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Human personal air pollution clouds in a naturally ventilated office during the COVID-19 pandemic

Shen Yang [a,*], Akila Muthalagu [a,b], Viviana González Serrano [a], Dusan Licina [a]

[a] Human-Oriented Built Environment Lab, School of Architecture, Civil and Environmental Engineering, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland
[b] Environmental Systems Group, Department of Civil Engineering, Indian Institute of Technology Hyderabad, Kandi, India

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ABSTRACT

Personal cloud, termed as the difference in air pollutant concentrations between breathing zone and room sites, represents the bias in approximating personal inhalation exposure that is linked to accuracy of health risk assessment. This study performed a two-week field experiment in a naturally ventilated office during the COVID-19 pandemic to assess occupants' exposure to common air pollutants and to determine factors contributing to the personal cloud effect. During occupied periods, indoor average concentrations of endotoxin (0.09 EU/m^3), TVOC (231 μg/m^3), CO_2 (630 ppm), and PM_{10} (14 μg/m^3) were below the recommended limits, except for formaldehyde (58 μg/m^3). Personal exposure concentrations, however, were significantly different from, and mostly higher than, concentrations measured at room stationary sampling sites. Although three participants shared the same office, their personal air pollution clouds were mutually distinct. The mean personal cloud magnitude ranged within 0–0.05 EU/m^3, 35–192 μg/m^3, 32–120 ppm, and 4–9 μg/m^3 for endotoxin, TVOC, CO_2, and PM_{10}, respectively, and was independent from room concentrations. The use of hand sanitizer was strongly associated with an elevated personal cloud of endotoxin and alcohol-based VOCs. Reduced occupancy density in the office resulted in more pronounced personal CO_2 clouds. The representativeness of room stationary sampling for capturing dynamic personal exposures was as low as 28% and 5% for CO_2 and PM_{10}, respectively. The findings of our study highlight the necessity of considering the personal cloud effect when assessing personal exposure in offices.

1. Introduction

Air pollution is one of the great threats of humans' age, given the strong link with premature mortality and reduced life expectancy [1–3]. Due to the fact that humans spend most of their time indoors [4], indoor air pollution accounts for a dominant proportion of their daily exposure [5]. Indoor air contaminants include radioactive (e.g., radon [6]), gaseous, and particulate pollutants. Volatile organic compounds (VOCs) are the most prevalent gaseous pollutant indoors [7,8], originating from various materials, humans and their activities, and intrusion from outdoors [9–12]. Exposure to VOCs is related to irritation and respiratory symptoms [13,14], whereas some chemicals are carcinogenic [15]. CO_2 is another well-known gaseous pollutant, mainly emanating from human exhalation and indoor combustion. Elevated exposure to indoor CO_2 has been associated with decreased human productivity and cognitive performance [16–19]. Airborne particles are known carcinogens [20], and exposure to PM_{10} (particles with aerodynamic diameter ≤10 μm) can result in respiratory and cardiopulmonary health issues [21,22]. Endotoxin, a cell wall bound component of gram-negative bacteria, is an important biological component of PM_{10} [23]. Endotoxin can adversely affect human health by activating the alveolar macrophages to release cytokines, which are chemoattractants leading to a cascade of inflammatory effects [24].

Personal exposure and air quality in office environments are of special interest owing to their enormous economic implications [25]. Although multiple studies quantified air pollutant concentrations by field measurements in offices [26–33], their results may not accurately characterize employees' personal exposures. An important, yet overlooked, exposure determinant is the spatial variability of indoor pollutant concentrations. Perfect air mixing is not expected in office environments [34,35]. Owing to spatial and temporal relationships among air pollutant sources, sampling locations, and human breathing zone, exposure estimation based on measurements at stationary room sampling stations could introduce bias. "Personal cloud" effect refers to

* Corresponding author.
E-mail address: shen.yang@epfl.ch (S. Yang).

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the difference in air pollutant concentrations between breathing zone and room sites, representing the bias of approximating personal exposure governed by spatial air pollution gradients. It is firstly introduced by Özkaynak et al. to describe the excess of PM_{10} concentrations (~50 $\mu g/m^3$) measured at personal sites relative to the room average values in residences from their seminal PTEAM study [36]. The personal air pollution clouds can result from exogenous sources related to human activities and proximity to localized emissions, such as cooking and smoking [37–43]. In addition, endogenous sources can also contribute to the personal cloud, which mainly refer to emissions from human breathing, skin, clothing, and applied personal care products. Humans are potent sources of CO_2, VOCs, particles, and microbes in indoor environments [44–55]. The released air pollutants can elevate the pollution level in the peri-human microenvironment, and cause elevated personal exposures relative to the room background level. Such a personal cloud effect has been documented in personal exposure studies in residential homes, mostly for particles and few for VOCs [36,56–60]. The RIOPA (Relationships of Indoor, Outdoor, and Personal Air) study involving 219 homes in the US found similar median indoor and outdoor stationary measurements of $PM_{2.5}$ (14.4 and 15.5 $\mu g/m^3$, respectively), whereas the median personal $PM_{2.5}$ concentration reached 31.4 $\mu g/m^3$, two times higher than the stationary measurements [61]. An endotoxin measurement campaign in schools for children with asthma found significant higher personal exposures than indoor/outdoor stationary levels of endotoxin (geometric mean 0.07 vs. 0.02 EU/m^3) and concluded the importance of considering personal endotoxin cloud to monitor exposure of vulnerable populations [62]. There have also been investigations on the personal cloud of particles and CO_2 using controlled chamber study [63,64] and computational fluid dynamics (CFD) simulation [65]. In a controlled chamber simulating office environments, Pantelic et al. [63] detected a median personal CO_2 cloud magnitude at 200–500 ppm, depending on metabolic generation, posture, and breathing pattern. Similarly, Licina et al. [64] reported a personal PM_{10} exposure increment of 1.6–13 $\mu g/m^3$ above the room average levels in a simulated office using an environmental chamber. However, studies exploring the personal cloud effect in field office environments are limited.

Offices generally have lower occupancy density relative to other public indoor spaces, such as cars and aircraft cabins. Along with generally limited movements, it is expected that spatial air pollution gradients in offices, and therefore the personal cloud magnitude, are relatively more pronounced [64,66]. The EXPOLIS study reported differences in PM composition and VOC concentrations between measurements at personal sites and in workplaces in Helsinki [67,68]. They found that personal measurements of D-Limonene, alpha pinene, and hexanal were significantly higher than workplace concentrations, but lower for styrene, hexane, and cyclohexane. A recent study in Swiss offices detected the existence of personal PM_{10} at a magnitude ranging 5–37 $\mu g/m^3$, whereas personal CO_2 clouds were mainly found in private or low-occupancy offices [69]. Nevertheless, to our knowledge, field tests in offices exploring CO_2, VOCs, particles, and endotoxin through both personal and room-average measurements have not been documented. In addition, interpersonal differences and factors driving the personal cloud effect in the office environment, especially in the context of the COVID-19 pandemic, are yet to be explored.

The COVID-19 pandemic has brought more public attention to indoor air quality [70–73]. The influence of COVID-19 on occupants' health and exposure is not occurring only through the direct path of airborne transmission, but also via indirect ways. The pandemic has altered office settings (such as occupancy level and ventilation rates) and occupant behaviors (such as the use of hand sanitizers and more frequent opening of windows). Personal exposure in offices during the pandemic has been, nevertheless, poorly investigated, and most of the investigation has focused on homes and home offices where people spend increasing time [74–77]. Additionally, while the use of hand sanitizers has been widely recommended, it remains unclear about the potential unintended consequences on indoor air quality and inhalation exposure [78]. Therefore, there is a clear need and value in performing the field office experiments during the COVID-19 pandemic to understand how the pandemic influences personal exposures.

The objective of the study is to quantify personal exposures and to understand the personal cloud effects of CO_2, individual VOCs, particles, and endotoxin in a real office environment during the COVID-19 pandemic when office settings and occupant behaviors have been altered. This study also intends to investigate interpersonal differences, factors driving the personal cloud effect, and representativeness of room sampling for personal exposure estimation. We performed measurements of endotoxin, individual VOCs, CO_2, and PM_{10} at personal sampling sites of three participants and four stationary sampling sites inside a naturally ventilated office to characterize personal exposure, the magnitude of personal air pollution clouds, and the relationship between indoor stationary measurements and personal exposures. We also conducted a semi-controlled experiment for one participant to investigate the influence on the personal PM_{10} cloud of four normal daily activities that have the potential to impact human emissions of particles. This study is the first to investigate concentrations of the four air pollutants at both personal and room stationary sites in office environments during the COVID-19 pandemic. The results of this study are of potential use for improved personal exposure assessment in offices and for improved indoor environment control to mitigate inhalation exposures during and after the COVID-19 pandemic.

2. Materials and methods

2.1. Study site

The office involved in the field test was located on the second floor of an office building in Switzerland. The building previously served as a factory site renovated in 2015. The office had an area of 42.3 m^2 and an inner height of 2.8 m. The office was designed to be occupied by a maximum of six people. During the COVID-19 pandemic, the office followed the "half occupancy" regulation to have three occupants (P1, P2, and P3) regularly working inside. The layout of the office is shown in Fig. 1. The office was furnished with a sofa, a coffee table, four desks and chairs, and six cabinets. A set of water-supplied radiant panels installed on the ceiling provided heating and cooling for the office. A thermostat was equipped on the east wall to automatically control the office temperature, normally set at 21 °C. The office was naturally ventilated through two windows embedded in the north wall. The office was located in the inner zone of the building without direct connection with the outdoors, and thus the air exchange occurred between the office and the atrium space of the building.

2.2. Experimental design and setup

The field experiment ran continuously for two weeks in November 2020 during the COVID-19 pandemic. The first week of standard office work aimed to investigate the daily nature of personal cloud and the potential disparities among occupants. The second week, which was the replicate of the first week with the exception of one participant (P1) who performed semi-controlled experiments, aimed to probe the influence of four factors on personal PM_{10} cloud: 1) wearing clothing that was previously worn during office working; 2) applying body cream; 3) using hand sanitizer; and 4) wearing clothing that was worn during home cooking. Relative to clean clothing, wearing worn clothing is expected to resuspend more particles from clothing surfaces [79–82] and thus elevate PM_{10} concentration in the breathing zone. Similarly, cooking activities can generate small particles [37,39] that could be deposited on clothing and subsequently resuspended. On the contrary, applying body cream is associated with lower PM_{10} mass emissions from humans [83] and is thus presumed to reduce the PM_{10} gradient in the peri-human microenvironment. Using hand sanitizer can inactivate microbes on

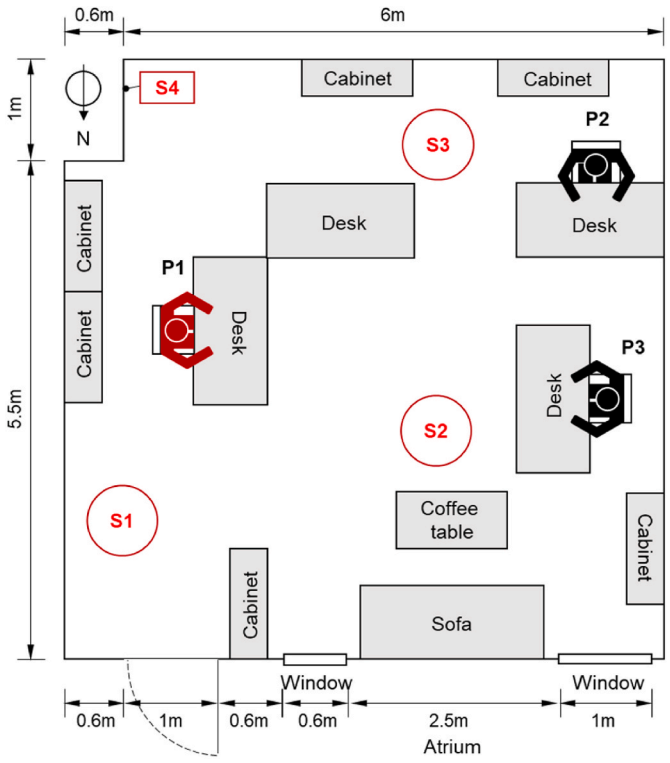

Fig. 1. The layout of the office. The office was regularly occupied by three occupants (P1, P2, and P3) during the COVID-19 pandemic. There were four stationary sites for sampling indoor air pollutants: S1, S2, S3, and S4, of which S4 was the location of the room thermostat. Personal exposure was sampled at breathing zones of P1, P2, and P3. The atrium measurement station was located 10 cm outside the window. P1 was involved in the semi-controlled experiment in the second week. Geometrical data were annotated in the layout. In addition, the sizes of main furniture were as follows (L × W × H, m): desk (1.6 × 0.8 × 0.8), cabinet (1.2 × 0.4 × 1.1), coffee table (1.0 × 0.6 × 0.5), and sofa (2.0 × 0.8 × 0.8).

human skin [84] (important constituents of human-released particles [52,85]), and therefore alter personal PM_{10} cloud. The schedule of the two-week field tests and details of the semi-controlled experiment is presented in Table S1.

We measured airborne concentrations of endotoxin, VOCs, CO_2, and PM_{10} at one outdoor site, four stationary sites in the office, and three personal sites. The indoor measurement locations are shown in Fig. 1. The room sampling stations (S1 – S4) were positioned over a metallic stand with a height of 1–1.2 m. To investigate the spatial air pollutant distribution, the principle of setting up the room stationary sampling points was to evenly distribute them inside the room space without disturbing the walking path of the participants. Specifically, S4 was located near the thermostat of the room to mimic the situation of integrating air quality sensors with the existing thermostat. The stations were placed at least 1.5 m distant from each participant to avoid the impact of human emissions and activities on the stationary sampling. Previous studies showed that the convective boundary layer of a seated person has a thickness of 0.4–0.5 m relative to a human body [86–88]. Therefore, the personal sampling sites were all within 0.5 m from the participants as an attempt to capture air pollutant concentrations in the peri-human microenvironment, as illustrated in Fig. S1. Specifically, endotoxin and VOC samplers were attached to participants' clothing and were thus more easily influenced by the human thermal boundary layer. To avoid the influence of expiratory flows and participants' talking, personal CO_2 and PM_{10} were monitored at a desk station within 0.5 m from the participants, which is in line with a previous study [69].

During the first week, at both personal and room stationary sites, we collected endotoxin samples using an impactor with size cutoff of 10 μm (Model 200, SKC Inc., UK) and an air pump (AirCheck TOUCH, SKC Inc., UK) working at 4 L/min. The polycarbonate filter within the impactor had 37 mm diameter with 0.8 μm pore size (SKC Inc., UK). The impactors were cleaned before each sampling to avoid carryover contamination. We replaced a new filter every day in the impactor to avoid any loss due to desiccation and also to avoid contamination through settling. At the personal sites, we asked the participants to run the sampling pumps only when they were seated at the desk. To collect indoor and outdoor endotoxins during the occupied period, the pumps were turned on when the first participant arrived at the office and turned off when the office became unoccupied. After sampling, the collected samples were kept in an airtight polyethylene zip lock bag and stored in a refrigerator at −20 °C. At the end of the sampling week, all the filters from a single impactor were bundled into one sample due to the low biomass concentration. In addition to the sampling during the occupied period, we also collected samples overnight when the office was unoccupied to elucidate the influence of occupancy periods on indoor endotoxin levels. The VOCs were collected using passive sampling badges (TOXpro SA, Switzerland) compliant with ISO 16017–2 [89] and ISO 16000-4 standards [90], which consisted of one VOCs passive sampler (carbon molecular sieve) and one aldehyde passive sampler (2, 4-dinitrophenylhydrazine impregnated silica gel). Similar to endotoxin sampling, participants were asked to open the caps of the samplers at personal sampling sites only when they were working at the desks. The samplers at the room stationary sites and the outdoor station were closed when the office was unoccupied. Both endotoxin and VOC samples were analyzed offline in laboratories after the field experiment, as described in Section 2.3.

The CO_2 levels at personal and room stationary sites were continuously measured by a portable sensor (MX1102A data logger, HOBO, US) with an accuracy of ±50 ppm at 1-min intervals. Time-resolved PM_{10} number concentrations at the personal site of P1 and the room station S4 were recorded using a Mini Wide-Range Aerosol Spectrometer (MiniWRAS, Grimm Aerosol, DE) with manufacturer-specified accuracy of ±3%. At other sampling sites, the PM_{10} levels were measured by optical particle counters (Model 804, MetOne Instruments Inc., US; accuracy: ±10%).

In addition to measuring air pollutant concentrations, we also recorded the occupancy status inside the office. We put an infrared occupancy data logger (UX90, HOBO, US) beneath the desk of each participant to determine the participants' presence at the desk. We also asked the participants to track their activities during the working hours using an application (timetrack.io) so that we could cross-check the occupancy activities in the studied office. According to the recorded activity track, participants were mostly (>80% of the time) seated in their working stations during the experiment. Specifically, we asked the participants to record when they used hand sanitizers. Participant P1 was also in charge of using the application to record events when there was an obvious change of the office environment, such as window/door opening status or change of occupancy.

2.3. Sample and data analysis

Endotoxin concentration in the collected samples was analyzed by means of a chromogenic endotoxin assay kit (Genscript, US; detection limit: 0.01 EU/mL). The analyzing materials were sterilized at 180 °C overnight to avoid endotoxin contamination. Particles collected on the filters were extracted in endotoxin-free water with 0.05% tween-20 in pyrogen-free centrifuge tubes: filters were racked at 37 °C for 10 min, followed by centrifugation of 1000 g for 5 min. The extracts were added to an endotoxin-free glass tube and the concentration was measured at 37 °C [91]. The level of endotoxin was determined based on an enzyme-substrate reaction followed by colorimetric analysis using a spectrophotometer at 545 nm wavelength. Endotoxin standards were prepared with Limulus amebocyte lysate (LAL) assay water. Endotoxin

concentration was normalized by sampled air volume and reported in units per cubic meter of air (EU/m^3). All the samples were analyzed in duplicates. The limit of quantification of the analyzing method was 0.01 EU, corresponding to an endotoxin level of 0.001 EU/m^3, considering the average sampling air volume of ~10 m^3 in this study.

The samplers for VOCs and aldehyde were returned to the laboratory under ISO 17025 [92] accreditation scheme (Advanced Chemical Sensors Co. Ltd, Florida, US) and analyzed after solvent desorption. The VOCs were analyzed by gas chromatography (GC) with a mass selective (MS) detector for identification and quantification (GC-MS, Shimadzu Model GC/QP-2010). TVOC was identified as the total amount of compounds detected in the VOCs passive sampler, and the concentration was quantified as toluene equivalent. The level of aldehyde, including formaldehyde, acetaldehyde, acrolein, propionaldehyde, butyraldehyde, benzaldehyde, glutaraldehyde, and hexaldehyde, was analyzed by high-performance liquid chromatography (HPLC) with UV detection (Waters Alliance 2695 Separation Module, Waters XBridge). The measurement accuracy of the sampling and analysis of VOCs and aldehydes was within 25%, with a limit of quantification (LOQ) ranging from 0.2 to 0.3 μg/m^3. Details about analyzing procedure of the VOC and aldehyde samples can be found in the recent study [7].

Size-segregated particle data collected from the spectrometer (22 log-even distributed size bins from 0.25 to 9.4 μm) and OPCs (three size bins: 0.3–1 μm, 1–2.5 μm, and 2.5–10 μm) were converted from particle number to PM_{10} mass concentration. We assumed that the mass-weighted distribution, density (1 g/cm^3) and shape of the particles (spherical) were constant across the size ranges [93]. After the conversion, the PM_{10} mass data were adjusted with a correction factor obtained from our recent PM_{10} measurements in Swiss offices using standard filter sampling and analyzing methods for PM_{10} mass concentrations [69].

The personal cloud magnitude was calculated by subtracting the average air pollutant concentrations measured at room stationary sites from that at personal sites when participants worked at desks. The magnitudes of personal endotoxin and VOC clouds were obtained as averages across the first week, whereas CO_2 and PM_{10} personal clouds had time-resolved profiles owing to real-time measurements during the two weeks.

For endotoxin and VOCs, the personal cloud was considered significant if the concentration measured at personal sites fell out of the range of mean ± 3 × standard deviation (mean ± 3SD) from the room sampling stations. For CO_2 and PM_{10}, due to the large dataset, we applied a one-sample *t*-test to examine the significance of their personal cloud magnitudes. In addition, to investigate the difference among participants for personal CO_2 and PM_{10} clouds, we compared their magnitudes using a two-sample *t*-test ($N = 2$) and a one-way ANOVA test ($N > 2$). This also applied to examine the influence of four factors on personal PM_{10} cloud in the semi-control experiment. Furthermore, we obtained the R^2 (coefficient of determination) between each personal and room stationary measurements for CO_2 and PM_{10}, in order to quantify the representativeness of indoor sampling stations for capturing the dynamic personal exposure.

2.4. Quality control and assurance

All the instruments were calibrated by their manufacturers within three months before the field test. Prior to the study, we performed a flow check and a zero check for the particle spectrometer and OPCs. The flow rate of the sampling air pump for endotoxin was calibrated with a flow meter (Defender 520, Mesa Labs Inc.). We kept a field blank filter without sampling in the first week to have the background level of endotoxin. The CO_2 monitors were calibrated with outdoor CO_2 levels as recommended by the manufacturer.

We conducted side-by-side tests for CO_2 and PM_{10} measurements to correct the difference among devices. The instruments were placed in an 0.75 m^3 environmental chamber with a mixing fan to ensure uniform air distribution. Inside the chamber, they were exposed to the same levels of airborne particles and CO_2 in order to derive adjustment factors of their performance. The configuration of PM_{10} measurements during calibration (the length and angle of sampling tubing) was kept the same as that in the field test so that the deposition losses of particles can be considered in the adjustment factors.

3. Results and discussion

3.1. Exposures and personal endotoxin clouds

The weeklong average endotoxin level measured at room stations in the occupied office was 0.09 ± 0.01 EU/m^3, shown in Fig. 2, which was in the lower range of values reported in schools and offices (0.07–9.30 EU/m^3) [94] and far below the recommended occupational exposure limit for airborne endotoxin at 90 EU/m^3 [95]. This concentration was considerably higher than that detected in the unoccupied office (0.02 EU/m^3), and also in the atrium (0.06 EU/m^3), which was comparable to outdoor levels (0.001–2.6 EU/m^3) [96]. It suggests that the participants and their activities contributed to the indoor endotoxin levels. Although the participants stayed in the same office, endotoxin concentrations at their personal sampling sites substantially differed. P2 had the highest exposure to endotoxin at 0.15 EU/m^3, which was 4 and 15 times higher than P3 (0.04 EU/m^3) and P1 (0.01 EU/m^3), respectively. The endotoxin exposure levels were strongly associated with the number of times of using hand sanitizer (Fig. 2). Alcohol-based sanitizers can kill microbes colonized on human skin by destroying their cell membranes [84], which could lead to their release of endotoxin into the human breathing zone. This suggests that inhalation exposures to endotoxins in offices could be influenced by the use of hand sanitizers. The difference between room stationary and personal endotoxin levels indicates that the room average overestimated the exposure to endotoxin for P1 and P3 but underestimated for P2. Previous measurement campaign in schools [97] also reported inconsistent relationships among personal, indoor, and outdoor endotoxin levels – Depending on individuals and school

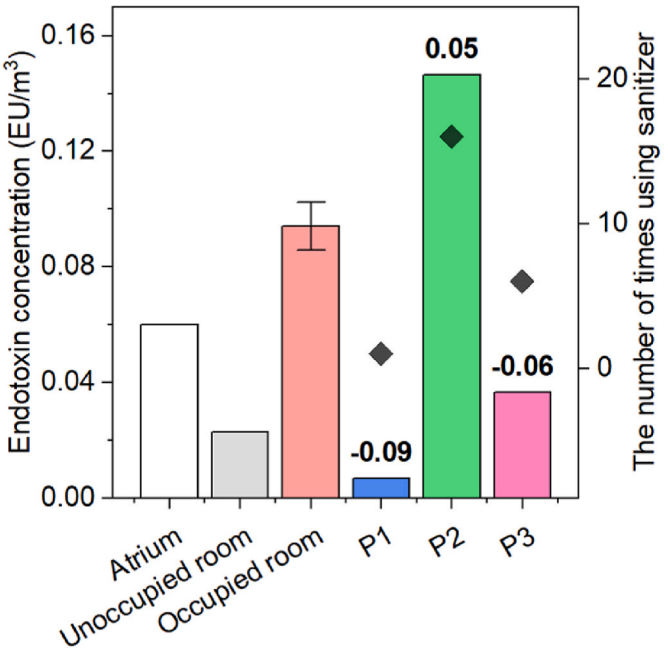

Fig. 2. Endotoxin concentrations detected at the atrium, indoor (unoccupied and occupied), and personal sites (P1, P2, and P3). Numerical labels on the top of P1, P2, and P3 bars indicate magnitudes of individual personal endotoxin clouds (difference between personal and room average levels). Diamonds represent the number of times using sanitizer for each participant during the sampling period in the first week (right axis). The error bar of the occupied room represents the standard deviation of room stationary samples.

locations, personal endotoxin exposure could be either higher or lower than indoor and outdoor concentrations. Therefore, to assess occupants' exposure to endotoxin, personal sampling is recommended.

3.2. Exposures and personal VOC clouds

We detected an overall of 13 compounds using the passive sampling kits at the stationary and personal sites in the office, as listed in Table 1. The indoor concentrations of most compounds were higher than the atrium, except for glutaraldehyde, demonstrating the contribution of indoor sources to the VOC level buildup in the office. Ethyl alcohol showed the highest level (1379 ± 115 μg/m^3) among all the detected compounds, followed by isopropyl alcohol (113 ± 10 μg/m^3). Such levels were 16 times and 2 times higher than the concentrations reported in a recent Swiss office study before the pandemic, respectively [69]. These two alcohols are commonly found in hand sanitizers. The concentrations of formaldehyde (58 ± 5 μg/m^3) and alpha-pinene (58 ± 4 μg/m^3) were found to be substantially higher in this study relative to other office measurements [26,29,69]. This may be owing to the fact that both the interior and exterior walls of the studied office and the adjacent rooms were made of wooden boards that off-gas formaldehyde and alpha-pinene. Formaldehyde concentration was below the maximum indoor recommended limit from the World Health Organization (WHO) [98], in France [99] of 100 μg/m^3 and in Switzerland of 125 μg/m^3 [100]. However, the concentration exceeded the acute exposure limit value of 55 μg/m^3 and also went far beyond the 8-h and chronic exposure threshold of 9 μg/m^3 proposed by the Office of Environment Health Hazard Assessment (OEHHA, US) [101]. Exposure to such levels of formaldehyde can lead to acute eye irritation and chronic respiratory discomfort [102]. The indoor average TVOC concentration (231 ± 20 μg/m^3) was in the range of that reported in office measurements in literature [69]. The room TVOC level was far below the recommended limit of 1000 μg/m^3 in Switzerland [100] and within the range of the lower and upper limits from Germany (200 and 300 μg/m^3, respectively) [103]. Other compounds, such as toluene, acetone, and acetaldehyde, generally exhibited concentrations similar to that detected in other office measurements and were below the exposure threshold limits [98,103,104].

The VOC levels at personal sites deviated from the room stationary sites. Personal exposure of P2 and P3 to TVOC (359 and 422 μg/m^3, respectively) exceeded the upper limit of 300 μg/m^3, although the room-average levels remained below. Out of 13 detected VOCs, 10 compounds showed significant differences between personal and stationary sampling sites for at least one participant. Most of the compounds had significantly higher concentrations at personal sites relative to the room average, evidenced by their positive personal cloud magnitudes, except for 1-butyl alcohol (Table 1). The most elevated personal cloud was found for ethyl alcohol (220–1026 μg/m^3), followed by isopropyl alcohol (21–177 μg/m^3). The personal clouds of these two alcohols were more significant for P2 and P3, relative to P1, likely because the former two used the hand sanitizers more frequently (Fig. 2).

Acetone and acetaldehyde were associated with significant personal clouds (9–20 and 6–10 μg/m^3, respectively) across all three participants. These two compounds are usually found in human breath [47,105]. A small fraction of human exhaled air could be re-inhaled and thus led to elevated concentrations in the breathing zone [106]. Moreover, acetone and acetaldehyde are also known as products from ozone-human reactions taking place around the human envelope, which could also contribute to the personal cloud effect [107]. Butyraldehyde was not detected in the stationary sampling sites but in all three personal sites, resulting in a personal butyraldehyde cloud of 3–11 μg/m^3, probably originating from human-related sources, such as from food [108]. Similarly, benzaldehyde, which is commonly used in personal care products [109], was only found in personal sites of P1 (6 μg/m^3) and P2 (11 μg/m^3). The excess concentrations of personal formaldehyde and alpha-pinene levels relative to the room average (15–26 and 14 μg/m^3, respectively) could be attributed to the proximity of participants' seats to the wooden walls, a potent source of these two compounds. The results reveal that participants' exposure to VOCs differed from each other, even though they shared the same office. Approximating by sampling at indoor stationary sites can result in a significant underestimation of personal exposure to VOCs.

3.3. Exposures and personal CO_2 clouds

Fig. 3 shows that in a typical working day, the indoor average CO_2 concentration ranged from 450 to 1000 ppm, generally below the recommended limit [110]. The indoor CO_2 level rose during occupancy hours and especially in the afternoon hours when the office door was closed. When the door was opened around 16:00, there was a sharp decrease in CO_2 concentration. The CO_2 concentration measured at the personal site generally followed the trend of the room average. When the office was unoccupied, the differences between measured CO_2 levels at P1's desk and the room stationary sites were within the instrument uncertainty. However, the personal CO_2 levels were generally higher than the room average when P1 was seated at the working station. The room average values could not capture specific periods as well as all intermittent peaks occurring in the vicinity of P1. The personal cloud was mainly caused by the exhaled CO_2 from the participant, which created a "CO_2 bubble" in the peri-human microenvironment. This result suggests that using office average CO_2 concentrations underestimates workers' personal exposures.

We found the existence of personal CO_2 clouds for all three participants, as demonstrated in Fig. 4a. P1 had the largest CO_2 personal cloud

Table 1
Detected VOC concentrations (μg/m^3) at stationary and personal sites in the office, and VOC personal cloud magnitudes (difference between personal and room-average levels, μg/m^3). Bolded values represent statistical significance of the personal cloud.

Compound	Stationary sites		Personal sites			Personal cloud magnitude		
	Atrium	Indoor average (SD)	P1	P2	P3	P1	P2	P3
Ethyl alcohol	1015	1379 (115)	1599	2312	2404	220	**933**	**1026**
Acetone	21	26 (3)	35	41	46	**9**	**15**	**20**
Isopropyl alcohol	88	113 (10)	134	198	290	21	**85**	**177**
Cyclopentane	0	4 (3)	0	0	8	−4	−4	4
1-Butyl alcohol	0	16 (1)	19	0	0	3	**−16**	**−16**
Toluene	7	8 (0)	8	7	9	0	−1	**1**
Alpha-pinene	50	58 (4)	63	54	72	5	−4	**14**
Formaldehyde	54	58 (5)	74	66	84	**15**	7	**26**
Acetaldehyde	18	19 (1)	25	26	28	**6**	**8**	**10**
Propionaldehyde	5	1 (2)	5	6	7	3	5	5
Butyraldehyde	3	0 (0)	3	4	11	**3**	**4**	**11**
Benzaldehyde	0	0 (0)	6	11	0	**6**	**11**	0
Glutaraldehyde	11	0 (0)	0	0	0	0	0	0
Total (as Toluene)	168	231 (20)	265	359	422	35	**128**	**192**

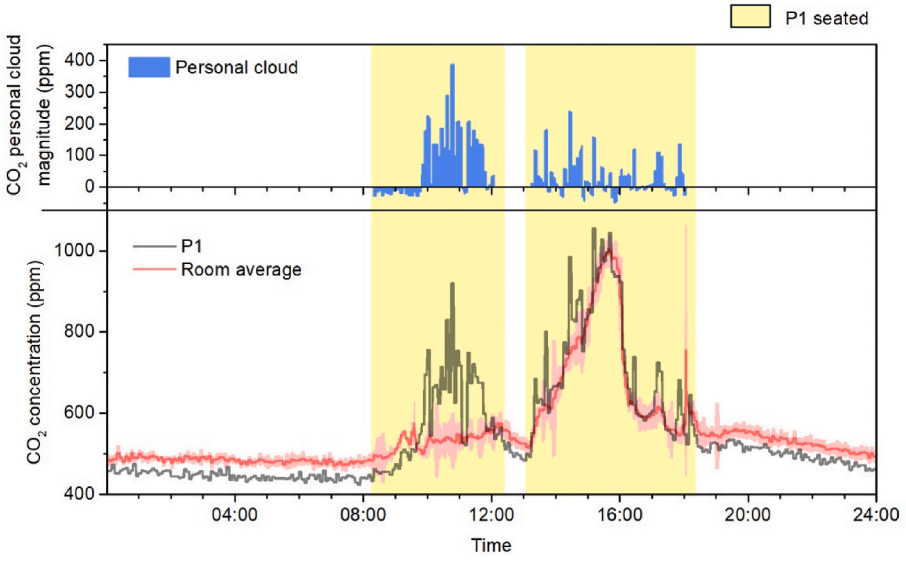

Fig. 3. Example of time-series of room average and P1 personal CO_2 concentrations, and the magnitude of CO_2 personal cloud (difference between personal and room average levels) in a day. The red band represents the standard deviation of CO_2 values at room stationary sampling sites. The yellow background indicates the period when P1 was seated in the office. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

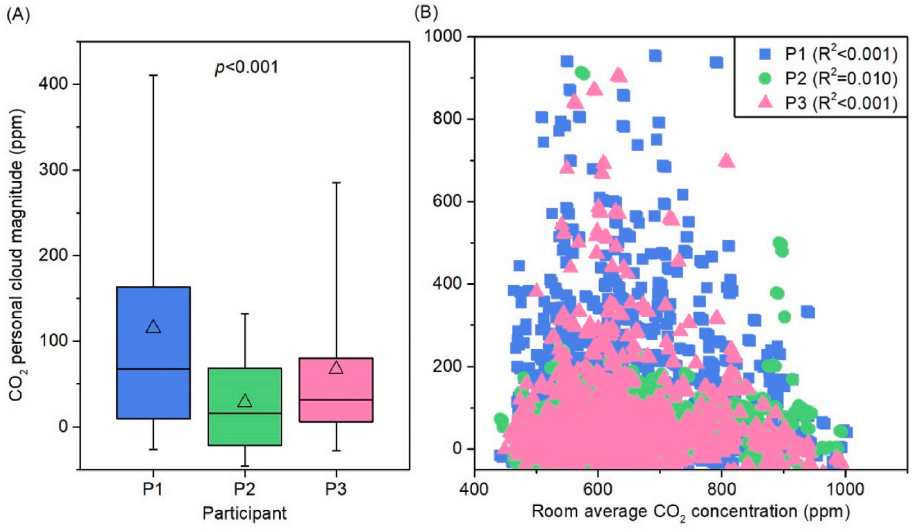

Fig. 4. CO_2 personal cloud of three participants (A) and the correlation with room average CO_2 concentration of 1-min resolution (B). The triangle in the box plot represents the mean value. The difference in CO_2 personal cloud magnitude was significant among the three participants ($p < 0.001$). R^2 represents the coefficient of determination.

magnitude (mean: 120 ppm; median: 65 ppm), followed by P3 (mean: 65 ppm; median: 36 ppm) and by P2 (mean: 32 ppm; median: 20 ppm), relative to the mean indoor CO_2 level at 630 ppm. The values are generally in line with the recent office studies, with a mean and median CO_2 personal cloud magnitude ranging from 0 to 200 ppm [69]. The difference among the three participants for their CO_2 personal cloud was statistically significant ($p < 0.001$), revealing the individual uniqueness of their peri-human microenvironment. The recorded disparity could be owing to the difference in the breathing style and local air mixing.

We further examined the dependence of CO_2 personal cloud magnitude on the room average CO_2 level (Fig. 4b). It can be clearly seen that the correlation between the CO_2 personal cloud magnitude and room average CO_2 concentration was extremely weak, with R^2 no larger than 0.01. It suggests that for the room average CO_2 level below 1000 ppm in a shared office, the CO_2 personal cloud magnitude was independent of the indoor average level, but more associated with local air around humans and their breathing style. However, as seen in Fig. 4b, when the room average CO_2 concentration approached the maximum of around 1000 ppm, the magnitude of CO_2 personal cloud generally became lower. We can presume that more occupants or closed door/window – the two potential causes of elevated indoor CO_2 level – resulted in more uniform spatial CO_2 distribution and consequently diminished CO_2 personal cloud. To check the influence of occupancy density on personal CO_2 cloud, we categorized the personal CO_2 cloud dataset of P1 based on the occupancy number in the office. It is found that the CO_2 personal cloud magnitude of P1 was significantly lower when the office was occupied by three participants, relative to only one or two participants (Fig. S2). Such an association with occupancy density echoes the finding from the literature [69] that CO_2 personal cloud was more obvious for participants working in private offices relative to shared offices.

3.4. Exposures and personal PM_{10} clouds

In a typical working day, the room average PM_{10} concentration ranged from 1 to 40 μg/m^3 (Fig. 5). Elevated PM_{10} levels occurred when the office was occupied, whereas a gradual decrease in PM_{10} concentration followed after the participants left the office. The average indoor PM_{10} concentration during the occupied period in the selected working day was around 10 μg/m^3, which was below the recommended limit of 20 μg/m^3 [98]. The value was in line with the range reported in other office air quality measurement campaigns [26,29,69]. The PM_{10} concentration at the personal sampling site of P1 shared a similar trend as the room average concentrations but in the higher range. Such an obvious disparity illustrates a substantial underestimation of personal PM_{10} exposure using the room average sampling.

Similar to P1, we also detected a discernible PM_{10} personal cloud for the other two participants, however, with distinct magnitudes, as shown in Fig. 6a. The mean indoor PM_{10} concentration was 14 μg/m^3, while the mean PM_{10} level of P1's personal site was considerably higher, leading to a personal PM_{10} cloud with a median value of 5 μg/m^3 and a mean value of 9 μg/m^3. The mean and median PM_{10} personal cloud magnitudes for P2 were both ~7 μg/m^3, whereas those for P3 were 4 and 3 μg/m^3, respectively. These values fall into the range of that reported in a previous office and chamber study [64,69], but in general lower than that found in residences [36,56]. Statistical tests showed that the difference in the PM_{10} personal cloud among the three participants was significant ($p < 0.001$). Similar to CO_2, the magnitude of PM_{10} personal cloud was also independent of room average level of PM_{10}, as evidenced by the weak correlation ($R^2 < 0.17$) shown in Fig. 6b. The PM_{10} personal cloud in the office may mainly originate from coarse particle detachment from human skin and clothing [49], and particle resuspension from desks, floor, and other human-contact surfaces due to body movement [81,111,112]. Therefore, the personal PM_{10} cloud magnitude would be mainly associated with human shedding rate, local surfaces, and body movement. Unlike the personal CO_2 cloud, the influence of occupancy density on the PM_{10} personal cloud magnitude was not significant, as illustrated in Fig. S3. It indicates that personal PM_{10} personal cloud may be mainly associated with local human-related emissions rather than air disturbance by others, which will be discussed hereinafter (Section 3.5).

We performed semi-controlled experiments for P1 in the office to investigate the influence of four factors potentially associated with human shedding on PM_{10} personal cloud magnitude. The results presented in Fig. 7, however, illustrate that these factors did not play a significant role in PM_{10} personal cloud magnitude during regular office work. Although the median magnitude of PM_{10} personal cloud was lower when the participant applied body cream or used hand sanitizer, the mean values in all scenarios were similar at 8–9 μg/m^3, without significant difference ($p = 0.94$). It suggests that none of the studied four factors strongly impacts personal exposure to PM_{10}. Their influence may be covered by local body movements, which can easily increase PM_{10} emissions in the peri-human microenvironment by multiple times due to elevated shedding rate and resuspension [64]. Nevertheless, as the experiments were semi-controlled and performed in a field study, the results need to be interpreted with caution.

3.5. Correlations between personal and stationary sampling

The aforementioned results have demonstrated an overall underestimation of personal exposure using the room-average measurement. It is, however, useful to know if stationary location can better capture personal exposure in the office. Given the large dataset, we performed a matrix of correlation analysis between personal and room stationary measured concentrations of CO_2 and PM_{10}, shown in Fig. 8.

The representativeness of stationary sampling for personal exposure to CO_2 varied in the range of 28–80%, depending on locations and participants (Fig. 8a). Among the three participants, personal CO_2 exposure of P2 was more effectively captured by the room stationary sampling, given the relatively strong correlation between the personal and stationary measurements ($R^2 = 0.70$–0.80). On the other hand, CO_2 measurement at room stations only accounted for ~30% of the variation of P1's personal exposure to CO_2. In terms of sampling locations, S2 and S3 presented an overall better prediction of personal exposure to CO_2 than the other two locations, likely because they were placed in between the three participants and close to the center of the office, where gases could be better mixed.

The representativeness of stationary sampling for personal PM_{10} exposure was worse than for CO_2, except for P2, as illustrated in Fig. 8b. Stationary samples could only explain 5–7% and 7–25% for the variation of personal exposure to PM_{10} of P1 and P3, respectively. In addition, the personal PM_{10} exposure was more easily captured by the stations closest to the participants, such as S1 for P1 and S2/S3 for P3, though with low reliability. Different from the molecular diffusivity of CO_2,

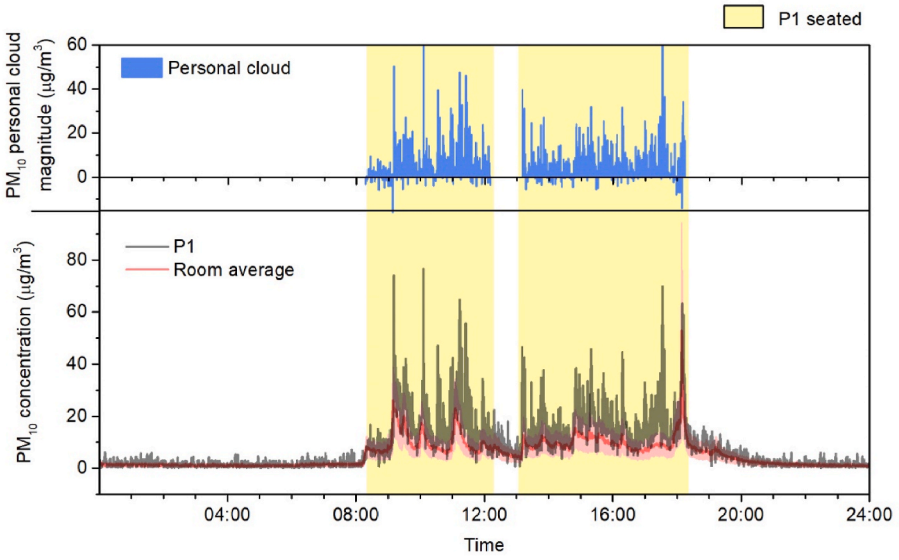

Fig. 5. Example of time-series of room average and P1 personal PM_{10} concentrations, and the magnitude of PM_{10} personal cloud (difference between personal and room average levels) in a day. The red band represents the standard deviation of PM_{10} values at room stationary sampling sites. The yellow background indicates the period when P1 was seated in the office. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

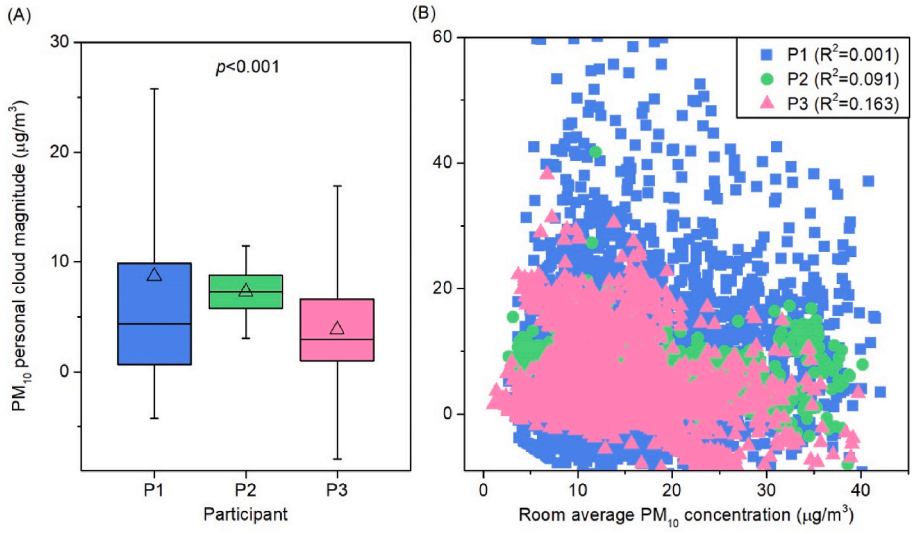

Fig. 6. PM_{10} personal cloud of three participants (A) and the correlation with room average PM_{10} concentration of 1-min resolution (B). The triangle in the box plot represents the mean value. The difference in PM_{10} personal cloud magnitude was significant among three participants ($p < 0.001$). R^2 represents the coefficient of determination.

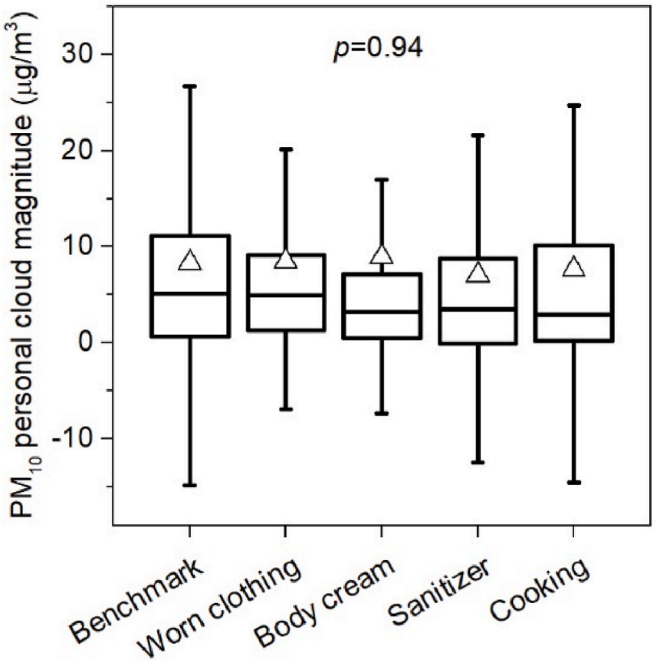

Fig. 7. Influence of four factors on PM_{10} personal cloud magnitude of one participant (P1). The factors include wearing clothing that was previously worn during office working, applying body cream before experiments, using hand sanitizer during the experiments, and wearing clothing that was worn during cooking. The triangle in the box plot represents the mean value. The difference was not significant ($p = 0.94$).

aerosol particles, especially coarse ones, have several orders of magnitude lower diffusion coefficients and are subject to gravitational settlement. Therefore, relative to CO_2, large particles emanated from humans and their activities may hardly reach distant room sampling stations but easily confined in the peri-human microenvironment. It consequently leads to elevated concentration gradient and deviated variation of PM_{10} concentrations. This also partially explains that the influence of occupancy density on personal cloud magnitude was significant for CO_2 but not for PM_{10} (Figs. S2 and S3).

The results suggest that the reliability of room stationary measurements representing personal exposure is strongly dependent on individuals and their relative location to the sampling stations, which is in line with a previous chamber study [34]. It is worth noting that in the studied office, if air quality sensors were incorporated with the wall-mounted thermostat (location S4), they would largely misinterpret personal exposure to air pollutants, based on the worst correlation between measurements at personal sites and the S4 location, for both CO_2 and PM_{10}.

3.6. Study implications and limitations

Relative to stationary air quality monitors located at remote ambient sites, indoor environments have been in the center of focus towards improved exposure assessment. To further refine exposure assessment, we need to take the indoor spatial and temporal variations of air pollutant concentrations into consideration. Although previous chamber studies have indicated that the use of mixing fans and people walking indoors can diminish or even eliminate personal CO_2 and PM_{10} clouds [63,64], these two conditions are not commonly encountered in typical offices. The study results demonstrate that substantial personal air pollution clouds exist in the office environment. Therefore, we can expect an underestimation of exposures by using room stationary sampling sites.

On the other hand, there is value in exploring scenarios in which indoor stationary sampling sites have the potential to approximate personal exposure. This requires a comprehensive understanding of interconnected factors such as absolute and relative locations of sensors and occupants, and the influence of occupants and ventilation on air pollutant dispersion. Our results reveal that the personal cloud magnitude is strongly dependent on individual occupants about their emissions and the local microenvironment. Hence, a deeper understanding of human emissions of air pollutants and their influence on peri-human microenvironments is warranted. The COVID-19 pandemic has altered office settings and occupant behaviors to some extent. Reduced occupancy density has the potential to introduce higher personal CO_2 cloud in the office environment and thus brings challenge to approximating personal CO_2 exposure with indoor stationary measurements. In addition, our results demonstrate that the use of hand sanitizers is strongly associated with elevated exposure to endotoxin and alcohol-based VOCs, which merits closer attention to diminish these unintended

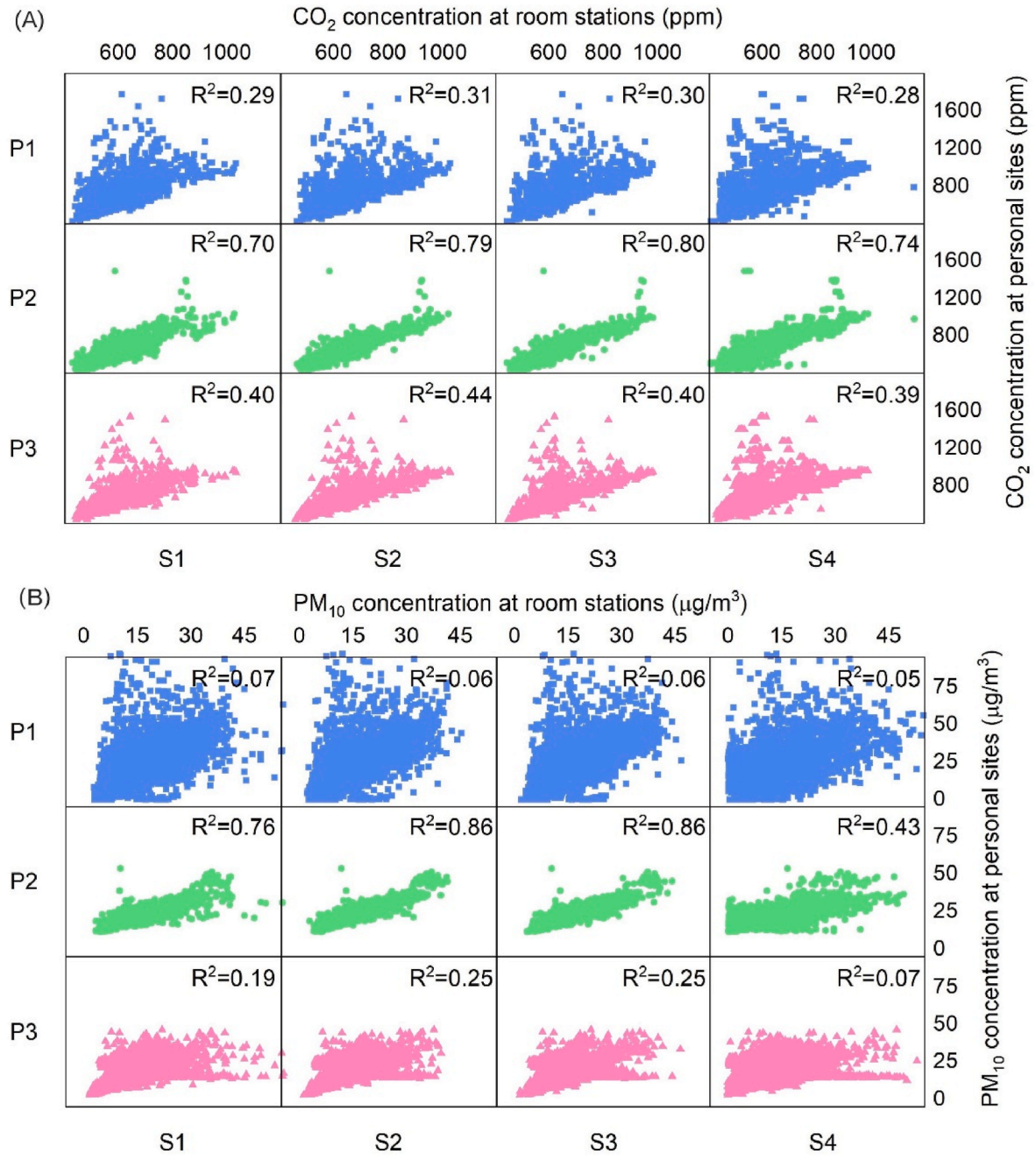

Fig. 8. Scatter plot of (A) CO_2 concentrations and (B) PM_{10} concentrations measured at personal sites (P1, P2, and P3) and room stationary sites (S1, S2, S3, and S4). Refer to Fig. 1 for specific locations of the personal and room stationary sites in the office. R^2 represents the coefficient of determination.

effects. Occupants can consider to regularly wash their hands to avoid using hand sanitizer too often. They may also put their hands distant from their breathing zone for a while after using hand sanitizer to mitigate the exposure to alcohol-based VOCs. However, due to lack of real-time monitoring of endotoxin and VOCs in participants' breathing zone, it was unclear how long the emissions would last. Future studies should experimentally test the emission characteristics of endotoxin and VOCs from hand sanitizer application. In addition, a more comprehensive health risk analysis is warranted to balance the mitigation potential of using hand sanitizer during a pandemic and exposure to unintended emission products. Finally, this study focused on a naturally ventilated office, where there was a large variation in air change rates (see Section S1 and Table S2). In mechanically ventilated offices, although the personal cloud magnitude may differ depending on the relative location of individuals to air diffusers, the existence of personal cloud effect is still expected, as it has been revealed in chamber studies with mechanical ventilation [34,63,64].

In interpreting the study results, several limitations should be acknowledged. The experiments were conducted across two weeks (one week for endotoxin and VOCs) in the transition season from autumn to winter. The short-term measurements may not be representative of the yearlong exposures, especially considering seasonal variations of air pollution levels in offices [29]. Another limitation is that the 5-day passive sampling was not able to capture all traceable VOCs and thus led to a small number of detected compounds. In addition, the offline quantification of VOCs and endotoxin cannot provide time-resolved data, so the variation of the personal cloud cannot be captured. Furthermore, personal cloud is defined by the concentration difference

between the breathing zone and the room average. However, the non-uniformity of airflow distribution in the peri-human microenvironment brings uncertainties and challenges in determining concentrations in the "breathing zone" and measuring the exact level of air pollutants in the inhaled air [65,87,88]. Licina et al. [113] revealed that measurements at a 0.4–0.5 m distance from a manikin's mouth generally underestimate the exact inhaled air pollutant level, which depends on the location of pollutant sources, room air temperature, table positioning, and body inclination. Hence, the personal cloud magnitudes reported in this study may be seen as a lower-bound approximation for air pollutants associated with human respiratory and dermal emissions. Future studies should explore a degree to which PM_{10} and CO_2 measurements on the working desks can accurately represent inhalation exposures [63,114]. Finally, in the semi-controlled experiments during week 2, we manipulated P1's clothing and activity, whereas the body movement, ventilation and other participants' activities were not regulated. Therefore, the results need to be interpreted with caution. A fully controlled chamber study can more effectively elucidate the influence of these factors on personal clouds. Despite these limitations, our study has provided a valuable dataset of personal exposure and office air quality for future studies and has highlighted the need for considering the personal cloud effect in offices.

4. Conclusions

We performed a two-week field experiment in a naturally ventilated office during the COVID-19 pandemic to assess occupants' personal exposure to multiple air pollutants and to understand factors that contribute to the personal cloud effect. Results showed that personal-level (in the close vicinity of occupants) concentrations were significantly higher than those measured at room stationary sampling sites for the majority of air pollutants. The mean personal cloud magnitude ranged within 35–192 $\mu g/m^3$, 32–120 ppm, and 4–9 $\mu g/m^3$ for TVOC, CO_2, and PM_{10}, respectively. During the pandemic, the use of hand sanitizer was associated with elevated personal clouds of endotoxin (0–0.05 EU/m^3) and alcohol-based VOCs. Although the three participants shared the same office, their personal air pollution clouds were mutually distinct. The representativeness of room stationary sampling for capturing dynamic personal exposure was low – 28% for CO_2 and 5% for PM_{10}. The findings of our study highlight the necessity for considering how inhalation exposures are influenced by spatially dependent indoor emissions. Further efforts are needed to probe the optimal locations of indoor stationary measurements that can accurately represent inhalation exposures. Such efforts would support a refined assessment of exposure conditions and associated health risks.

CRediT authorship contribution statement

Shen Yang: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Akila Muthalagu:** Writing – review & editing, Methodology, Formal analysis. **Viviana González Serrano:** Writing – review & editing, Methodology. **Dusan Licina:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.buildenv.2023.110280.

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