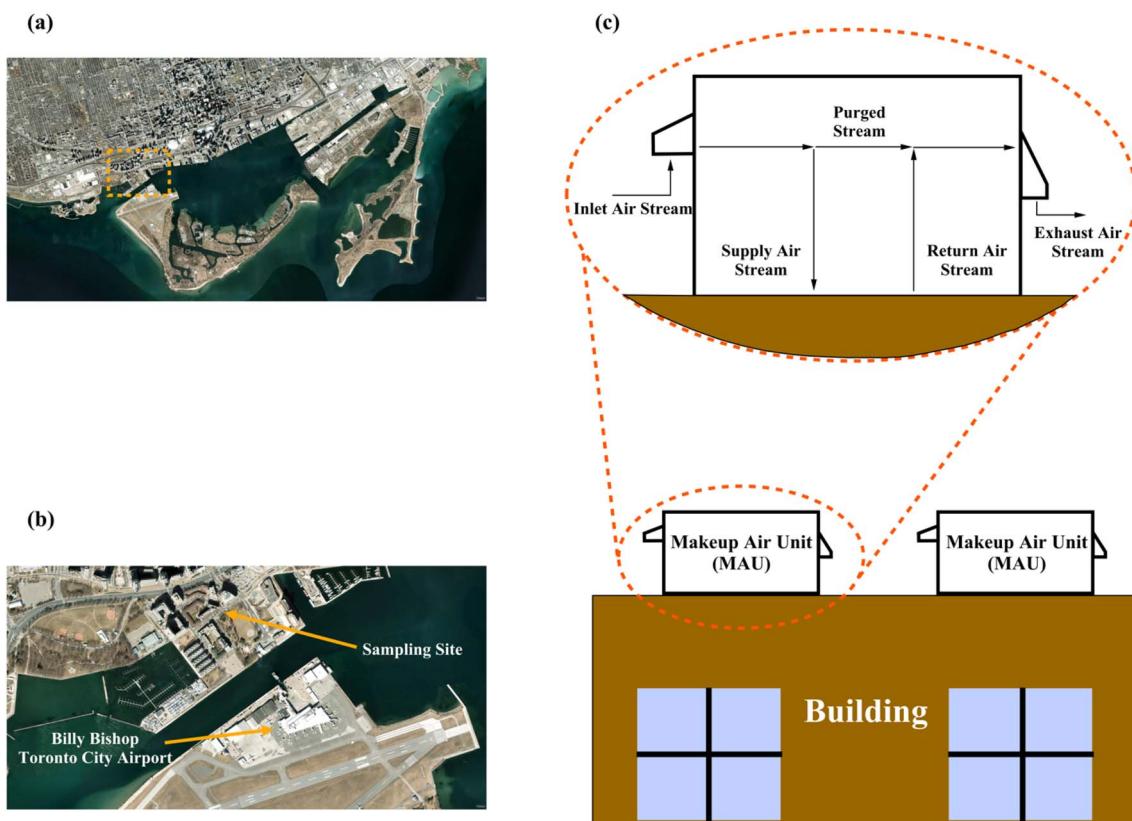

Fig. 1 Satellite images (generated by ARCGIS Pro software) of (a) the Bathurst Quay neighborhood, where the sampling site was located in downtown Toronto, Canada, and (b) the sampling site and the nearby Billy Bishop Toronto City Airport (magnified from the section designated by the dashed rectangle in panel (a)) and (c) a schematic of the building's mechanical ventilation system Make up Air Units (MAUs) with their connected air streams.

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were analyzed to assess the occurrence of dynamic emissions. This approach aligns with our study's focus on characterizing bulk emissions through integrated sampling rather than capturing highly transient fluctuations.

The adsorbent tubes were placed in tightly capped Teflon containers and shipped in cold-pack chests to the laboratory. All samples were analyzed within 24 hours after collection. See Section S1.2† for more information on the sampling procedures and details. In total, 129 samples, 18 analytical blanks, and 72 field blanks were collected during the campaign. The blanks were utilized in analytical instrument calibration and quality control/quality assurance procedures (Section S2.3†). Sampling took place under normal operating conditions of the building's mechanical ventilation system, ensuring representative data on air exchange and VOC transport. Based on air handler specifications and the building's estimated indoor volume, the total building air exchange rate is 0.52 h^{-1}, a value confirmed by manufacturer performance records. While air exchange rates can vary seasonally, natural ventilation was expected to be minimal during the colder months of this study.

2.3 Chemical analysis and quantification

The samples were analyzed using thermal desorption-gas chromatography/mass spectrometry (TD-GC/MS). The technical details of the analysis are included in Section S2.† Briefly, the sampled species were thermally desorbed *in situ* (Gerstel TDS3) and injected into a non-polar gas chromatography column along with helium as the carrier gas (Agilent 7890). GC eluents were ionized and analyzed using a quadrupole mass spectrometry detector (Agilent 5977). The species were identified and quantified through a software-aided search for chemicals based on their chromatographic retention indices and mass. Note that while real time analysis techniques, such as the approach adopted by Stinson *et al.*,[25] offer improved time resolution, that factor is not a critical advantage for this study, which is focused on the integrated analysis of emissions from a building and comparison to emission inventories. Furthermore, the challenges of deploying large and sensitive online instrumentation on the roof of a residential building during winter are logistically discouraging. Overall, our sampling approach can be much more easily and widely adopted by others and can facilitate broader comparisons in future studies.

2.4 Source apportionment through principal component analysis

Principal component analysis (PCA) was used to identify source-related trends in the dataset by reducing the dimensionality of VOC concentration data while preserving major variance patterns. This method was chosen for its ability to differentiate between mixed sources without requiring prior assumptions about emission profiles. The analysis included species with detection frequencies above 80% to ensure statistical robustness. The resulting principal components were interpreted based on species loadings, allowing for source characterization (*e.g.*, VCP-related *vs.* traffic-related emissions). A detailed breakdown of the

PCA methodology, including preprocessing steps and factor selection criteria, is provided in the ESI (Section S7).†

3 Results and discussion

3.1 Contrasting species occurrence and levels between the exhaust and inlet streams

Fig. 2(a) provides insight into the occurrence of species in the inlet and exhaust streams. A higher cumulative frequency at a given detection threshold suggests a greater overall occurrence of species. The observed shift in the exhaust stream towards higher cumulative frequencies indicates that more species were consistently present in the exhaust compared to the inlet. Fig. 2(b), in contrast, quantifies the overall signal intensity per sample volume across different volatility categories. Analysis of the exhaust stream samples often revealed higher concentrations of gaseous species compared to inlet data. Furthermore, the detection frequency was generally higher in the exhaust stream (Fig. 2(a)). As shown in Fig. 2(a), 37 out of 47 species quantified from the exhaust samples were detectable in at least 90% of analyses, whereas 27 out of 42 quantified species from the inlet samples had detection frequencies exceeding 90%.

The detected species are categorized based on their volatility in Fig. 2(b). We classified species based on their effective saturation concentration (C^*) at 298 K (Table S1†). Following Matsui *et al.*,[32] species with log $C^* > 6.5$ were categorized as VOCs, those with $2.5 < \log C^* \leq 6.5$ as IVOCs, and those with $0.5 < \log C^* \leq 3.5$ as SVOCs. Accordingly, we classified species more volatile than *n*-dodecane (log $C^* \sim 6.6$)[33] as VOCs. Those with volatilities between *n*-dodecane and *n*-docosane (log $C^* \sim 2.4$)[33] were classified as intermediate volatile organic compounds (IVOCs), and those less volatile than *n*-docosane were considered to be semivolatile organic compounds (SVOCs). Fig. 2(b) shows the total ion chromatogram (TIC) area, as a surrogate for total detected species mass, per unit volume of sampled air for inlet and exhaust sample series. VOC levels in the exhaust stream were significantly (*p*-value < 0.05) higher than in the inlet, with the exhaust median concentration being 22% higher. This suggests considerable VOC emission rates across the building envelope, making the building a net VOC source. In contrast, the means and medians of SVOCs and IVOCs were comparable between exhaust and inlet samples (less than 15% difference) with no statistically significant difference detected between the inlet and exhaust data. This closer parity is likely related to the elevated IVOC/SVOC emissions from outdoor combustion sources involving higher temperatures facilitating the release of less volatile species. Another potential cause could be the increased indoor sorptive sinks from air-to-surface partitioning, leading to losses of IVOCs/SVOCs. Overall, consistent with the results from previous studies,[21] the gaseous organic concentration profile associated with indoor emissions within the building envelope is more diversified and elevated compared to the ambient air, suggesting the significance of buildings as net sources to the outdoor environment.

Note that the discussion above regarding the relation between species concentrations and the corresponding

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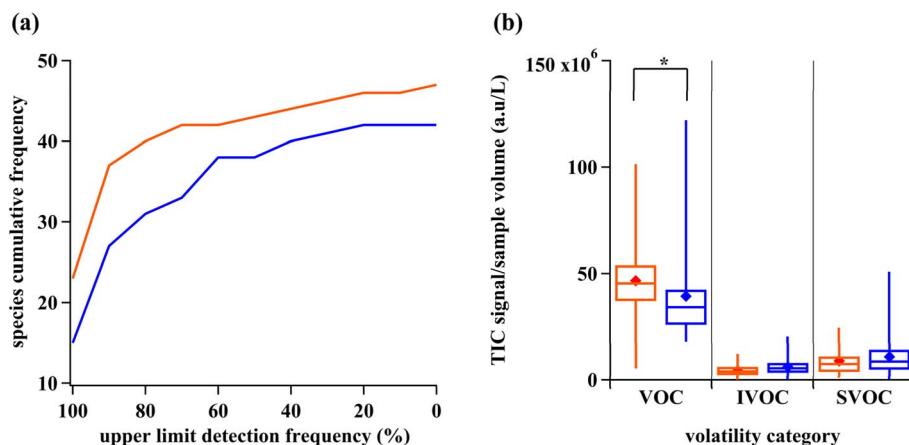
Fig. 2 (a) Cumulative number of detected species in the MAU's inlet and exhaust streams as a function of detection frequency. A species' detection frequency represents the fraction of samples in which it was detected. Higher cumulative frequencies for the exhaust stream indicate that more species were consistently present compared to the inlet stream. (b) Total Ion Chromatogram (TIC) signal in arbitrary units (a.u) per unit volume of sampled air for MAU's inlet and exhaust samples across three volatility categories: Volatile Organic Compounds (VOCs), Intermediate-Volatile Organic Compounds (IVOCs), and Semi-Volatile Organic Compounds (SVOCs). The box plot whiskers represent data minima and maxima, while diamonds indicate means. The asterisk denotes a statistically significant ($p < 0.05$) difference between inlet and exhaust data (Section S8†).

emission rates should be interpreted in light of different dilution and dispersion patterns across indoor and outdoor spaces. Accordingly, lower inlet stream concentrations may also result from increased dilution of outdoor air. Nevertheless, we believe that the elevated exhaust concentrations, as shown in Fig. 2–4, are primarily affected by net gaseous emissions from the building, as this notion is confirmed through comparing building emission rates calculated from the MAU's inlet and exhaust concentrations with emission inventories (Section 3.4).

3.2 Diurnal trends of VCP tracers and light aromatic species from the MAU's air streams

As mentioned above, VCPs, for which more than half of the emissive activity is predicted to occur indoors,[16] account for a large source of organic gaseous emissions.[10,34] Previous research has illustrated the impact of VCP emissions on urban air pollution *via* mobile and stationary samplings of ambient air;[12,35] However, the buildings' contribution is understudied. This section presents MAU's exhaust and inlet concentrations of species identified in previous studies as VCP-tracers[36] to underscore the role of buildings as mediators for air quality impacts of some key VCP sources (Fig. 3). The results are followed by the same analysis for the aromatic group of benzene, toluene, ethylbenzene, and xylenes (BTEX), known to be associated with combustion sources such as traffic exhaust emissions[37] (Fig. 4). During each of our 18 sampling visits, we collected 4 inlet and 4 exhaust air samples, leading to a total of 72 data points per two-hour interval across the campaign. All species shown in Fig. 3 and 4 were detected in every sample, ensuring consistency in sample size. The figures illustrate minimum, 5th percentile, median, 95th percentile, and maximum values to capture both routine and episodic emissions.

The exhaust and inlet concentration profiles shown in Fig. 3 and 4 are likely influenced by indoor and outdoor emission sources, respectively, which may differ in their daily patterns. Among the indoor sources, emissions from architectural and furniture coatings (*i.e.*, products that are applied to building's stationary structures and their accessories), paints, adhesives, and sealants occur steadily, leading to less temporal concentration variability. In contrast, episodic emissions associated with cleaning (*e.g.*, monoterpenoids and chlorinated organic compounds),[38] cooking (*e.g.*, ethanol and aldehydes),[9,39] and other human activities lead to more dynamic concentration profiles. For instance, Akteruzzaman *et al.* showed transient total VOC enhancements ranging from 7.95 to 35.66 ppm resulting from different indoor cooking practices.[40] For prominent outdoor source activity trends, traffic emissions peak during rush hours, especially in weekday early mornings.[41] Outdoor industrial and commercial emissions are expected to follow the business hours of the associated emitting facilities.

Fig. 3 shows the diurnal variability of concentration distributions of the VCP tracers octamethylcyclotetrasiloxane (hereafter referred to as D4), decamethylcyclopentasiloxane (hereafter referred to as D5), and monoterpenoids, which reflect emissions from solvent-based adhesives, personal care products, and fragranced products, respectively.[36]

Adhesives are known to exhibit a dual-phase emission pattern: an intense release of volatiles in the first few hours following application, followed by a prolonged, attenuated phase of lower emissions.[16,34] Given that the studied building has been occupied for over two decades, the dominant source of adhesive-related emissions is expected to be residual emissions from long-applied materials such as flooring, furniture, and structural components, leading to relatively stable median D4

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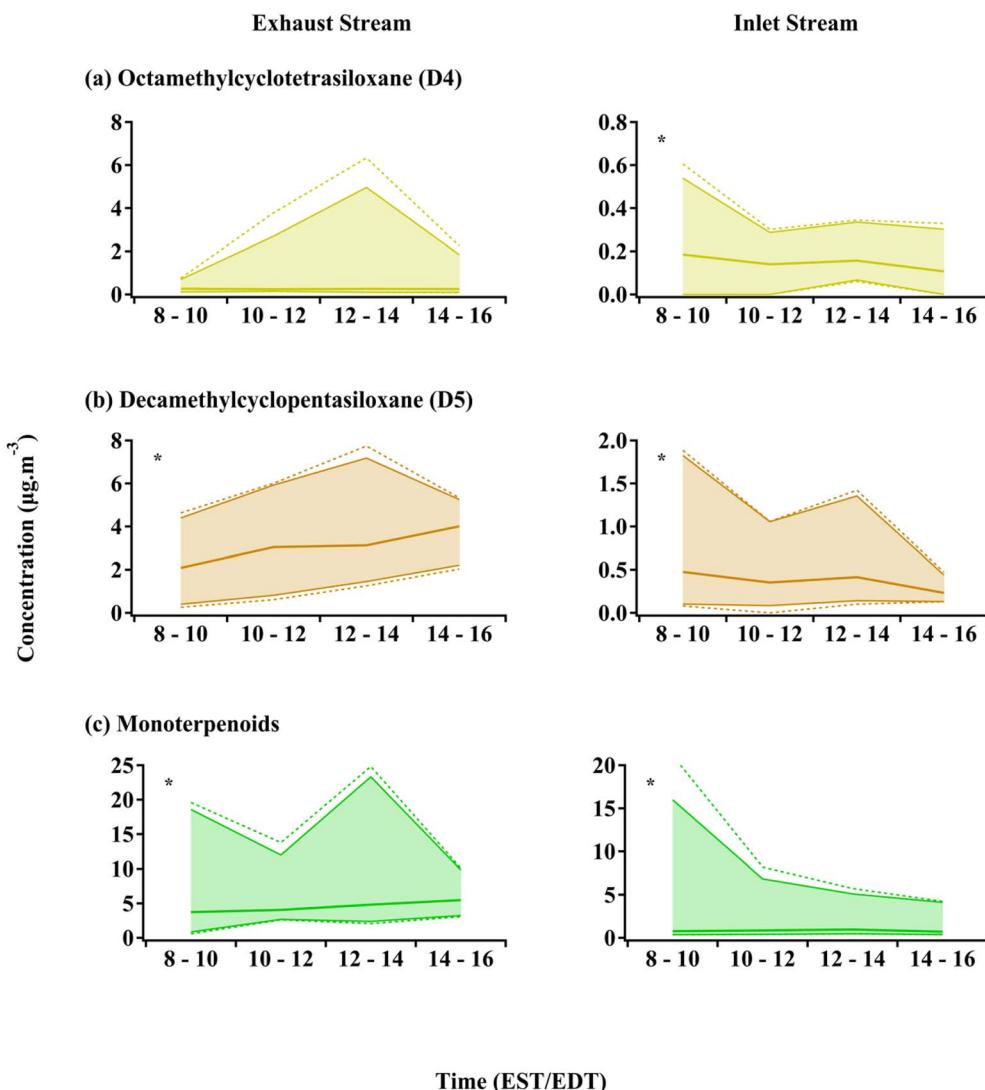
Fig. 3 Diurnal concentration variability profiles of Volatile Chemical Product (VCP) tracers for MAU's inlet and exhaust streams. The inspected VCP tracers are (a) octamethylcyclotetrasiloxane (D4), (b) decamethylcyclopentasiloxane (D5), and (c) monoterpenoids (see Section S2.2† for the full list of measured monoterpenoids). Each time interval represents 72 data points ($n = 72$), derived from 4 samples per visit across 18 visits. The lower and upper dashed lines correspond to data minima and maxima, respectively. The shaded bands represent the 5th to 95th percentile variability ranges. The thick lines within the bands indicate data medians. An asterisk on the upper left corner of a panel suggests statistically significant ($p < 0.05$) diurnal variability. See Section S8† for details of statistical significance analysis.

levels throughout the day. Accordingly, Fig. 3(a) shows no significant diurnal variability in D4 median concentrations from the exhaust stream. Nevertheless, there is an elevated D4 exhaust concentration in the 12:00–14:00 interval in Fig. 3(a), more than 30 times larger than the median concentration within the same interval. This condition resulted from a single outlier related to one of the samples (March 7, 2023), which is suspected to be caused by a freshly applied adhesive product within the building at that time. The maximum D4 concentration over the 14:00 to 16:00 interval occurred on the same day, which agrees with the fresh adhesive hypothesis above as it has been shown that such emissions peak during the first hours of product application, then plateau to lower steady levels.[42] Among the inlet concentration profiles, D4 levels during the 14:00 to 16:00 interval are significantly (p-value < 0.05) lower

than the other sampled periods, with late-afternoon concentrations lower by 20–40%. Since indoor adhesive-related emissions tend to follow a stable, long-term decay profile, and no corresponding trend was observed in the exhaust stream, we argue that the late-afternoon variations in the inlet stream are primarily associated with fluctuations in industrial D4 emissions rather than indoor sources. Given that industrial coating emissions are the dominant outdoor source of D4 (Table S6†), it is plausible that this decline is influenced by variations in industrial emission rates during business hours.

D5 concentrations within the MAU's exhaust stream are expected to reflect personal care product use by the building residents and hence depend on indoor occupancy. Accordingly, we believe that the overall increase in median D5 concentrations from the exhaust stream occurs as a result of residents
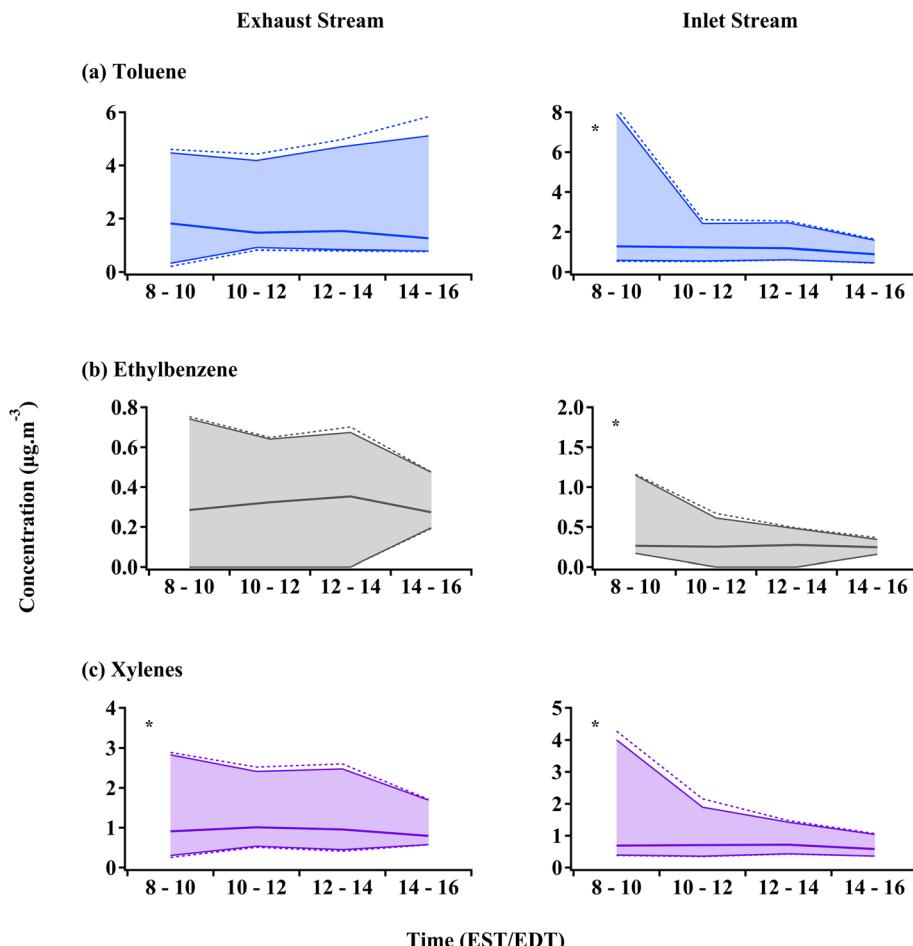
Fig. 4 Diurnal concentration variability profiles of simple aromatic compounds (a) toluene, (b) ethylbenzene, and (c) xylenes (including the *ortho*, *meta*, and *para* isomers) often associated with combustion emissions, especially traffic exhaust, for MAU's inlet and exhaust streams. The lower and upper dashed lines correspond to data minima and maxima, respectively. Each time interval represents 72 data points ($n = 72$), derived from 4 samples per visit across 18 visits. The shaded bands represent the 5th to 95th percentile variability ranges. The thick lines within the bands indicate data medians. An asterisk on the upper left corner of a panel suggests statistically significant ($p < 0.05$) diurnal variability. See Section S8† for details of statistical significance analysis.

returning from work during the later hours of the day. In agreement with the latter interpretation, D5 exhaust concentrations during the 8:00 to 10:00 am interval were higher among the weekend samples and less variable than those the weekdays (Section S3†). To explain the observed diurnal patterns of D5 concentrations in the inlet data, it is important to consider its volatility and emission dynamics. With an estimated vapour pressure of about 20 Pa at the room temperature,[43] D5 is not expected to evaporate immediately after application. Instead, following the indoor use of personal care products, such as shampoos, lotions, and deodorants, D5 may continue off-gassing from the skin and clothing. Therefore, although our sampling period did not cover intervals before 8:00 am, residual enhanced emissions from the earlier hours are likely to drive D5 inlet concentrations peak during the 8:00–10:00 am interval (Fig. 3(b)). This interpretation aligns with previous urban air quality studies, which have reported morning-time enhancements in ambient D5 concentrations linked to personal care product emissions from commuters[44] and the temporal trend of

monoterpenes, which also peak in the morning. Since monoterpenes are key fragrance ingredients in personal care products and their biogenic emissions are negligible during the cold months of the year, their co-occurrence with D5 reinforces the hypothesis of morning-time emissions from commuters. The subsequent D5 inlet (outdoor) concentration peak during the 12:00 to 14:00 interval, similar to the trends previously observed in New York City,[36] may have resulted from the re-application of some personal care products (*e.g.*, skin care and cosmetics) around lunchtime. Note that this trend does not necessarily imply that personal care products are applied outdoors. Instead, we believe that the reapplication of D5 indoors (*e.g.*, in office buildings) leads to delayed emissions into the outdoor air *via* ventilation. Given that our study site is downwind of Toronto's financial district, it is reasonable to conclude that emissions from surrounding office spaces influence the observed D5 trends (Fig. 1 and S3(c)†). Although personal care product reapplication by residents within the studied building may partially contribute to D5 exhaust stream enhancements in

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Fig. 3(b), since 80% of our samples were collected on weekdays when many residents are away, outdoor-to-indoor transfer likely contributes at least in part to the observed exhaust stream enhancement.

The observed profiles of monoterpenoid concentrations in Fig. 3(c) are expected to result from anthropogenic emissions as the biogenic emissions from plants are expected to be minimal during cold months of the year.[45] We believe that indoor monoterpenoid levels, represented by exhaust stream data in Fig. 3(c), are associated with emissions from fragranced cleaning and personal care products. Previous indoor air quality studies suggest that cleaning emissions peak during the cleaning time and decay quickly within a few minutes afterwards.[19] The significant (p-value < 0.05) diurnal variability of monoterpenoids within the MAU's exhaust stream in Fig. 3(b) is consistent with this dynamic nature of cleaning emissions. Note that the MAU's exhaust diurnal profile is not expected to synchronize with cleaning activities because of the air retention time in the mechanical ventilation system (Section S5.2†). The significantly higher (p-value < 0.05) monoterpenoid levels within the inlet data during the 8:00 to 10:00 interval likely originate from fragranced personal care product emissions from early-morning commuters. In addition to emissions from fragranced products, the monoterpenoid diurnal variability profiles in the MAU's inlet data may partly be affected by traffic emissions[46] although this scenario is less applicable to our measurements as our sampling location was not often upwind of the nearby major roads (Section S1.3†). In general, distinguishing between traffic and fragranced emissions from outdoor commuters is not straightforward because they occur concurrently.[44] Further data on fuel monoterpenoid emission factors can assist future studies to elucidate the traffic contribution by comparing measured ambient concentrations with estimated vehicular emissions. Note that higher monoterpenoid concentrations (18–$20\ \mu g\ m^{-3}$) were observed in 3 out of 18 sampling days. These outliers are not representative of typical morning trends, and while they may arise from alternative sources, we lack sufficient data to definitively identify their origin.

While the species depicted in Fig. 3 are associated with VCP sources, the aromatic compounds in Fig. 4, which belong to the BTEX group, are often assumed to be related to traffic emissions.[37] We excluded benzene from our quantifications because the sampling conditions (Section S1.1†) led to exceeding the critical adsorbent breakthrough volume for benzene, discrediting quantification results for this species. Inlet median concentrations of toluene, ethylbenzene, and xylenes were more comparable with the exhaust values, as opposed to pronounced exhaust-inlet differences for VCP tracers in Fig. 3. Upper limit aromatic inlet concentrations during the 8:00 to 10:00 am interval were higher than exhaust values, especially for toluene, likely due to elevated traffic emissions during the morning rush hour. The enhanced morning concentrations in the inlet data in Fig. 4 drive the significant inlet-stream diurnal variability for toluene, ethylbenzene, and xylenes. However, no significant diurnal variability was observed in toluene and ethylbenzene levels within the exhaust stream, indicating the role of indoor

sources in damping the dynamic effects of traffic emissions. We believe that VCP indoor emissions were the main contributors to this condition (Section 3.4). Among the indoor VCPs, the significant sources of toluene and ethylbenzene are printing inks, coatings, and some general-purpose surface cleaners (Section S4.1†). Although, in the exhaust stream data, xylenes showed a different trend from toluene and ethylbenzene, we believe the xylenes concentration profile, mainly driven by *meta* and *para* isomers (Section S3†), is still primarily influenced by indoor VCP emissions. Since the *meta* and *para* isomers are more abundant in some cleaner formulations,[47] the diurnal variation among the xylenes from the exhaust stream in Fig. 4 is likely due to episodic emissions related to such sources. Overall, the concentration profiles in Fig. 4 show some influence from indoor sources of light aromatic compounds in the exhaust air stream. This observation suggests that emerging sources, such as VCPs, are influential to the indoor quality even for species that are widely attributed to more recognized sources such as traffic.

3.3 Source apportionment analysis

Given the complex interplay of various indoor and outdoor emissions in shaping the observed concentration profiles, a source apportionment analysis is helpful to identify the pivotal emission sources. As it was mentioned in Section 2.4, to systematically identify key VOC sources, we applied principal component analysis (PCA), a statistical technique that reduces the dimensionality of complex datasets while preserving the essential variability in the data. By transforming our high-dimensional dataset—consisting of numerous quantified species—into a small number of principal components (PCs), PCA enables us to infer dominant sources that contribute significantly to observed concentration patterns. The PCA results indicate that the exhaust stream emissions are driven by five major PCs, explaining a total of 94% of the data variability, while the inlet stream emissions are captured by four PCs, accounting for 88% of the variability (Tables S8 and S9†). Fig. 5 and 6 present the results of principal component analysis (PCA) for the exhaust and inlet streams, respectively. The PCA loadings indicate which species contribute most strongly to each principal component (PC), while the score profiles illustrate how these components vary over time. The order of PCs in Fig. 5 and 6 follows their importance, with PC1 explaining the largest proportion of variability. All loadings are standardized to the range $[-1,1]$ to facilitate comparison across species. The diurnal score variations in Fig. 5 and 6 provide further insight into the temporal dynamics of VOC emissions. PCs dominated by less dynamic sources, such as coatings, tend to exhibit stable score profiles, reflecting their continuous emission behavior, whereas PCs influenced by transient emissions from personal care products or traffic display more pronounced time-dependent fluctuations. Similar graphs for the remaining PCs are included in the ESI (Section S6).† The following paragraphs present and discuss the PCA evidence regarding the effect of various evaporative and combustion sources on the measured air pollutant concentrations.
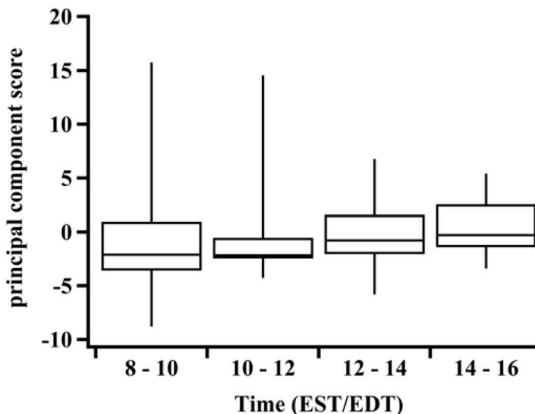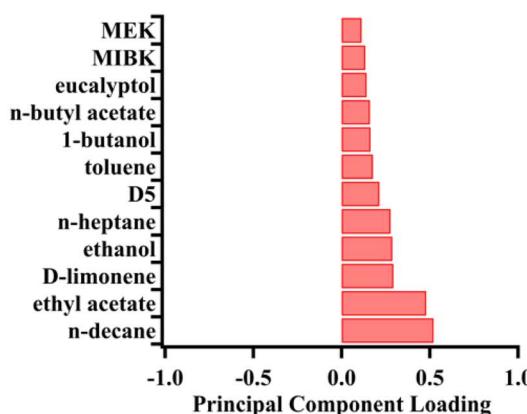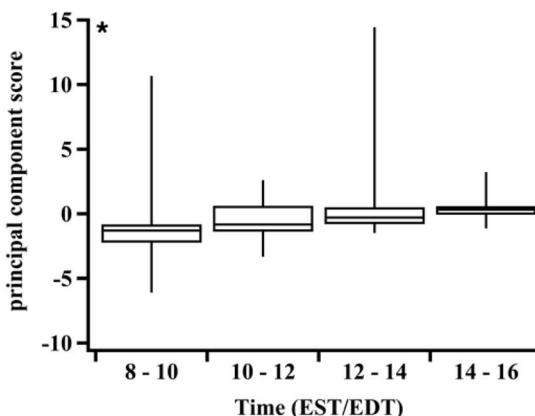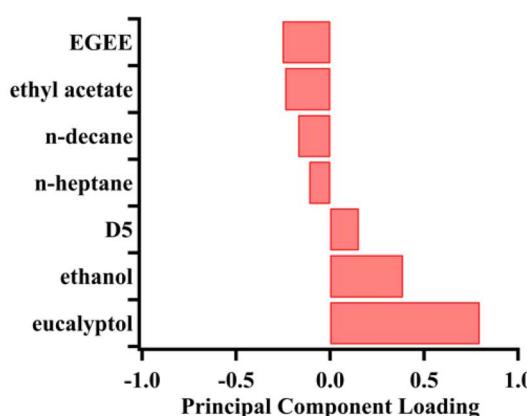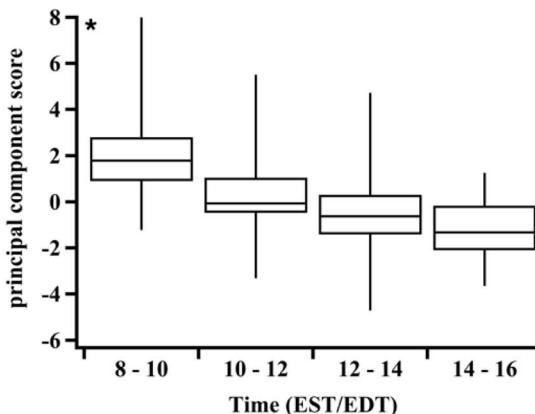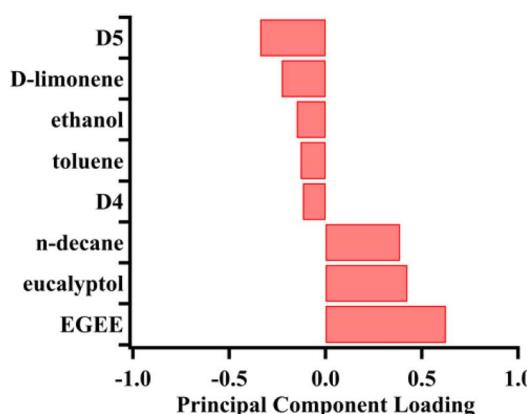
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Fig. 5   Principal component analysis (PCA) loading and score profiles for the (a) first (40% of data variability), (b) second (18% of data variability), and (c) third (12% of data variability) principal components (PCs) associated with the MAU's exhaust airstream data. PC loadings are standardized to values within the [−1,1] range. MEK, MIBK, and EGEE stand for methyl ethyl ketone, methyl isobutyl ketone, and ethylene glycol monoethyl ether, respectively. The box plot whiskers on PC score profiles correspond to score minima and maxima. An asterisk on the upper left corner of a score profile suggests statistically significant ($p < 0.05$) PCA score diurnal variability. See Section S8† for details of statistical significance analysis.

Among the evaporative sources, coating emissions have a notable impact on PCs shown in Fig. 5 and 6. Coatings have been identified in previous research as the most emitting source among the VCPs.[16,34] Fig. 5(a) illustrates that the first principal component (PC1) of the exhaust stream—representing the most important PC, as it explains the largest fraction of data
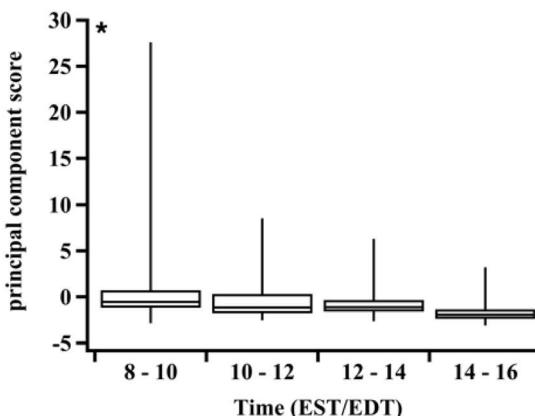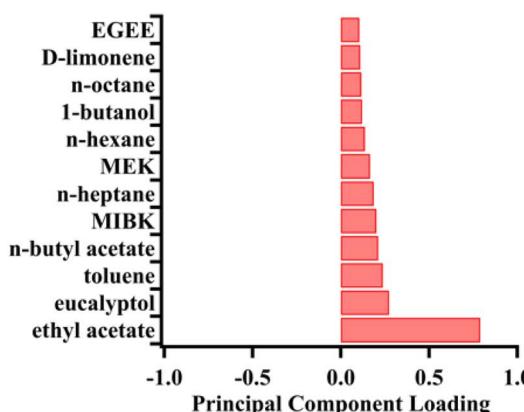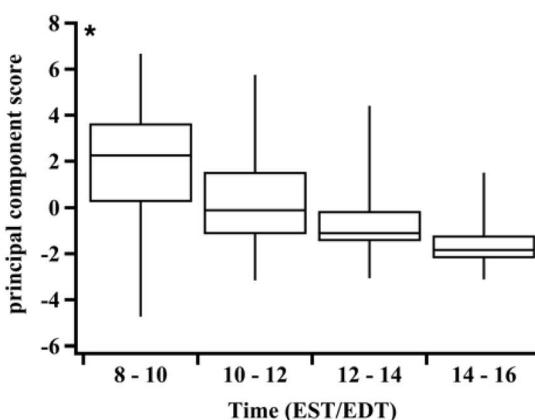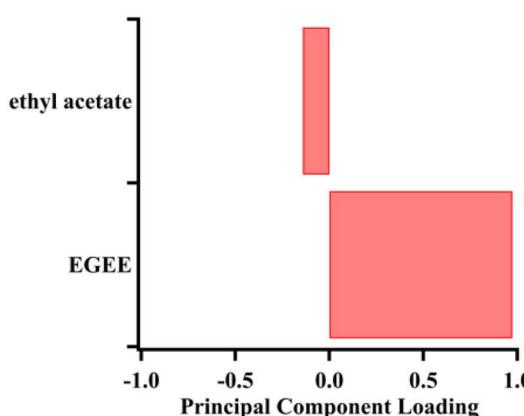
Fig. 6 Principal component analysis (PCA) loading and score profiles for the (a) first (63% of data variability) and (b) second (17% of data variability) principal components (PCs) associated with the MAU's inlet airstream data. PC loadings are standardized to values within the [−1,1] range. MEK, MIBK, and EGEE stand for methyl ethyl ketone, methyl isobutyl ketone, and ethylene glycol monoethyl ether, respectively. The box plot whiskers on PC score profiles correspond to score minima and maxima. An asterisk on the upper left corner of a score profile suggests statistically significant ($p < 0.05$) PCA score diurnal variability. See Section S8† for details of statistical significance analysis.

variability—is predominantly influenced by *n*-decane and ethyl acetate. Both compounds are found in VCP formulations, especially coatings,[16,47] as well as fuel blends.[48] We believe the impact of traffic on the exhaust stream's first PC is minor compared to coatings because other essential traffic emissions such as normal- and iso-octane are either absent from Fig. 5(a) or feature reduced loadings, such as toluene. Furthermore, the first exhaust's stream PC score demonstrates no significant diurnal variability in agreement with the steady emission pattern associated with coatings.

Given the importance of VCPs among indoor emission sources and the previous research findings that cleaners and personal care products are the highest emitting VCP sources following coatings, these sources are also expected to influence exhaust stream's PCs. Fig. 5(b) depicts that the exhaust stream's PC2 (18% variability) is primarily associated with D5 and monoterpenoids, such as eucalyptol, alongside ethanol, a known marker for cleaning agents (see Table S6†). The diurnal profile of PC2 scores exhibits clear trends, with an elevated

upper bound during the 8:00–10:00 am interval and higher median values in the late afternoon. These patterns align with human activity, such as morning and evening personal care product use and periodic cleaning activities within the building. Additionally, ethanol emissions from cooking during lunchtime may further contribute to PC2 score variations.

Note that in addition to ethanol, cooking emissions are known to include monoterpenes and aldehydes, particularly saturated aldehydes such as hexanal and heptanal, which were detected in this study. Less volatile compounds emitted from cooking, including fatty acids and organic aerosol particulates, were not measured due to the instrumentation's focus on volatile gaseous organics. Moreover, the variability in cooking emissions—depending on the food type, cooking method, and procedure—makes precise source attribution difficult in the absence of direct observational data. In contrast, VCP formulations exhibit less variability, allowing for more robust and reliable source attribution in discussions of building emissions. Emissions from cleaners and personal care products are

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associated with indoor occupancy and tend to decrease during the middle of the weekdays, when some building residents are away at work. As expected, the data indicate that the exhaust stream's second PC scores, as well as D5 and monoterpenoid concentrations, are more comparable across the different time intervals during the weekends (Section S3†).

The effect of traffic, as a key outdoor source among the combustion emissions, is primarily reflected in the inlet stream's PC1 (63% of data variability), whose loading profile features fuel ingredients such as toluene and *n*-octane, fuel additives such as methyl ethyl ketone (MEK), methyl isobutyl ketone (MIBK), and butyl acetate,[49,50] as well as vehicle oil additives such as ethyl acetate (Fig. 6(a)).[47] The elevated upper bound of PC1 scores during the 8:00–10:00 am interval coincides with the morning rush hour, indicating a substantial contribution from vehicular emissions. The presence of coating-related markers suggests that ventilation from surrounding buildings also plays a role in shaping ambient air composition. Thus, the first inlet PC corresponds not only to coatings, as discussed above, but also to traffic emissions.

The PC loading profiles depicted in Fig. 5(c) and 6(b) are primarily associated with ethylene glycol monoethyl ether (EGEE). The PC score diurnal variability profiles of these two factors are also similar, involving monotonically decreasing medians. Hence, we think these PCs are affected by the same emission source(s) that are associated with EGEE. EGEE emissions may originate from antifreeze systems used in vehicles and aircraft.[47] However, the EGEE diurnal concentration pattern (Section S3†) does not resemble traffic emission dynamics, characterized by elevated activity during the morning and late afternoon rush hours. Aircraft emissions are also unlikely to have an appreciable effect on the observed EGEE levels as the nearby Billy Bishop local airport is seldom upwind of the sampling site (Fig. 1 and S3†). We also rule out the effect of EGEE emissions from VCP sources. Among VCP sources, EGEE is almost totally associated with coating emissions (Table S3†). Industrial coating emissions may feature more time variability compared to architectural coatings, whose steady emissions are not reconcilable with the significant diurnal variabilities in PC scores, as shown in Fig. 5(c) and 6(b). However, we could not identify any major facility within 10 kilometers of the sampling site with emission activities involving EGEE upon inspecting the National Pollutant Release Inventory (NPRI).[51] These PCs may be impacted by a complex interplay between multiple sources, including those mentioned above. Hence, we designate the exhaust's third PC and the inlet's second PC as EGEE-related principal components in the absence of enough evidence to accurately attribute them to a specific emission source.

Overall, by elucidating key species and their variation time trends, the PCA illustrates that exhaust stream profiles associated with building emissions are pivotally affected by VCP emissions related to coatings, cleaners, and personal care products. Coatings and adhesives primarily emit species such as D4 and glycol ethers, cleaners release oxygenated VOCs such as ethanol, glycol ethers, and monoterpenes, while personal care products are major sources of D5, monoterpenes, and fragrance compounds.[16,36] A detailed breakdown of species-source associations is provided in Table S6.† Given the variability in formulations and product usage, some overlap exists between emissions from different sources. Meanwhile, the inlet stream PCA, which reflects ambient air effects, identifies traffic and coating emissions as the primary contributors to the observed data.

3.4 Comparing observed building net emissions to VCP inventory data

The PCA results described above identified VCPs as key contributors to the MAU's exhaust stream composition. Furthermore, the total organic gaseous species concentration comparison across the inlet and exhaust streams suggested the building as a net emission source (Fig. 2). The role of indoor VCP sources in driving the building emissions can be further substantiated by comparing this study's observations to an applicable VCP emission inventory. Considering the building as a control volume, we utilized mass and energy balance equations involving the MAU's exhaust and inlet concentrations to calculate species-resolved building net emissions (see Section S5† for the calculation details). We compared the resulting emission rates with indoor VCP emissions expected from a Toronto building of the same size based on a published VCP emission inventory for Canada[16] in Fig. 7 (see Section S4.2† for more information). The median observations and emission inventory predictions differed by less than 15% for recognized VCP emission tracers such as D-limonene, fragrances (assumed to be equivalent to monoterpenoids), and volatile methyl siloxanes, *i.e.*, D4 and D5 (Fig. 7(d)). Although the parity across observations and emission inventory estimates was weaker for other species, the difference remained within one order of magnitude, except for *n*-undecane and *n*-dodecane. We could not identify major non-VCP sources for *n*-undecane. However, *n*-dodecane has flavoring applications.[48] Thus, *n*-dodecane underestimation shown in Fig. 7(a) may be related to cooking emissions. Overall, we consider the deviations across the observations and estimations in Fig. 7 acceptable, given the uncertainties of the VCP emission inventory estimations.[16]

Despite the median deviations across observations and predictions, their variation ranges overlap for most of the species shown in Fig. 7, including some normal alkanes (Fig. 7(a)) and aromatics (Fig. 7(c)). Thus, although the latter two classes of compounds are often assumed to originate from traffic, our findings suggest that their ambient levels are also influenced by building emissions. This observation indicates that the mitigation strategies to combat air pollution caused by traffic organic gaseous emissions may need to be complemented by addressing VCP sources from buildings to address public health concerns associated with urban air quality.

Although we consider the studied building to fairly represent an average multi-unit residence in Toronto, generalizing the results shown in Fig. 7 to other buildings must be carried out with caution. Indoor VCP emissions vary per variables such as building age, airtightness, occupancy level, ventilation rate and mode, as well as typical indoor activities. Our results may not be

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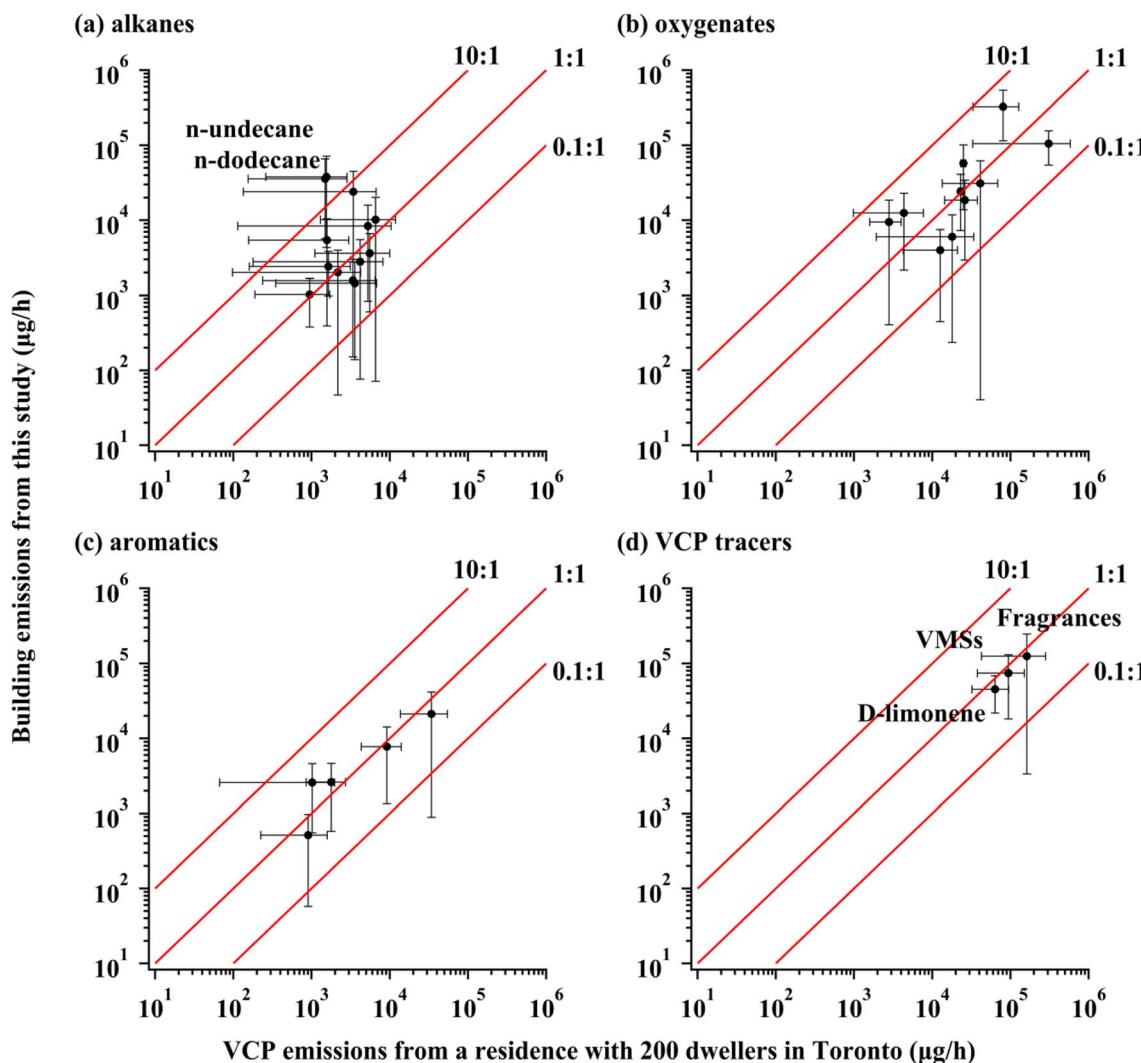
Fig. 7 Parity plots for (a) alkanes, (b) oxygenates, (c) aromatics, and (d) VCP tracers comparing net building emissions calculated from the mechanical ventilation rate (see Section S5†) and MAU's inlet and exhaust stream concentrations *versus* the indoor VCP emission predictions for a building with the same number of occupants in Toronto, Canada, based on the VCP emission inventory developed by Askari and Chan.[16] The points and error bars correspond to the median and 5th to 95th percentile intervals, respectively.

applicable to indoor spaces differing appreciably from the studied building regarding the parameters above.

3.5 Impact of building emissions on ambient air quality

The previous sections established the role of buildings as net emission sources in Toronto, a typical metropolitan area in North America. This conclusion begs the question of whether the building emissions are strong enough to impact ambient air pollutant trends. Fig. 8 shows the exhaust-to-inlet concentration ratios for compounds featuring at least in one of the PC loadings of Fig. 5 and 6. The median ratios in Fig. 8 are close to or higher than unity, indicating elevated exhaust concentrations for these species, in agreement with the overall trends discussed in Section 3.1. The occasional instances of exhaust-to-inlet ratios below one in Fig. 8 suggest conditions where the building is a net sink for the species of interest. We believe indoor species sinks, time-dependent phenomena not

considered through our steady-state analysis (see Section S5†), and alternative ventilation paths are among the factors that can explain this observation.

A correlation analysis across effective MAU's exhaust and inlet stream concentrations helps to illustrate the impact of building emissions on ambient air quality as a significant correlation, in combination with concentration ratios discussed above, suggest ambient levels to be influenced by building emissions. The air residence time within the building envelope is approximately 2 hours (Section S5.2†). Therefore, the exhaust stream concentrations were paired with inlet stream values sampled two hours earlier for the correlation analysis, leading to the exhaust stream's 8:00 to 10:00 am and inlet stream's 2:00 to 4:00 pm series to be excluded from the correlation analysis due to the lack of corresponding data. Fig. 8 shows significant correlations (p-value < 0.05) between indoor and outdoor concentrations for eleven out of sixteen species. This

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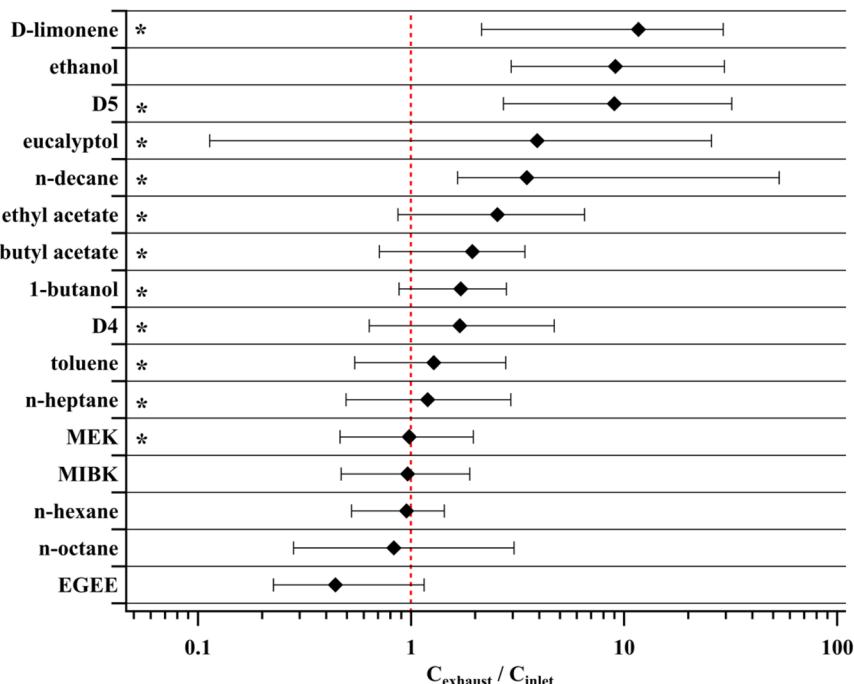
Fig. 8 Concentration ratios for effective indoor levels relative to outdoor values for species featuring in PC scores of MAU's inlet and exhaust data in Fig. 5 and 6. The error bars correspond to the 5th to 95th percentiles. An asterisk next to the species name suggests a statistically significant ($p < 0.05$) correlation between effective indoor and outdoor concentrations. MEK, MIBK, and EGEE stand for methyl ethyl ketone, methyl isobutyl ketone, and ethylene glycol monoethyl ether, respectively.

observation is in favor of a common source influencing both indoor and ambient concentrations. The lack of a significant correlation for ethanol, n-hexane and n-octane may arise from their emissions occurring primarily from traffic. Overall, given the insights presented above about net emissions from the building, we believe that the correlation results in Fig. 8 confirm the influence of indoor emissions, especially from VCPs, on ambient air quality and underscore the buildings as critical factors in determining air pollution in urban regions with a high density of built area. Beyond their direct impact, building emissions also play a role in the formation of secondary pollutants such as ozone and SOA. For instance, monoterpenes, which are emitted from personal care products and cleaning agents, are well-documented precursors for both SOA and O_3 formation through atmospheric oxidation.[52] Additionally, D5 siloxane, a key compound in personal care products, has been found to contribute to SOA formation under ambient conditions.[53]

4 Conclusions

This study presents experimental evidence to illustrate the emerging role of buildings as net emission sources of organic gaseous pollutants. We collected air samples from the inlet and exhaust streams of a mechanical ventilation system of a multi-unit residence in downtown Toronto. Volatile organic compounds were more prevalent and concentrated in the exhaust stream compared to the inlet. 37 out of 47 quantified species were detected in at least 90% of exhaust samples,

whereas this detection frequency applied to only 27 out of 42 species in the inlet stream. Total VOC concentrations were significantly ($p < 0.05$) higher in the exhaust stream, ranging from 0.1 to 30 $\mu g\ m^{-3}$ depending on species volatility. Source apportionment results confirmed volatile chemical products (VCPs) as the primary drivers of exhaust concentrations, with coatings, cleaners, and personal care products emerging as dominant contributors. Specifically, siloxanes (D4 and D5) from personal care products, monoterpenoids (e.g., D-limonene and α-pinene) from fragranced cleaners, and oxygenates (e.g., ethanol and ethyl acetate) from coatings and cleaning agents were the most abundant species in the exhaust stream. Building net emissions were within the predicted bounds of indoor VCP emissions estimated from a published inventory for Canada, with parity errors smaller than 15% for VCP tracer species.[16] Significant correlations were observed between exhaust and inlet concentrations for most of the key species, suggesting the impact of building emissions on ambient air quality. Overall, the results of this study support the critical role of indoor emissions associated with VCPs, as an emerging source of organic air pollutants, when the emissions are transported outdoors via building ventilation paths. Future regulations concerning VOCs should include indoor sources such as coatings, adhesives, cleaners, and personal care products by mandating product reformulations or design changes. Furthermore, knowledge mobilization efforts to make people aware about lifestyle choices, such as using heavily scented products, will be useful to control and mitigate air pollution impacts associated with residents' indoor activities.

As research efforts aimed at improving air quality delve further into investigating significant anthropogenic emission sources, exploring the contribution of building emissions will enhance our understanding of urban air pollution. Scaled-up building emission predictions can be constrained experimentally (e.g., using eddy covariance) to examine the collective effect of buildings on ambient air pollution in densely populated urban regions. We suggest that future research extend the air sampling periods beyond the morning to afternoon interval considered in this study and utilize online sampling techniques to illustrate dynamic trends, especially for sources such as cleaners and personal care products. Future studies can also be better informed by gathering information about building architectural emission sources, such as paints and construction materials, in addition to data on timing and details of indoor emissive activities such as cleaning, cooking, and using personal care products. Our study was focused on indoor-to-outdoor pollutant transfer through mechanical ventilation. It is crucial to investigate the implications of alternative transfer paths that may cross the building envelope via other pathways, such as natural ventilation. For example, emissions from an open window located on a middle floor of a tall building may make building emissions subject to enhanced street canyon effects compared to the rooftop.[54] Computational fluid dynamics models coupled with dispersion calculations can investigate the significance of such phenomena for various scenarios.[55] Overall, conducting more extensive measurements and investigating the different ventilation paths and intensities, which may involve more interdisciplinary research in collaboration with building sciences experts, are key to improving our understanding of the impact of buildings on ambient air pollution.

Data availability

Data for this article, including measurements of VOC concentrations in the exhaust and inlet streams, are available at Figshare: **https://doi.org/10.6084/m9.figshare.24328816**.

Conflicts of interest

The authors declare no financial competing interest.

Acknowledgements

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