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Emission rates and the personal cloud effect associated with particle release from the perihuman environment

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Abstract

Inhalation exposure to elevated particulate matter levels is correlated with deleterious health and well-being outcomes. Despite growing evidence that identifies humans as sources of coarse airborne particles, the extent to which personal exposures are influenced by particle releases near occupants is unknown. In a controlled chamber, we monitored airborne total particle levels with high temporal and particle-size resolution for a range of simulated occupant activities. We also sampled directly from the subject's breathing zone to characterize exposures. A material-balance model showed that a sitting occupant released 8 million particles/h in the diameter range 1-10 µm. Elevated emissions were associated with increased intensity of upper body movements and with walking. Emissions were correlated with exposure, but not linearly. The personal PM_{10} exposure increment above the room-average levels was 1.6-13 µg/m^3 during sitting, owing to spatial heterogeneity of particulate matter concentrations, a feature that was absent during walking. The personal cloud was more discernible among larger particles, as would be expected for shedding from skin and clothing. Manipulating papers and clothing fabric was a strong source of airborne particles. An increase in personal exposure was observed owing to particle mass exchange associated with a second room occupant.

Keywords

Human emissions, Personal exposure, Activity type, Particle size distribution, Cross-contamination, Particle sources

Practical implications

Better understanding of the nature, scope and significance of human personal clouds is valuable for enhanced prediction and control of personal exposure, which is important in relation to how indoor air quality influences human health. The results of this work are of potential use in indoor air quality models and for improved ventilation design. The work also contributes to a better understanding of human occupants as sources of airborne particles, which could strengthen the design and interpretation of future air pollution exposure studies.

Introduction

Inhalation exposure to particulate matter is associated with important health concerns. Yet major challenges remain to accurately characterize exposure. A potentially important factor is spatial variability of indoor particle concentrations. When mixing is systematically incomplete, substantial concentration gradients may persist in proximity to emission sources. Depending on the spatial relationships among monitor location, particle sources, and the human breathing zone, errors in assessing inhalation exposure may result.[1-3]

The term "personal cloud" is used here to mean an excess of particle mass concentration in the vicinity of a person relative to room-average levels. In a seminal study of personal exposure to airborne particulate matter, Özkaynak et al.[4] identified the personal cloud as a potentially important factor. They wrote: "Population-weighted daytime personal PM_{10} exposures averaged 150 ± 9 µg/m^3, compared with concurrent indoor and outdoor concentrations of 95 ± 6 µg/m^3. This [difference] suggested the existence of excess mass near the person, a 'personal cloud' that appeared related to personal activities." The results of that study suggest that the personal cloud might have contributed ~ 50 µg/m^3 to the average PM_{10}

exposure of subjects in their study, a clear indication of the potential significance of the issue. Yet, many questions remain unanswered about the nature and significance of the personal cloud.

In principle, excess particle mass in the perihuman zone may arise from exogenous and endogenous air pollution sources that occur as a result of personal activities. Exogenous sources involve activities such as cooking, vacuuming, making a bed, secondary exposure from a nearby smoker, resuspension from flooring attributable to walking, or from other activities that generate localized emissions of airborne particulate matter.[1,5-7] In a study by Ferro et al.,[7] the influence of exogenous sources, expressed through the ratio of inhaled to room-average concentrations of $PM_{2.5}$, varied from 1.2 for vacuuming to 4.2 when folding a blanket.

Endogenous sources include human skin and clothing that can generate airborne skin fragments, shed previously deposited particulate matter, and create airborne particles through frictional interaction of clothing fibres. Rodes et al.[1] drew attention to emissions from the human body as an important confounding source of the personal cloud and stressed the need for data on their influence. Yet, a quarter century later, particulate matter emissions from the human envelope have not been effectively quantified as constituents of the personal cloud.

Early research identified humans as important contributors to the indoor total and biological airborne particle load through their shedding of bacteria-laden skin flakes into the air.[8] The shedding rate was associated with human activity level, type of clothing and lipids on the skin surface.[9,10] Recent studies have utilized culture independent quantitative polymerase chain reaction methods to quantify microbial emissions associated with human

occupancy indoors.[11,12] Although providing useful information about overall human-associated emissions, integrated sampling approaches are unable to resolve short-term, intermittent and highly irregular processes that influence shedding rates, and that are key for developing a deeper understanding of the personal cloud. Only a few studies have quantified human emissions of total and biological airborne particles with a high temporal and particle-size resolution.[13-15] You et al.[13] identified a positive correlation between the vigour of human activity and the emission rates of total airborne particles. Bhangar et al.[14,15] used a laser-induced fluorescence-based ultraviolet aerodynamic particle sizer (UV-APS) to quantify human emission rates of fluorescent biological aerosol particles and reported that human skin and clothes are important sources indoors.

To quantify the effect on the personal cloud of shedding from and near the human envelope, we undertook a series of experiments that allowed assessment of separate contributions of the body envelope from any exogenous sources of particulate matter. To assess the relative importance of the type and vigour of activities, human subjects undertook scripted behaviours. The collection of size-resolved, real-time data allowed for exploration of dynamic processes that are important for exposure assessment. By measuring particle levels at multiple locations — in the breathing zone, at stationary locations in the room and in the exhaust vent — we sought to elucidate the relationships between spatial pollution distribution and type of activity, particle size, human exposure and the personal cloud. The secondary purpose of this study was to expand the otherwise scarce body of literature with new measurement results for human emissions of size-resolved airborne particles for a set of scripted activities. The results also provide insight regarding the individual contribution of potentially important exogenous air pollution sources, such as handling paper and clothing fabrics, as well as the transmission of human-associated aerosol particles between occupants.

To our knowledge, this is the first study to document the contribution of particle release from the perihuman environment to the personal cloud effect. The results of this work are potentially useful for interpreting the health risks associated with indoor air quality, for improving accuracy in exposure assessment, and for developing improved measures to control exposure to coarse mode airborne particles, which include bioaerosols.

Methods

The definition and interpretation of the personal cloud vary in the literature. Some studies refer to the personal cloud as the particulate matter contribution from human skin and clothing to the room air (otherwise referred to as "human particle emissions").[13,16] Other studies define the personal cloud as a ratio of measurements between a personal and a stationary monitor.[7] The ratio of personal to room average particle levels is not expected to be a stable indicator of the personal cloud effect, as that ratio would be sensitive to exogenous factors that could vary strongly from one condition to another. Our study adopts the representation of the personal cloud as an additive mass-concentration increment (in $\mu g/m^3$), specifically defined as the enhancement of breathing zone concentration above the room average condition as associated with spatially varying concentration fields. We anticipate that this measure is a relatively stable outcome variable, meaning that the results presented in this work have the merit of being applicable to describe similar circumstances in other indoor environments beyond those directly tested here.

Study site

The study was conducted in a controlled environmental chamber with floor area of 21 m^2 and 2.4 m ceiling height, corresponding to an interior volume of approximately 50 m^3. The chamber floor is covered with hard vinyl tiles, compatible with the research goal of minimizing coarse particle resuspension.[17,18] The flooring was thoroughly cleaned with water prior to the start of experiments. The experimental room, which is tightly sealed, is situated inside a larger, thermally conditioned volume, which serves to protect the envelope from solar and wind effects. The internal heat sources from lighting and equipment were minimized (<100 W) and were kept constant throughout the experiments. The chamber was furnished with two tables, two laptop computers and two chairs, centrally positioned (Figure 1a).

The chamber is served by a dedicated heating, ventilation and air conditioning system that continuously delivers 100% outdoor air at a constant flow rate. To isolate the effect of human skin and clothes shedding from particulate matter in outdoor air, the supply air was drawn through a particle filter with a manufacturer-specified efficiency of ≥ 95% for 0.3 µm particles. Conditioned air, with temperature and humidity control, was supplied to the chamber through two ceiling-mounted air diffusers and exhausted through an opening in the wall at 1.8 m height (Figure 1a). For this study, the indoor dry-bulb temperature and relative humidity set points were 21 ± 1.5 ˚C and 40 ± 5%, respectively. Overall, the chamber was configured so that it would be uninfluenced by any outdoor pollution source. Consequently, the only source of variation in indoor particle levels was caused by the presence and activities of the occupant(s).

Experimental design and sampling

The effects of occupant activity on the emission rates and the personal cloud were assessed through 12 distinct two-hour experiments conducted between December 2015 and January 2016. Each treatment was replicated three times for a total of 36 experimental runs. To ensure that the indoor pollutant concentration was independent of diurnal variations, time-of-day effect was probed. As the effect was found to be insignificant, three treatments were scheduled for each day. Potential for bias and confounding were minimized through the use of a randomized block experimental design, with two isolated factors: activity type (sitting or walking) and occupancy number (one or two), as summarized in Table S1.
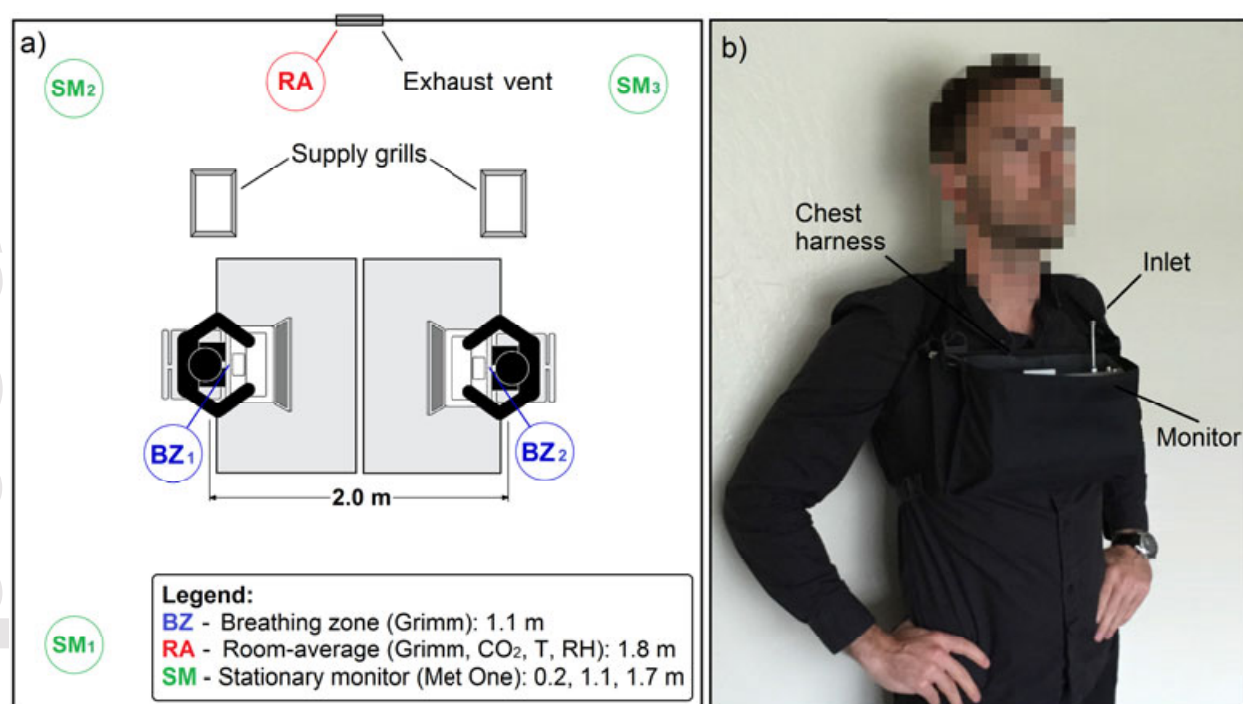

Fig. 1 (a) Environmental chamber configuration and sampling locations; and (b) breathing zone personal monitor (BZ) placement within 0.15 m from the mouth. Stationary monitors SM_1, SM_2 and SM_3 were located at the three heights, 0.2, 1.1, and 1.7 m, respectively. Note that the schematic layout (a) presents one (ID8) out of several investigated configurations, as reported in Table 1.

To limit any effect of particle resuspension from contact surfaces, the chamber floor and furniture were thoroughly cleaned with water prior to experiments. As the focus of the study was on particle shedding from the human envelope, resuspension from flooring was

also minimized through measures to limit floor dust, including any that might be tracked-in on shoes. Clean shoes used only indoors were employed during these experiments and tracked-in dust was further controlled through the use of a daily-replaced sticky doormat at the entrance of the experimental room. The chamber floor was cleaned with a sticky roller at the end of each day.

Occupant clothing consisted of black pants and long-sleeve shirt, both made of 100% cotton (Figure 1b). The material weave pattern is known to influence particle resuspension from clothing, while the clothing fiber type does not have significant influence.[13,19] Clothing conditions in our study were maintained constant throughout the treatments. Before each day's experiments, the subject clothing was laundered, tumble-dried and exposed to indoor (residential) air for 10 hours. These experiments were not designed to examine the influence of varying clothing conditions, otherwise known to be an influential factor.[13,15,19] We intend to report in a companion paper the influence of clothing coverage, color and whitening agents on emission rates of total and fluorescent biological airborne particles.

The experiments were designed to quantify human envelope emission rates and the personal cloud as a function of occupant activities. Seven activities were explored with a single occupant and three activities were tested with two occupants, as summarized in Table 1. These seven activities were included in the study matrix: sitting still, sitting with moderate movement, sitting with intensive movement, sitting with intensive movement involving arranging office papers, sitting with intensive movement involving manipulation of a clothing fabric, walking at a slow pace (80 steps/min), and walking at a brisk pace (110 steps/min). Detailed description and time of each activity is summarized in Table S2 in the supporting information. Each activity was continued for 30 min followed by an unoccupied period of 1.5

h to permit monitoring of particle concentration decay. Two supplementary experiments were designed to probe the effect of human proximity (1 and 2 m distance) and relative contribution of occupancy-associated emissions to the personal exposure and personal cloud of a receptor.

The test subjects were one female (age: 29; height: 1.72 m; weight: 62 kg) and one male (age: 29; height: 1.85 m; weight: 80 kg) who followed defined procedures prior to and throughout the experiments. The night before each experiment, subjects were asked to shower at a specific time and not to apply personal care products. On the day of experiments, subjects would put on the prepared experimental clothing (including the personal aerosol monitor), and then enter the chamber to perform scripted activities. Each activity was executed at a prescribed pace, regulated by means of a metronome. The male subject undertook all experiments that involved a single participant. A human subject's protocol was prepared and approved by the Human Subjects Committee at Lawrence Berkeley National Laboratory, where the experiments were conducted (Approval ID: 346H001 - 2SP2016).

Real-time, size-resolved aerosol spectrometers (models 11-A and 1.108, GRIMM Aerosol Technik GmbH, Ainring, Germany) were employed to measure concentrations of airborne particles in the vicinity of the mouth to approximate inhaled air (also referred to as the "breathing zone") as shown in Figure 1b, and in the exhaust vent to represent room-average concentrations. Airborne particles were resolved in 8 size groups based on optical diameter (0.3-0.5 µm, 0.5-1 µm, 1-2 µm, 2-3 µm, 3-4 µm, 4-5 µm, 5-7.5 µm, 7.5-10 µm). Concurrent indoor and outdoor CO_2 levels were measured with real-time gas analyzers (LI-COR Biosciences, Lincoln, NE, USA). The dry-bulb temperature and relative humidity monitor detected room-average values in the exhaust vent. During the experiments, we also sampled particle number concentrations with stationary optical particle counters (model Met One HHPC 6+, Beckman Coulter Life Sciences, Palatine, IL, USA) at three peripheral

locations (Figure 1a) to assess the degree of mixing of airborne particles in the room. All measurements were carried out with 1-min resolution to capture the unsteady responses to dynamically changing conditions.

Data interpretation

To estimate particle mass from measured number concentration, we assumed that particles are spherical with density of 1.0 g/cm³. As the average airborne particle density is in the range 1-2.5 g/cm [3,20] mass estimates reported here should be deemed as lower-bound values. Another assumption made was that the mass-weighted size distribution, dM/d(log d_p), is constant within each particle size group. The PM$_{10}$ mass levels were computed by summing the particle mass concentrations bigger than 0.3 µm and smaller than 10 µm. Particles smaller than 0.3 µm are considered to insignificantly impact total PM$_{10}$ mass concentration in these experiments owing to two considerations: (a) negligible intrusion of particles of outdoor origin and (b) weak contribution to total particle mass emissions as a consequence of human movement by submicron particles, as supported by the experimental results and by expectations for mechanical generation of particulate matter.

A mass-balance model (equation (1)) was applied to particle number concentrations measured in the room exhaust air to calculate the source emission rates from human activities. For this analysis, we assumed that the incompressible volume of air (V) in the room is perfectly mixed.

$$\frac{dN_{in,i}(t)}{dt} = \frac{ER_i(t)}{V} + ap_i N_{out,i}(t) - (a + k_i)dN_{in,i}(t)$$

(1)

In equation (1), $N_{in,i}(t)$ is the indoor particle number concentration in the room air in size channel i (particles per m³); $ER_i(t)$ is the per-occupant emission rate (particles/h) in

channel i; a is the air exchange rate (1/h); p_i is the particle penetration efficiency from outdoors for channel i (-); $N_{out,i}(t)$ is the particle number concentration in outdoor air in channel i (particles per m^3); and k_i is the particle deposition loss-rate coefficient for channel i (1/h). Equation (1) is applied by first multiplying all terms by the time increment, dt, and then integrating each term over the period of the experimental run (0 to T). Particle penetration from outdoors is negligible for these experiments owing to near complete removal by the filter in the supply air, as confirmed by measurement of very low indoor particle levels during unoccupied periods.

Enhanced concentrations associated with the personal cloud contribute little to the room averaged concentrations even though they may contribute substantially to occupant exposures. The error associated with applying the well-mixed assumption was probed by applying a two-compartment model (perihuman zone and rest of the room) with results presented in Table S3. Overall, the associated error was acceptably small, below 13%.

In summary, the time-averaged size-resolved emission rate during the active source period (T_s) was evaluated as shown in equation (2):

$$\overline{ER_i(T_s)} = \frac{T}{T_s}V\left[\frac{dN_{in,i}(T) - dN_{in,i}(0)}{T} + (a + k_i)\overline{N_{in,i}(T)}\right]$$

(2)

Here, the overbar indicates a time-average of the parameter for the time period from 0 to either the end of the active-emissions period (T_s) or to the end of the experimental run (T).

To evaluate the air-exchange rate (a), a 10-min pulse of carbon dioxide (CO_2) was released into the supply duct via a flow controller (model MC-10SLPM-D/5V, Alicat Scientific, Tucson, AZ, USA) delivering 10 liters per minute. An exponential decay was fit to the increment of indoor CO_2 concentration above the outdoor level for the period after injection and the rate constant was equated to the air-exchange rate, which we found to be 2.3

± 0.05 per hour (n = 5 replicates). Spot checks throughout the measurement campaign based on human-generated metabolic CO_2 levels substantiated that the ventilation rate was constant, as expected.

In preliminary experiments, we investigated the influence of variable air-exchange rates on the air speed measured at multiple locations in the chamber. The measured speed was consistently less than 0.1 m/s, indicating that near-occupant air flows would be dominated by the individual's thermal plume and movement, rather than by air flows induced by the mechanical ventilation system.

Indoor particle deposition loss-rate coefficients (k_i) were estimated for each treatment based on size-specific particle number concentration decay rates during the post-occupancy period. The value of k_i was assessed as the negative slope of the natural logarithm of $N_{in,i}$ versus decay time, minus the air-exchange rate. These results are believed to be a lower-bound estimate of k_i during occupancy period due to an excluded increment of deposition onto the human subject(s). The empirical estimates of size-resolved k_i obtained for sitting and walking activities (enclosed in Table S4) were compared with the deposition loss-rate coefficients reported by Thatcher et al.[21], as shown in Figure S1. The estimates of $ER_i(t)$ were derived with empirically determined k_i values and with those adopted from Thatcher et al.[21] to assess the degree of uncertainty owing to incomplete knowledge regarding particle deposition rates. The results agreed to within 5% for the number emission rates and to within 20% for the mass emissions. The results reported here rely on the empirically determined k_i values from the present experiments.

Metabolic CO_2 emission rates (ER_{CO_2}) were determined for each treatment based on an integral material balance expression similar to equation (2), with $k = 0$, and with the average net CO_2 level indoors represented by the difference between indoor and outdoor levels.

Data collected with calibrated aerosol spectrometers and stationary optical particle counters were corrected using adjustment factors from side-by-side tests of instrument performance (Table S5). The CO_2 instrument response was confirmed through exposure to calibration gases at 0 and 1000 ppm.

Results and Discussion

Figure 2 illustrates the effect of three types of occupant activity on time-resolved 1-min mean breathing zone and room-average PM_{10} levels. When a subject performed seated activities, inhaled concentrations were consistently above the room-average value. With the low background level in the chamber, the relative difference was as high as an order of magnitude when the subject performed stretching. Conversely, low activity periods such as computer work were characterized by a minimal increment to personal PM_{10} concentrations. The substantial concentration enhancements in the perihuman zone are attributed to weak mixing when the occupant is seated. Once detached from skin or clothing, particles progress to the surroundings at a slow pace, thus elevating the likelihood to become inhaled. The high relative differences in PM_{10} mass concentrations between the two types of seated activities are primarily attributed to exogenous particle emissions from handling papers and clothing fabrics. The activity periods that involve manipulation of the fabric are characterized with sharp intermittent peaks in the breathing zone particle concentration. This aspect will be discussed in subsequent sections along with the exposure contributions from handling paper.
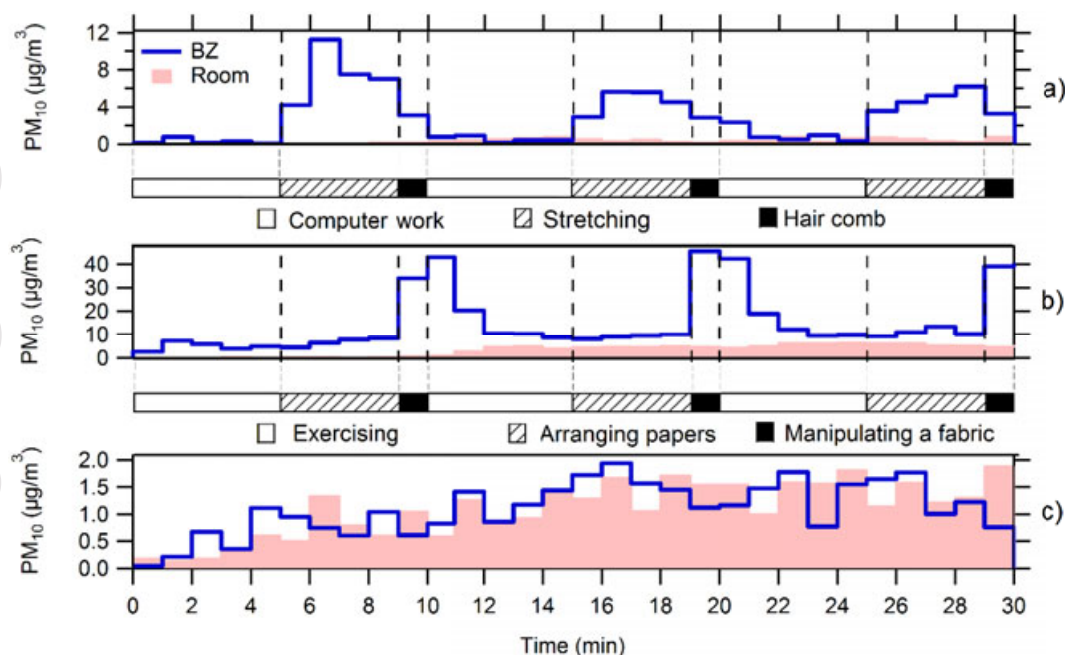
Fig. 2 Time series representation (1-min resolution) of the breathing zone (BZ) and room-average (Room) PM_{10} mass concentrations when a subject (a) performed seated moderate movements (referred to as ID = 2 in Table 1), (b) performed seated intensive movements with fabric and papers (ID = 5), and (c) walked at a pace of 110 steps/min (ID = 7). Vertical dashed lines represent transitions between specific occupancy movements, within the same activity type. Note the different vertical axis scales among the three frames.

A walking subject (Figure 2c) apparently stirred the room air well, disrupting any evident spatial gradients of particulate matter. A relatively low contribution to the room-average PM_{10} mass concentrations from walking is attributable to minimized resuspension from the floor for these experiments. In other experiments in which particle release from the floor was not minimized, resuspension was found to be responsible for up to 70% of total airborne particle emissions induced by walking.[15]

Emission rates from different occupant activities

Figure 3 compares size-resolved emission rates of total particles by count (upper frames) and mass (lower frames) when the occupant performed seated moderate movements (left frames), seated intensive movements (middle frames) and walked at a pace of 80

steps/min (right frames). As anticipated, walking was linked to higher emission rates relative to moderate sitting activity owing to the increased vigour of bodily movements. Considering all particles in the optical diameter range 0.3-10 µm, the mean emission rate for walking was 90 ± 14 million particles per h, 60% higher than the emission rate from the sitting moderate activity (see also Table 1). The estimated emission rates of particles larger than 1 µm from walking and sitting with moderate bodily movements were 20 ± 2 million particles per h and 8 ± 3 million particles per h, with corresponding estimated mass emission rates of 0.45 and 0.17 mg/h, respectively. There was no significant difference in the emission rates for the two walking intensities. When reassessed to only consider particles larger than 5 µm, the emission rates from walking (80 steps/min) and sitting (moderate movement) were 1.5 ± 0.1 million particles per h and 0.6 ± 0.4 million particles per h, respectively. These results match well the emissions (for particle diameters > 5 µm) reported by Bhangar et al.[14], summarized to be 3 ± 1 million particles per h for walking on a clean plastic sheet, and 0.7 ± 0.4 million particles per h for a sitting activity.

Table 1 summarizes human emission rates for a full set of occupant activities. Particle emissions could barely be detected when the subject minimized movement (referred to "seated still" in Table 1). This finding substantiates an expectation that human bodily movement is a dominant factor inducing particle detachment from skin and clothing. That particle emissions are detected at all under "seated, still" conditions may reflect (a) imperfectly still subject, and/or (b) emissions from respiratory flows. Although expiratory droplets are effective in transporting biological agents into a room,[22] past studies suggest that emissions associated with tidal breathing contribute little to emissions of supermicron particulate matter.[23]
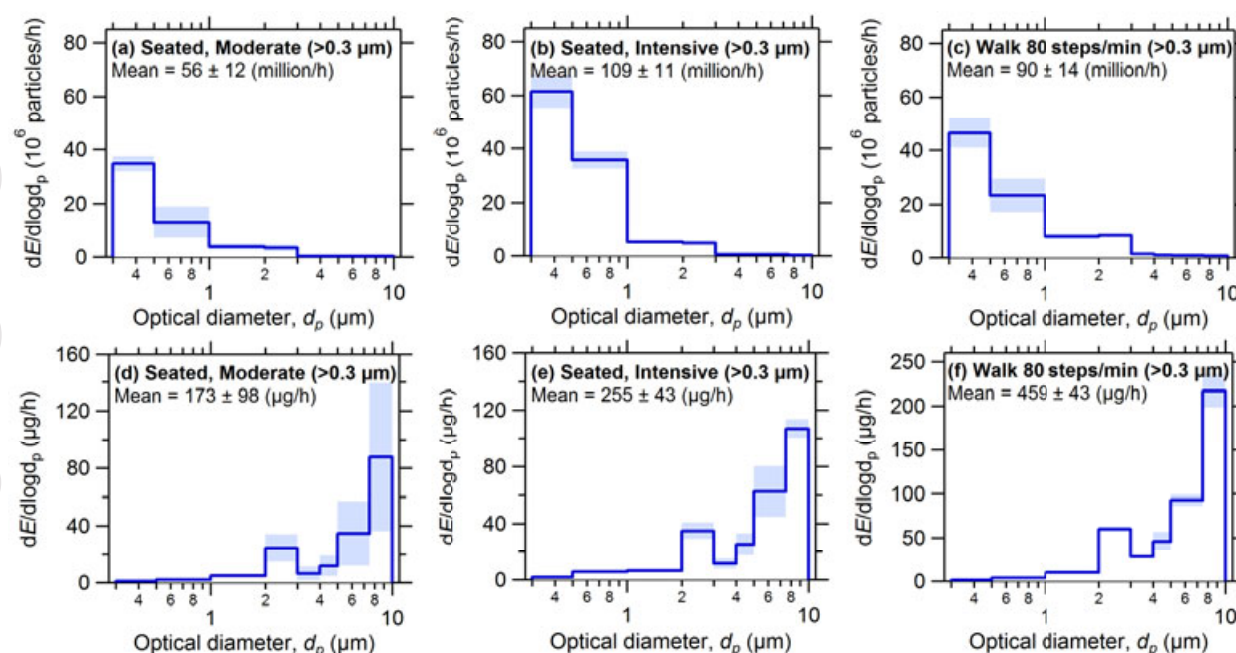
Fig. 3 Size-resolved particle emission rates by count (upper frames) and mass (lower frames) associated with one human occupant performing moderate seated movements (left frames; ID = 2), intensive seated movements (middle frames; ID=3) and walking at 80 steps/min (right frames; ID = 6) in the chamber. The mean ± standard deviation (represented by shaded area) across specified particle sizes are reported in each frame.

As a part of a quality assurance test, an ultraviolet aerodynamic particle sizer (UV-APS) (model 3314, TSI Inc., Shoreview, MN, USA) monitored number concentrations of total aerosol particles in the exhaust vent line. The results reported in Table 1 based on optical particle diameter agree well with the emission rates estimated based on aerodynamic diameter (from UV-APS); the differences range from 0% for walking at 80 steps/min to 40% for sitting with intensive movement. Details about the UV-APS measurements and the comparison of occupant emissions estimates based on the two instruments (GRIMM vs. UV-APS) are reported in Table S6.

In our studies, the seated male subject produced 37 gCO_2 per h, which increased to 58-63 gCO_2 per h for seated intensive bodily movements, and to 82-118 gCO_2 per h for walking (Table 1). These metabolic CO_2 emission rates were considerably higher than per subject average values reported by Bhangar et al.[15] for sitting (27 gCO_2 per h) and for walking (38 gCO_2 per h). The subjects in Bhangar et al. were female. Previously reported

discrepancies have been noted in metabolic CO_2 generation levels by body size, gender and

race.[24]

Table 1 Summary of the mean ± standard deviation of human emission rates of total particle number in the size range 0.3-10 µm ($ER_{>0.3}$), supermicron (>1 µm) total particle number ($ER_{>1}$), particle supermicron mass ($M_{>1}$), and carbon dioxide (ER_{CO2}) as a function of the number of occupants (Occ.) and type/intensity of activity [a].

ID [b]	Occupant activity [c]	Occ.	$ER_{>0.3}$ (10^6/h)	$ER_{>1}$ (10^6/h)	$M_{>1}$ (mg/h)	ER_{CO2} (g/h)
1	Seated, still	1	31	0.3	0.002	37
2	Seated, moderate movement	1	56 ± 12	8 ± 3	0.17 ± 0.10	59 ± 5
3	Seated, intensive movement	1	109 ± 11	12 ± 2	0.25 ± 0.04	58 ± 1
4	Seated, intensive movement with fabric	1	300 ± 48	61 ± 6	1.9 ± 0.3	65 ± 7
5	Seated, intensive movement with fabric and paper	1	370 ± 31	67 ± 1	1.9 ± 0.3	63 ± 10
6	Walk 80 steps/min	1	90 ± 14	20 ± 2	0.45 ± 0.04	82 ± 3
7	Walk 110 steps/min	1	110 ± 18	18 ± 7	0.49 ± 0.03	118 ± 10
8	Seated, moderate movement	2	53 ± 11	12 ± 3	0.26 ± 0.06	111 ± 9
9	Seated, intensive movement with fabric and paper	2	650 ± 67	147 ± 13	3.8 ± 0.3	112 ± 3
10	Walk 80 steps/min	2	180 ± 70	36 ± 15	1.1 ± 0.5	159 ± 11

[a] There were n = 3 replicates for each case; the duration of each activity was 30 minutes.
[b] Two supplementary experiments were designed to probe the effect of particle transmission from the source to the receptor at 1 and 2 m distance, referred to here as cross-contamination.
[c] Detailed description and the time-pattern of activities is summarized in Table S2 in the supporting information.

Human personal cloud: Effects of activity type and room air mixing

Figure 4 shows time-averaged, size-dependent particle concentrations in the breathing zone in comparison to the room-average concentrations for three conditions. Subject occupancy did not materially influence the mass concentration of airborne particles smaller than 2 µm, but did result in a distinct increase in mass concentrations of particles in the diameter range 2-10 µm. This particle-size range includes the dominant mode of indoor airborne bacteria,[11,12] which may be associated with scales of desquamated skin.[9,25] The walking subject contributed 1 µg/m^3 to the room particle mass concentrations averaged across the size range 0.3-10 µm.

A striking feature of Figure 4 is the comparison of breathing zone and room-average concentrations. Distinctively different outcomes are displayed for the two experiments with the subject seated as compared to the walking experiment. For the walking experiment, breathing zone concentrations were equivalent to the room-averaged value. That outcome could be a combined result of the detachment of the subject's personal convective boundary layer (also known as the "thermal plume") combined with more effective mixing of the room air induced by walking. Notably, as a consequence of the strong air mixing due to walking, the mean PM_{10} mass among three stationary monitors exhibited a low coefficient of variation (COV) of 5% (this pertains to both walking intensities examined). In the experiments entailing seated activities, the subject contributed to room-average mass concentrations at levels 30-60% of those induced by the walking subject; such differences are evidenced in the emission rates reported in Table 1. However, a notable personal cloud effect was discerned for the seated subject performing moderate and intensive bodily movements. The magnitude of the personal cloud in these cases was determined to be 2.2-2.3 µg/m^3. With less motion, as for a seated occupant compared with a walking occupant, spatial concentration gradients were more pronounced — promoting elevated concentrations in the breathing zone above the room-averaged condition. Less complete mixing in the room is exhibited in the higher COVs among the three stationary monitors for the seated subjects: 11% for intensive and 28% for moderate bodily movements. For the seated subject, sampling that was designed to characterize room-average concentrations would underestimate inhalation exposure. For conditions of these experiments, the magnitude of the underestimate for PM_{10} would be 2-3 µg/m^3.

Sufficient evidence from the literature suggests a strong link between indoor coarse particle concentrations and occupancy-associated emissions through body envelope shedding.[13,26,27] These empirical studies show that the mass rate of indoor emissions tends to

increase with particle size, at least within the range 0.3-10 µm diameter. The magnitude of
the personal cloud accordingly appeared more discernible within the larger particle-size
fractions, as displayed in frames (a) and (b) of Figure 4. Table S7 and S8 summarize the
mean size-specific particle number concentrations for a full set of activities as measured in
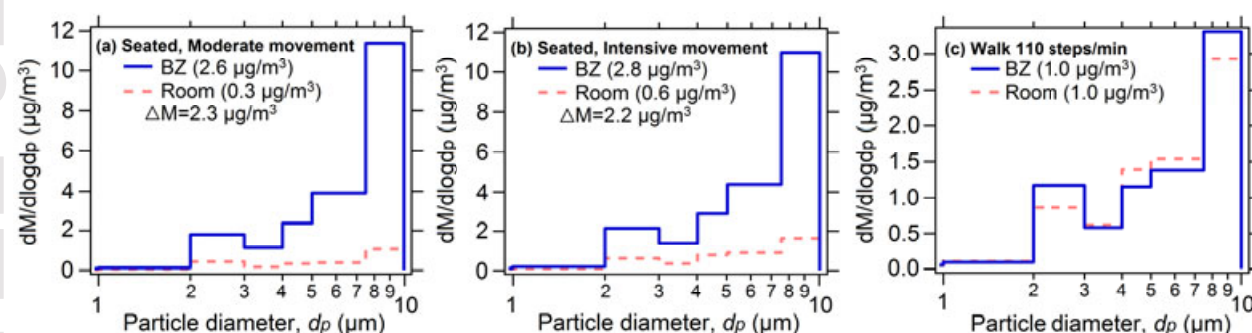the exhaust vent and in the subject breathing zone.

Fig. 4 Size distributions of the breathing zone and room-average particle mass concentrations
when the subject was a) seated with moderate movements (ID = 2), b) seated with intensive
movements (ID = 3), and c) walking at 110 steps/min (ID = 7). The personal cloud
magnitude, ΔM, represents the excess of breathing zone particle mass concentration relative
to the room-average concentrations.

The walking subject contributed more to the room-average particle mass
concentration than did the seated subject (Figure 4). Contributions from walking would likely
have been more prominent if measures had not been taken to minimize resuspension from
flooring and footwear. Notwithstanding the higher emission rates and room-average
concentrations of a walking person, inhaled concentrations of a seated subject were higher.
Also noteworthy, the seated subject performing moderate and intensive bodily movements
caused similar exposures, despite higher emission rates associated with the more intensive
activity. This result is supported by the more prominent spatial concentration gradients
among the three stationary monitors for moderate movements (COV=28%), as compared to
the more intensive movements (COV=11%).

Particle emissions and personal exposure from handling paper and clothing fabric

The quantitative contribution of particle emissions through release from skin and clothing was augmented by investigating the influence of handling two common exogenous materials: office paper and clothing fabric. Previous studies reported effects of fabric manipulation on indoor particle levels,[15,19,26] but no consideration was given to the potential influence on personal exposure. Particle emission rates associated with routine handling of paper and its contribution to inhalation exposure are unknown.

To probe the relative contribution of these sources, a seated subject performed intensive movements that included manipulation of paper and an article of clothing. Specifically, the paper manipulation activities entailed distributing onto a table, then collecting and arranging groups of printed stapled office papers at a prescribed pace. The paper was standard quality white A4, 75 g/m^2, exposed to an indoor office environment for variable duration. The clothing manipulation activity entailed repeated folding of an additional black cotton shirt (treated in the same pre-experimental manner as for the clothing that was worn) and having the occupant put it on (over worn experimental clothing) and take it off his or her body. The contributions to emissions from manipulating paper and clothing fabric were derived by subtraction, considering paired experiments in which only the manipulation changed. Specifically, runs ID5 and ID4 were analysed to assess emissions from paper manipulation and runs ID4 and ID3 were compared to evaluate emissions from manipulating clothing fabric (Table S2).

Office paper was observed to be an important source of coarse airborne particles when manipulated by an occupant (Figure 5a). Arranging papers for a period of 12 min caused an approximately equivalent increment in inhaled total particle mass as that generated through body envelope shedding for a period of 30 min (Figure 2b). When observed on a time-averaged basis (i.e., as if the duration of manipulating papers and human envelope shedding were equal), manipulating office papers contributed to an increment in personal

exposure to coarse particles that was 2.7× that caused by human envelope shedding, and this increase was sufficient to offset the shorter duration of the activity producing a comparable overall contribution to exposure. The average emission rate of airborne particles in the size range 1-10 µm from the manipulation of office paper was 0.1 million per minute. This rate is nearly twice the corresponding emission rates from the skin and clothing of a seated human performing moderate activities.
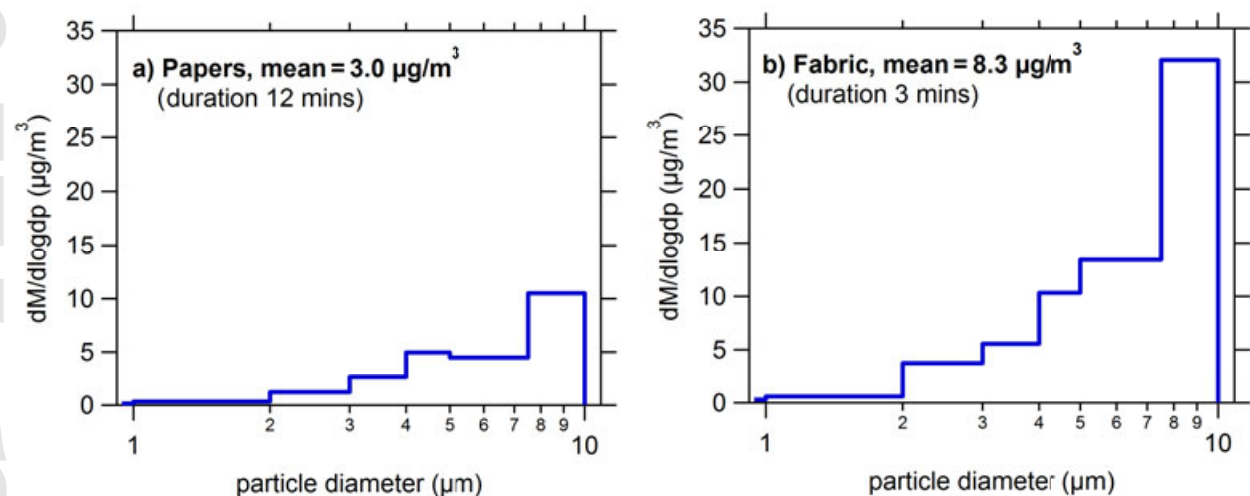

Fig. 5 Size distributions of average particle mass concentrations in the breathing zone of a seated occupant as a consequence of manipulating (a) paper for 12 minutes and (b) fabric for 3 minutes. Note that reported results represent 30-min averages of mean concentrations for the full activity period.

Clothing fabric manipulation through repeated folding/unfolding and putting on/taking off the shirt caused strong spikes in the breathing zone particle mass concentrations, peaking at above 40 µg/m^3 (Figure 2b). A 3-min fabric manipulation process increased the 30-min mean personal exposure by 8.3 µg/m^3 (Figure 5b), which translates to ~ 30× and ~ 10× increases in the time-integrated inhaled particle concentrations as compared with the respective contributions from human envelope shedding (intensive sitting activity, without paper or fabric manipulation) and manipulating paper, respectively. The corresponding mean emission rate of total particles in the size range 1-10 µm owing to fabric manipulation was 0.8 million per min. (The emission rates associated with fabric

manipulation would include the combined effects of particle detachment from fabric itself plus the emissions from the subject's worn clothing owing to enhanced friction.) This result substantiates previous research indicating that fabric manipulation can be a strong source of indoor aerosol particles.[19,28]

Influence of occupancy on room-average and personal cloud PM$_{10}$ mass concentrations

Figure 6 displays the room-average PM$_{10}$ mass concentration as a function of the number of occupants (zero, one or two) and activity type. The room-average PM$_{10}$ mass concentration exhibited a linear correlation with the number of occupants ($r^2 = 0.99$) across each activity. Two seated subjects performing moderate bodily movements contributed to PM$_{10}$ mass at a level that was ~ 50% less than a single walking subject.
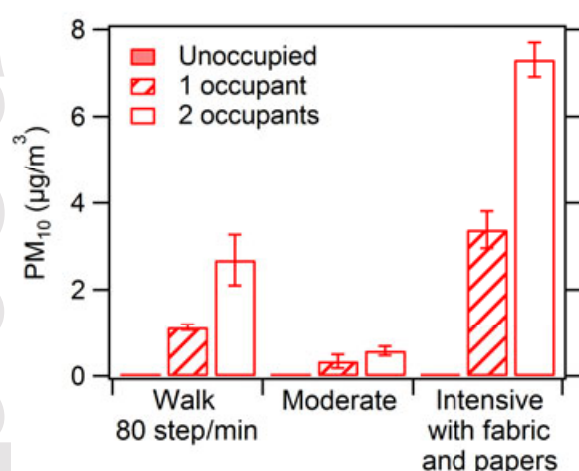

Fig. 6 Contribution of occupancy to the room-average PM$_{10}$ mass concentrations (± standard deviation) for three activity levels: walking at 80 step/min (ID = 6, ID = 10), sitting with moderate bodily movements (ID = 2, ID = 8); and sitting with intensive movement plus manipulation of fabric and paper (ID = 5, ID = 9). Data are averages of 30-min mean concentrations.

Figure 7 shows the PM$_{10}$ personal cloud magnitude of an occupant as a result of variable room occupancy and activity type. The increment of PM$_{10}$ mass in the breathing zone (personal cloud magnitude, *ΔM*) in the room when occupied by a single subject performing moderate and intensive bodily movements with papers and fabric was 2.5 and 12

µg/m^3, respectively. When two subjects performed these same activities simultaneously, the respective increment of PM$_{10}$ mass in the breathing zone decreased by 37% (to 1.6 µg/m^3) and by 19% (to 9.7 µg/m^3). This outcome suggests that the personal cloud magnitude might be higher at lower occupancy levels, perhaps in part owing to a lesser degree of room air mixing. Note that doubling the number of seated occupants who performed moderate body movements reduced the PM$_{10}$ coefficient of variation in the room from 28 to 4%. Although more research is needed to corroborate these findings, these results suggest an interpretation that occupancy levels may be inversely correlated with the size of the personal cloud.
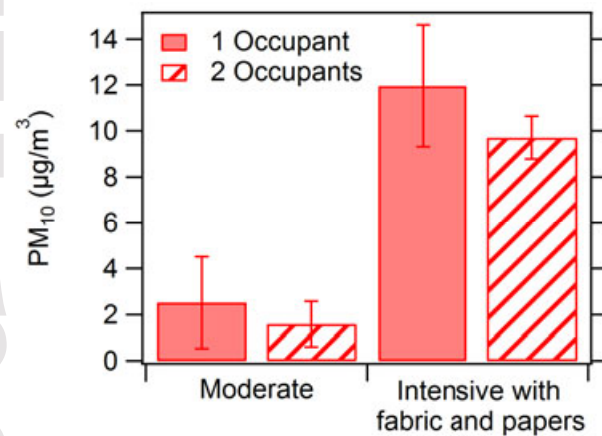

Fig. 7 The 30-min mean personal cloud PM$_{10}$ mass concentrations for two activity levels: sitting with moderate bodily movements (ID = 2, ID = 8) and sitting with intensive movement plus manipulation of fabric and paper (ID = 3, ID = 9).

The effect of proximity of the source to the receptor: cross-contamination

Particle release from the perihuman environment causes increases in the room-average concentrations, which would influence the exposure of other occupants in the room. That phenomenon is referred to here as cross-contamination. An important aspect when considering cross-contamination is the potential to contribute to the exposure of other occupants at a level above the room average (i.e., by enhancing other occupants' personal clouds). Such enhancement would depend on the spatial relationships between emitter and receptor. Although known to be a substantial contributor to the total aerosol load produced

through physical activities, the extent to which particles detached from the perihuman environment are transferred directly to the breathing zone of other occupants so as to contribute to those occupants' personal clouds has not been quantified. Our experiments with two occupants in the chamber provide an empirical basis for exploring such contributions to cross-contamination.

Figure 8 presents a quantification and comparison of the incremental personal exposure and the contribution to the personal cloud for two activity types. The left bar in each pair represents the total exposure from personal monitoring; the right bar reflects an enhancement in breathing zone concentration above the room average. For the left pair in each frame, the monitored subject is undertaking the activity. For the middle and right-hand pairs of bars, the monitored subject sits still while a second subject undertakes the activities at 1 and 2 m distance, respectively.

As clearly indicated in the figure, particles released through body envelope shedding are more important for autogenous exposure than for cross-contamination. In a near vicinity of a person (up to 0.45 m distance from the body), the convective boundary layer dominates the transport of particles upwards to the breathing zone, thus elevating personal exposure for particles released in the immediate perihuman space.[29-31] Particles that escape this boundary layer mix with the surrounding room air.[32] In these experiments, when a subject performed seated moderate bodily movements, the contribution to cross-contamination at 1 m distance was 11% of self-inhaled PM_{10} mass, and the value dropped further to 7% at 2 m distance. An increase in the shedding rate when the subject undertook intensified body movements was associated with a higher cross-contamination rate. Contribution to the PM_{10} mass in the breathing zone at 1 m was 3.4 µg/m^3 (22% of the self-inhaled concentration), and 2.6 µg/m^3 at 2 m distance (17% of the self-inhaled concentration). However, these enhancements are primarily attributable to increased room-averaged concentrations from the second occupant,

rather than from an increase specifically in the breathing zone of the monitored subject. Other studies have identified that the proximity of an exogenous source can meaningfully influence personal exposures.[2,33,34] Our results demonstrate than the human envelope shedding may contribute to cross-contamination, but that it has minimal influence on the personal cloud of other occupants even at 1 m distance.
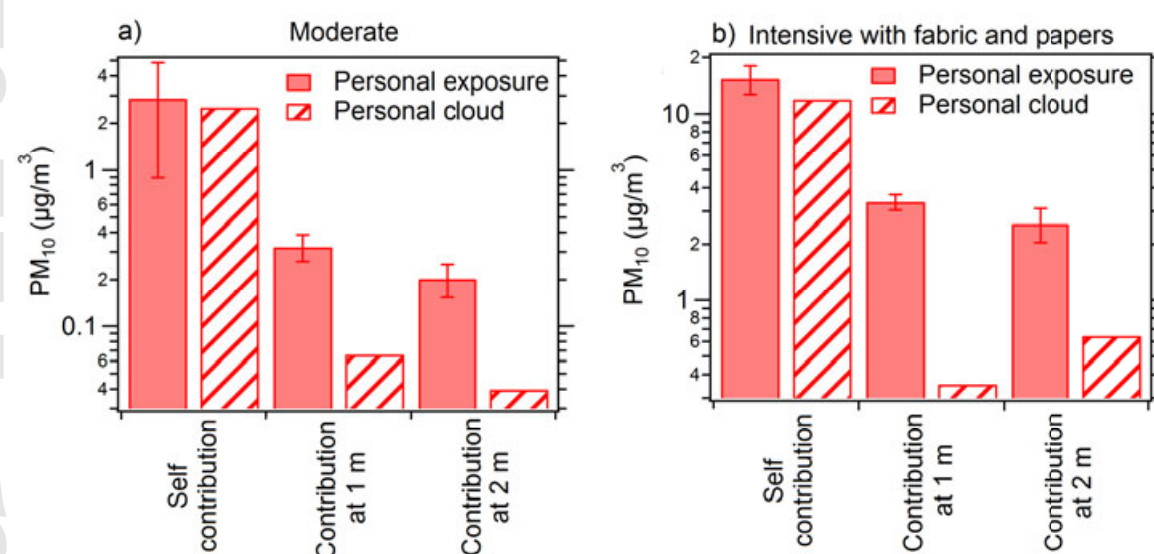

Fig. 8 Contribution to self-exposure and to the exposure of a subject seated at 1 and 2 m distance from the source for two activity types: (a) Seated, moderate movement (ID = 2), and (b) Seated, intensive movement with fabric and papers (ID = 5). The bars represent 30-min average contributions to exposure, based on measured particle concentrations in the breathing zone. When a second subject undertook activities in the chamber, at 1 m and 2 m distance, the monitored subject was seated and still, with negligible shedding rate.

Importance of human envelope shedding for exposure assessment

Environmental policies rely on ambient air quality data to predict human exposure, typically without taking account of indoor-outdoor relationships. Even studies that are designed to consider indoor-outdoor relationships often do not incorporate information about indoor spatial and temporal variations of airborne particle concentrations. Recent research has shown, for example, that airborne particle concentrations can be substantially higher during occupied periods than when spaces are unoccupied.[26,27,35,36] As a step towards more accurate exposure assessment, researchers have proposed that only the airborne particle concentrations

from occupied periods should be taken into account when incorporating information about indoor particle levels.[37] Our results suggest that stationary monitors designed to sample indoor air may also systematically underestimate human exposure to particulate matter in the coarse size fraction owing to spatially varying concentration fields. When a subject is seated and moderately active, it seems likely that there would be a continuous material contribution to the personal cloud owing to particle shedding from skin, clothing, contact surfaces, and (certain) manipulated objects. Estimates of the scale of the personal cloud effect range from 2.2 $\mu g/m^3$ for human envelope shedding, to 11 $\mu g/m^3$ when manipulating office paper and fabrics. This scale of effect is substantial when compared with total exposure estimates in particulate-matter health effect studies.

The particle diameter range 2-10 μm includes the dominant size mode of indoor airborne bacteria.[11,12] The human body is home to a diverse community of microorganisms, primarily bacteria,[38] but also fungi[39] and other organisms. Clothing can also contain microorganisms such as bacteria[40-42] and viruses[43] that, together with bacteria-laden skin flakes, fragments and fibers, get dispersed to the surroundings via occupants' activities owing to frictional forces involving fabric fibers, external contact surfaces, and/or the wearer's skin.[15,19,44,45] Increasing evidence identifies human occupancy as an important source of airborne bacterial and fungal DNA in indoor air.[11,12,16] Particles detached from the human envelope might also carry chemical and other potentially harmful agents, such as residual detergents and post-manufacturing hazardous substances that remain in clothing and textiles, and that are otherwise known to cause allergic sensitization and other health effects.[46,47] While the role of skin and clothing-associated microorganisms and chemicals on health is not sufficiently understood, it seems worthwhile to consider further the role of perihuman releases on occupant exposures.

Conclusions

Emissions from occupants' skin and clothing are potentially important contributors to aerosol particles indoors. In a controlled chamber study with low background particle levels, we measured the emission rates of total particles larger than 1 µm from a single occupant to be (average ± standard deviation) 20 ± 2.0 million particles per h for walking. When the subject was seated with moderate movement, the emission rate was 8 ± 3 million particles per h.

This study is the first to experimentally quantify the contributions of human envelope shedding to personal exposure and to the personal cloud effect. The personal cloud varied primarily in relation to the activity type. Despite the higher emissions associated with walking, the exposure of a sitting occupant in a low-background chamber was 2-13 µg/m^3 higher owing to heterogeneously distributed particulate matter. Emissions from a seated occupant were associated with unsteady increases in inhaled pollutant concentrations, producing a mean personal cloud magnitude of up to 2.3 µg/m^3. The personal cloud appeared discernible for particles larger than 2 µm in optical diameter, and, with regard to particle mass, the effect increased more for larger particles and with fewer room occupants. This evidence supports a view that — in spaces where occupants are primarily seated and when occupancy is sparse — the well-mixed representation of an indoor environment might yield systematic underestimates of human exposure to coarse airborne particulate matter.

The release of particles associated with the manipulation of fabrics and of office paper should be recognized as potentially important contributors to indoor particle emissions and elevated personal exposures. In particular, we found emissions of supermicron particles from handling paper to be 0.1 million particles per minute, about 2× higher than that associated with shedding from the human envelope itself. Manipulating a fabric appeared to be a more potent source of total aerosol particles, with an average emission rate of about 0.8 million

particles per minute.

Particle release from the perihuman environment also contributed to cross-contamination. The PM_{10} mass exchange between two occupants due to particle release from the skin and clothing was 0.32 µg/m^3 and increased to 3.4 µg/m^3 when it involved manipulating papers and fabric. This mass exchange diminished by 25-35% with increased distance between occupants from 1 to 2 m. It seems worthwhile to further investigate the nature of human associated particle emissions and how they influence indoor inhalation exposures of themselves and of other occupants, taking proper account of the proximity effects on cross-contamination.

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