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Human emissions of size-resolved fluorescent aerosol particles: Influence of personal and environmental factors

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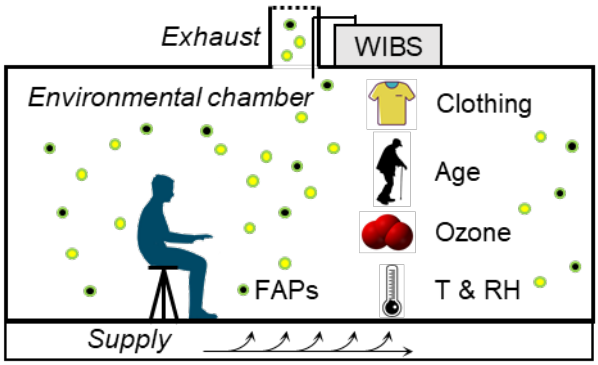
ABSTRACT

Human emissions of fluorescent aerosol particles (FAPs) can influence the biological burden of indoor air. Yet, quantification of FAP emissions from humans remains limited, along with poor understanding of the underlying emission mechanisms. To reduce the knowledge gap, we characterized human emissions of size-segregated FAPs (1-10 μm) and total particles in a climate chamber with low-background particle levels. We probed the influence of several personal factors (clothing coverage and age) and environmental parameters (level of ozone, air temperature and relative humidity) on particle emissions from human volunteers. A material-balance model showed that the mean emission rate ranged 5.3-16 × 10^6 fluorescent particles per person-h (0.30-1.2 mg per person-h), with a dominant size mode within 3-5 μm. Volunteers wearing long-sleeve shirts and pants produced 40% more FAPs relative to those wearing T-shirts and shorts. Particle emissions varied across the age groups: seniors (average age 70.5 years) generated 50% fewer FAPs compared to young adults (25.0 years) and teenagers (13.8 years). While we did not observe a measurable influence of ozone (0 vs. 40 ppb) on human FAP emissions, there was a strong influence of relative humidity (34 vs. 62%), with FAP emissions decreasing by 30-60% at higher humidity.

Keywords: Chamber study; Bioaerosols; Clothing; Age; Indoor climate; Ozone

GRAPHIC ABSTRACT



INTRODUCTION

Exposure to bioaerosols (biological aerosol particles) is an important factor influencing human health.[1,2] Although bioaerosol exposure can cause adverse health effects, such as infectious disease transmission and respiratory problems,[3,4] it has also been linked to protective effects for atopic conditions such as asthma.[5–9]

The indoor concentration of bioaerosols is influenced by a multitude of processes including ventilation, deposition, penetration from outdoors through the building envelope, and indoor emissions.[2,10] Among the various bioaerosol sources indoors, humans strongly contribute through a combination of extrinsic activity-related emissions[11–13] such as vacuuming[14,15] and cooking,[16,17] and intrinsic emissions produced by a combination of expiratory flow[18] and release from skin and clothing.[19–23] Human presence and associated activities can significantly impact the concentration and diversity of indoor bioaerosols in homes,[11,17,24,25] offices,[26,27] classrooms,[28–36] aircraft cabins,[37,38] and hospitals.[39,40] Yet, the intrinsic mechanisms of human bioaerosol emissions that determine human exposures remain poorly characterized. Recent advances in laser-induced fluorescence (LIF) techniques enable highly time- and size-resolved bioaerosol measurements.[2,41] Bioaerosols, particularly in coarse-mode, which exhibit laser-induced fluorescence, are designated as fluorescent biological aerosol particles (FBAPs).[42–44] However, LIF instruments have an inherent limitation when interpreting fluorescent particle measurements as a proxy for bioserosols. Hence, in this study, we term LIF-measured particles as fluorescent aeosol particles (FAPs). Few studies to date have used LIF instruments to characterize intrinsic human emissions of FAPs in controlled environments; and emission rates reported thus far highlight the importance of this source.[45,46]

Skin- and clothing-associated particles and microbes have been detected on indoor surfaces, in settled dust, and in air,[33,47–52] accounting for up to 40% of the total microbiome signature indoors.[53] The intensity of bioaerosol detachment from human surfaces and effectiveness of their transport is influenced by various personal and indoor environmental conditions, albeit to unknown extent. Early research suggested that clothing increases bioaerosol emissions by promoting the detachment of human skin flakes via friction.[54,55] More recent studies provided evidence that clothing also resuspends previously deposited bioaerosols, and can serve as a transport vector for bioaerosols.[56–59] Age may be also an important factor influencing shedding rate and overall FAP emissions, as skin surface conditions are known to change over the human lifetime.[60] Variation in environmental factors, such as ozone level, could be an additional factor influencing FAPs, owing to its ability to alter fluorescence intensity and spectra of bioaerosols,[61–63] though the effect on human-emitted FAPs is unknown. Air temperature and relative humidity are two additional factors that could alter human FAP emissions due to their known impact on particle adhesion and resuspension.[64–66]

Controlled studies that isolate human bioaerosol emissions from other indoor emissions and that probe the influence of personal and environmental parameters are lacking. Therefore, this study quantified the time- and size-resolved FAP emission rates from humans and investigated the influence of clothing level, age, presence of ozone, air temperature and relative humidity.

MATERIALS AND METHODS

The Climate Chamber

Within the ICHEAR (Indoor Chemical Human Emissions and Reactivity) project,[67,68]

we performed a series of experiments to quantify human emissions of FAPs in a 22.5 m^3 stainless-steel climate chamber (Figure S1). The chamber was ventilated with 100% outdoor air supplied through a perforated floor. A stainless-steel open mesh flooring was present on top of the perforated floor, making it suitable for reducing particle resuspension due to small surface area available for deposition and contact. The air was exhausted via an outlet in the ceiling where the particles were sampled. The outdoor air change rate in the chamber during the experiments was 3.2 ± 0.1 h^{-1}.[67] To minimize emissions other than from the human volunteers, the chamber was furnished with only a table, four wire mesh metal chairs and two pedestal mixing fans facing the chamber walls to ensure air mixing. Prior to the experiments, all the surfaces were thoroughly cleaned.[67] The heating, ventilation and air conditioning (HVAC) unit of the chamber was used to control chamber air temperature (T) and relative humidity (RH). The outdoor air was filtered with a newly installed F7 glass fiber particle filter, and a high efficiency molecular filter (activated carbon in loose-fill canisters), which ensured coarse particle- and ozone-free air supply (<10 particles/L and <1 ppb, respectively). Ozone was generated by delivering pure oxygen through a Jelight 600 UV lamp (Jelight Co. Inc., USA) in the chamber's supply air duct.

Experimental Design and Procedure

During the experiments, four volunteers sat around the table in the chamber and performed semi-scripted activities. The activities included use of their own smartphones or provided computer tablets, and standing up for 5 minutes every hour to stretch and walk within the chamber. They stayed in the chamber for 3 h (three ~1 h seated periods and two 5-min standing activities). Upon exiting the chamber, the measurements continued in the unoccupied chamber to determine the decay of particle concentrations and estimate the particle deposition onto the chamber surfaces. In total,

five groups of four human volunteers were recruited. Three groups consisted of young adults (A1-A3), one consisted of teenagers (T4) and one of seniors (S5).[67] Groups A1, A2, T4 and S5 were selected for investigations of FAP emissions. Groups A2, T4 and S5 included two males and two females, while group A1 consisted of three males and one female. To investigate the influence of clothing, age, ozone, T and RH on human emissions of FAPs, we conducted four sets of experiments with a total number of 20 experiments (Table S1).

Clothing and ozone. We tested two sets of loose-fitting clothing, named here as "long" and "short" clothing. The "long" clothing set included a sweat pant, long-sleeve shirt, and calf socks. The "short" clothing set contained shorts, a t-shirt, and ankle socks.[67] Both clothing assemblies were black in color in order to minimize interference of non-biological fluorescent signals, which can largely originate from white clothing.[69] Volunteers selected the clothing size that they normally used from the clothing sets otherwise identical for all volunteers.

Experiments were designed to probe the combined influence of ozone and clothing on human emissions of FAPs. Conditions with and without the presence of ozone were applied to both the "long" and "short" clothing scenarios. Group A2 wearing "long" clothing in the absence of ozone was considered as benchmark scenario. In experiments with ozone injection, a mean steady-state concentration of indoor ozone of 102 ± 2 ppb was established before volunteers entered the chamber. After the volunteers entered, ozone injection continued at a constant rate and ozone concentrations equilibrated at ~40 ppb.[67]

Age. We examined FAP emission rates from three age groups: 1) young adults, with the average age of 25.0 years (range 19-30); 2) teenagers (13.8 years, range 13-15); and 3) seniors (70.5 years, range 68-72). The average BMI index was 21.6 (range 20.0-23.9), 19.5 (range 19.1-20.4) and 25.6 (22.5-28.1) for young adults, teenagers and seniors, respectively.

T and RH. A 2×2 experimental matrix was designed to investigate the effect of T and RH. Moderate (28.7±0.7 °C) and high (32.5±0.1 °C) T, and low (34±2%) and high (62±1%) RH were considered. This set of experiments was performed with group A1, while ozone was absent in the chamber.

Apart from the 17 basic experiments (including 7 replicates), we performed 3 supplementary experiments with different groups of volunteers to validate the effects of ozone and clothing (Table S1). More details about the environmental chamber and the experimental procedures are described in Section S1 and in Bekö et al.[67]

Instrumentation

Fluorescent and total particles in the chamber were measured by the Wideband Integrated Bioaerosol Sensor (WIBS) with 0.3 L/min sampling flowrate (WIBS NEO, Droplet Measurement Technologies, US). The WIBS was placed on the roof of the chamber, as close as possible to the exhaust air duct to minimize coarse particle deposition inside the silicon sampling tube (length: total 67 cm, horizontal 17 cm). The end of the sampling tube was attached to the chamber's ventilation outlet in the ceiling, facing downwards to ensure effective capture of particles. Based on fluorescence intensities of the detected particles, we categorized them into eight types: "A", "B", "C", "AB", "AC", "BC", "ABC" and non-fluorescent,[70] as detailed in Section S2 and

Figure S2. Particles of different fluorescence types have been associated with distinct biological and abiotic nature.[69, 71-74]

The ozone concentration was monitored inside the chamber using an ozone monitor (Model 205, 2B Technologies, US) with a time resolution of 10 seconds at 2.0 L/min sampling flowrate. T, RH and CO_2 inside the chamber were continuously recorded every minute by a Vaisala GMW90 instrument (Vaisala Corporation, Finland) coupled with a data logger (HOBO UX120-006M, Onset Computer Corporation, US). In addition, we asked the volunteers to measure and record their skin water content (in %) once every hour using a Neon SK-5D skin analyzer, which determines skin wetness from the measured capacitance.[75,76]

Data Analysis

The raw data files from the WIBS consisted of size-resolved and fluorescence-categorized particle number concentrations based on the characteristics of each detected particle with millisecond resolution. From the WIBS particle detection size range 0.5-50 μm, we selected particles in the 1-10 μm range for further investigation. This was done because human-emitted FAPs predominantly fall into this size range, according to previous research.[36,45,46] We further aggregated the number concentrations into six size bins (1.0-2.0, 2.0-3.0, 3.0-4.0, 4.0-5.0, 5.0-7.5, 7.5-10.0 μm) to report emission rates. Particle loss in the sampling tube was estimated using Particle Loss Calculator, the accuracy of which has been validated against a complicated geometry of tubes.[77] The correction coefficient for each size bin is shown in Table S2.

The time-averaged per person emission rates of FAPs, *ER* (# per person-h), were computed based on the transient integral material-balance model,[24,25,46,78,79] shown in

Equation 1. In the equation, V is the chamber volume, m^3; τ is the experiment duration, h; $N(\tau)$ and $N(0)$ are the 5-minute average particle number concentrations[24] at the end and the start of the experiment, respectively, $\#/m^3$; $\overline{N}$ is the average concentration from the beginning of experiments until the end of occupied period, $\#/m^3$; a is the air change rate of the chamber, h^{-1}; β is the first-order particle deposition loss rate, h^{-1}; and n is the number of human volunteers. The particle deposition loss rate onto chamber surfaces β was obtained from the decay period after volunteers exited the chamber (listed in Table S3). The subscripts k and i represent particle fluorescence type and size bin, respectively.

$$\mathrm{ER}_{k,i} = \frac{V}{n}\left(\frac{N(\tau)-N(0)}{\tau} + (a+\beta_i)\overline{N_{k,i}}\right) \quad (1)$$

Equation 1 can also be applied to specifically evaluate human emissions of FAPs during sitting and standing activities. For this purpose, $N(\tau)$, $N(0)$, $\overline{N}$ and τ were also calculated for the two corresponding activities. We evaluated three sitting periods and two standing periods in each experiment. To estimate the emissions during the second and the third sitting periods, we used the last 15 minutes of sitting activities to avoid the influence of prior standing activities. We did not detect significant initial influx of particles caused by the short (~5 s) door-opening during entering the chamber (Figure 1) and this effect was considered negligible.

For the purpose of number-to-mass conversion, we assumed that particles are spherical with 1.0 g/cm^3 density, and that the mass-weighted size distribution is constant within each size bin.[80] Since the average airborne particle density is 1-2.5 g/cm^3,[81] (1.0-1.5 g/cm^3 for common bioaerosols including bacteria and yeast cells,[82–84]), and indoor bioaerosols can have a diversity of shapes (particularly large asphericity of coarse particles),[46,71,85,86] the mass concentrations reported here can be considered lower-bound estimates. To statistically evaluate the influence of investigated factors, we

performed the non-parametric Wilcoxon Mann-Whitney *U* test (SPSS 21) among emission rates during sitting periods for experiments with replicates.

Quality Control

Calibration of the WIBS was performed by the manufacturer (Droplet Measurement Technology) before the campaign. On-site checks of size calibration, sampling flow and zero count were conducted before and after the experiments. The size calibration was checked using monodispersed 1.005 μm and 2.005 μm polystyrene latex (PSL, Thermal Scientific, US), and showed < 10% error.

To ensure the robustness of the particle measurements, we performed measurements in parallel to WIBS using an aerosol spectrometer (Mini-WRAS, Grimm Aerosol Technik, Germany) at the chamber exhaust. The Mini-WRAS was capable of detecting aerosols ranging from 10 nm to 35 μm divided into 40 size channels. The preparation of the Mini-WRAS was identical to the WIBS, and disparity between the emission rates of total particles based on the measurement results of the two instruments was always below 12% (Table S4).

RESULTS AND DISCUSSION

Characteristics of Particle Emissions

Figure 1 shows time series of size-dependent (1-10 μm) total and fluorescent particle number concentrations measured in the chamber that was occupied with young adults (group A2) wearing "long" clothing (benchmark case). When the volunteers entered the chamber, there was a strong increase in the particle number concentration. During the first one-hour sitting period, the average FAP concentrations inside the chamber were around 0.4×10^6 m^{-3} with a dominant size mode between 3-6 μm. When the volunteers

stood up, the FAP and total particle concentrations increased sharply approximately 3 and 4 times, respectively, and peaked when the standing period ended. Every 5-minute standing event was followed by gradual decay of particle number concentration, with values returning to those seen prior to standing after 30 minutes. After the volunteers exited the chamber, the particle concentrations decreased gradually to the background, due to removal by ventilation and deposition.

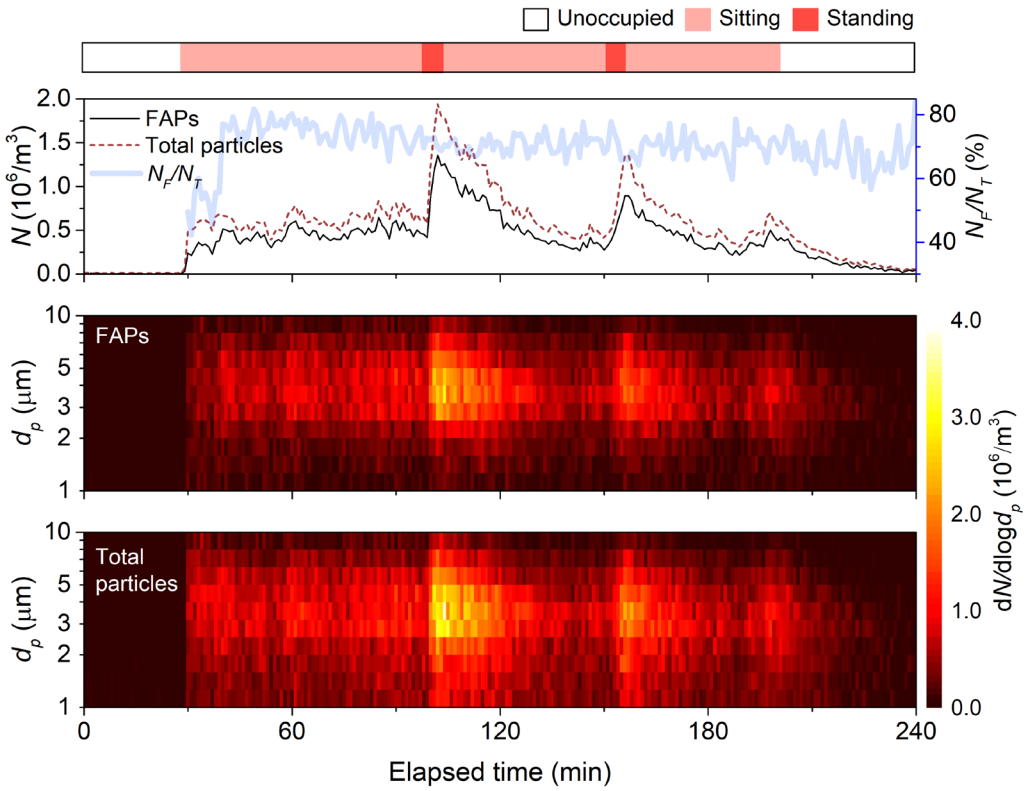


Figure 1. Time-series plots of FAP and total particle number concentrations (N, top left axis), fluorescent fraction (N_F/N_T, top right axis), FAP number size distributions (middle) and total particle number size distributions (bottom) across different activities. Benchmark experiment: young adults (A2) wearing long-sleeve shirts and pants with moderate T, low RH and without ozone.

Figure 2 shows the emission rates of size-resolved particles categorized according to fluorescence type during the entire three-hour period with occupancy in the benchmark case. The particle emission profile by count exhibited a lognormal peak with a dominant size mode between 3-4 μm. Particle emissions by mass were not discernible below 2

μm, but there was a distinct increase in the emission rates in the diameter range 3-10 μm. Within size range 1-2 μm, 46% of particles (by count) were fluorescent, and the proportion of particles exhibiting fluorescence gradually increased to 96% within the size bin 7.5-10 μm, owing to the generally increased fluorescence with growing particle volume.[69] Among the specific fluorescence categories, particles within 1-2 μm range had type "A" FAPs as the most abundant. Within the 2-4 μm size range, the "ABC" category was dominant, followed by "AB" and "A". The "ABC" type constituted the majority of FAPs larger than 4 μm, and more than 95% in the size range 7.5-10 μm. The "ABC" type particles constituted 86% of the FAP mass emissions. This is consistent with the results reported by Zhou et al.[46] While our results do not provide information about biological specificity of particles, the types "ABC", "AB" and "A" particles have been commonly associated with a mixture of biotic (bacteria, fungi, pollen, etc.) and abiotic sources (clothing fibers, detergents residue, soot and smoke, mineral dust, etc.).[69, 71–74] A comprehensive analysis of possible biological sources and abiotic interferences contributing to the detected FAPs can be found in Table S5.

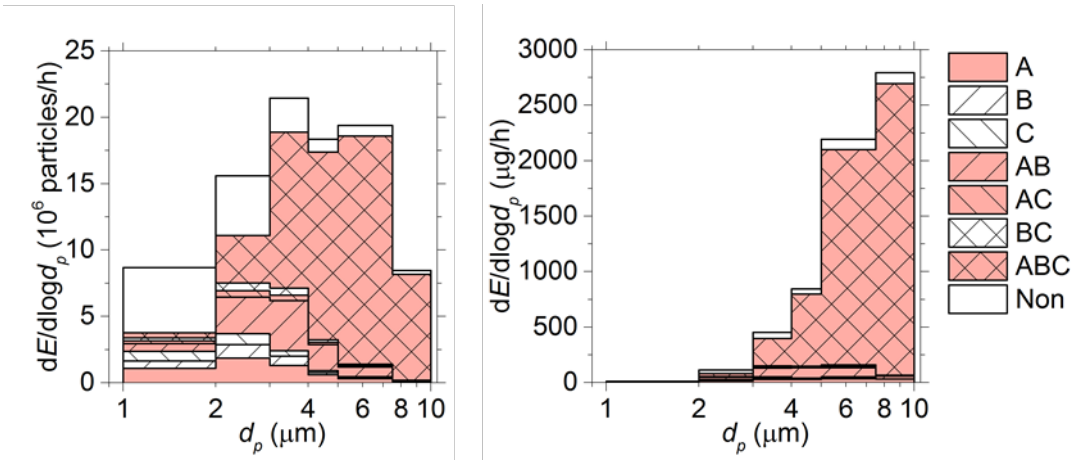


Figure 2. Size-resolved per person particle emission rates by count (left) and mass (right) during the entire occupied period for one experiment, differentiated by eight particle fluorescence types. Benchmark experiment: young adults (A2) wearing long-sleeve shirt and pants with moderate T,

low RH and no ozone.

The emission rates of standing volunteers were approximately 8 times higher for both FAPs and total particles in the 1-10 μm size range compared to sitting volunteers (Figure S3). The elevated emissions during standing are likely attributed to increased vigor of bodily movement and more intensive frictional interactions between the skin and clothing, as has been reported previously elsewhere.[45,78] The fluorescent fractions, defined as the ratio of FAPs to total particles, E_F/E_T, were consistently lower during standing activities (Figure S3c). We suspect that standing activity dislodged and resuspended a certain amount of particles that do not exhibit fluorescence from the clothing and other contact-surfaces in the chamber.

Influence of Personal and Environmental Factors

For the total period with occupancy, human volunteers emanated 5.3-16 × 10^6 FAPs per person-h across the various personal and environmental conditions, corresponding to mass emission rates within 0.30-1.2 mg per person-h (Table 1). FAPs constituted 64-82% of the total number of human-emitted particles, and 90-95% of total mass of emitted particles. Fluorescent particles of "ABC" type dominated FAP emissions across all the experiments (Table S7). Due to relatively short periods of standing activities, the emission rates during the total periods with occupancy were comparable or slightly higher than during the sitting periods. The emission rates obtained from the total periods are more representative of typical occupied conditions. However, since the standing activities could not be always fully controlled, in the following section, we report isolated emission rates for the sitting periods only. The lognormal fitting of the seated emission rates as a function of particle size bin can be seen in Table S6.

Table 1. Mean ± standard deviation of human emission rates per person of 1-10 μm FAP number (E_F), FAP mass (E_{F_m}), total particle number (E_T) and particle number of the dominant fluorescence type "ABC" (E_{ABC}). The results are reported for the entire three-hour periods with occupancy and for the sitting periods only. The values represent averages of all available data from all measured periods (N) for a given experimental condition/row (including replicate measurements when available). There were 3 sitting periods in each 3h period with occupancy.

Influencing factors and experimental conditions		**Entire three-hour periods with occupancy**					**Sitting periods**				
		N	***E_F*** **(10^6/h)**	***E_{F_m}*** **(mg/h)**	***E_T*** **(10^6/h)**	***E_{ABC}*** **(10^6/h)**	***N***	***E_F*** **(10^6/h)**	***E_{F_m}*** **(mg/h)**	***E_T*** **(10^6/h)**	***E_{ABC}*** **(10^6/h)**
Clothing and ozone [a]	"Long", without ozone	2	16±1	1.2±0.1	20±1	10±1	6	13±3	1.00±0.50	16±3	8.3±1.8
	"Short", without ozone	2	11±1	0.89±0.09	14±2	7.8±0.8	6	8.7±2.8	0.70±0.25	11±3	5.9±2.2
	"Long", with ozone	2	12±1	0.90±0.05	15±1	8.1±0.5	6	12±2	0.92±0.28	15±4	8.0±3.1
	"Short", with ozone	2	8.5±1.4	0.65±0.13	11±2	5.7±1.1	6	8.3±2.8	0.65±0.31	11±3	5.5±2.4
Age [b]	Young adults	2	16±1	1.2±0.1	20±1	10±1	6	13±3	1.00±0.50	16±3	8.3±1.8
	Teenagers	2	12±2	0.83±0.11	16±2	8.2±1.1	6	12±3	0.82±0.31	16±3	8.2±2.7
	Seniors	2	5.4±2.2	0.30±0.13	8.1±3.0	2.8±1.3	6	5.2±2.7	0.30±0.16	8.1±4.0	2.8±1.5
T and RH [c]	Moderate T, low RH	1	7.1	0.53	11	4.4	3	6.6±2.3	0.52±0.19	10±3	4.2±1.7
	Moderate T, high RH	2	5.5±1.2	0.40±0.08	8.6±1.9	3.1±0.7	6	4.8±1.8	0.34±0.17	7.6±2.5	2.7±1.3
	High T, low RH	1	12	0.85	16	7.7	3	12±4	0.86±0.33	16±5	7.5±2.9
	High T, high RH	1	5.3	0.39	7.5	3.0	3	4.8±1.7	0.33±0.14	6.8±2.2	2.6±1.3

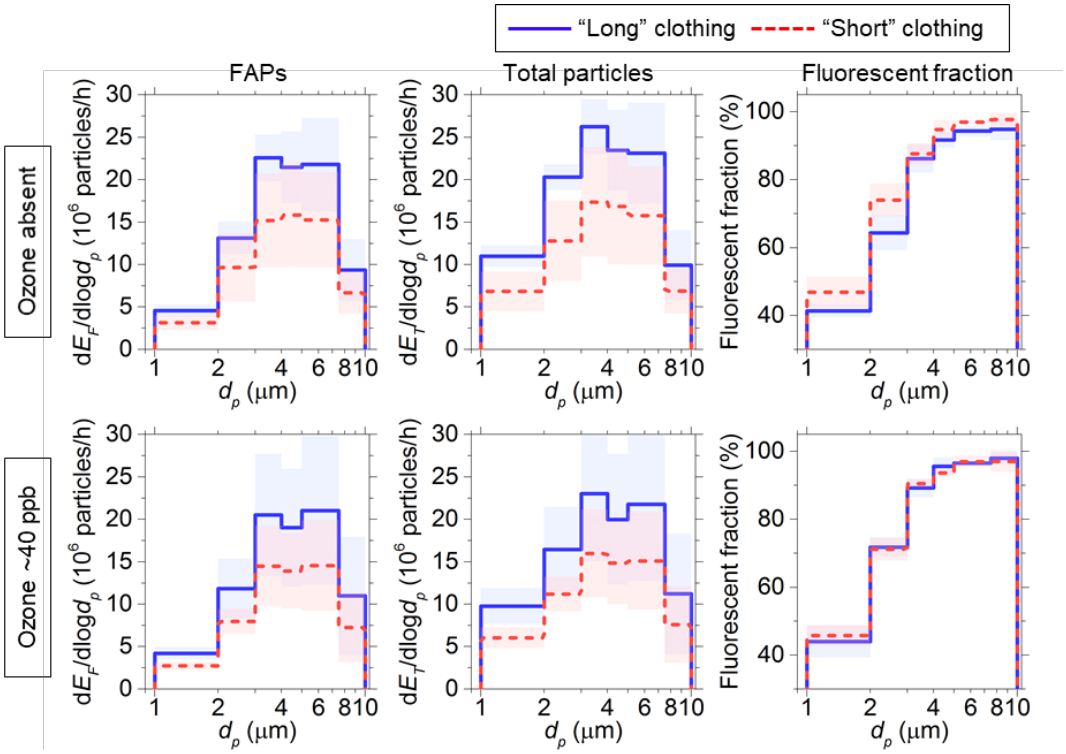
[a] Experiments with young adults, T: 25.5±0.5 °C, RH: 24±2 %. "Long" and "short" clothing refer to long-sleeve shirts and pants, and T-shirt and shorts, respectively. In cases with ozone, volunteers were exposed to ~40 ppb ozone.

[b] Experiments with long-sleeve shirts and pants, ozone absent, T: 26.6±1.1 °C, RH: 30±5 %.

[c] Experiments with young adults wearing long-sleeve shirts and pants, ozone absent, moderate T: 28.7±0.7 °C, high T: 32.5±0.1 °C, low RH: 34±2%, high RH: 62±1%.

Influence of clothing and ozone. Sitting young adults emanated considerably more particles when wearing long clothing than when wearing short clothing. The respective emission rates with ozone absent were 13 ± 3 vs. 8.7 ± 2.8 × 10^6 per person-h for FAPs (p<0.01), 16 ± 3 vs. 11 ± 3 × 10^6 per person-h for total particle number (p=0.02), and 8.3 ± 1.8 vs. 5.9 ± 2.2 × 10^6 per person-h for FAPs type "ABC" (p=0.04). The difference was statistically significant across the full 1-10 μm size range, as shown in Figure 3. The higher emission rates from volunteers wearing long clothing can be attributed to emissions from the clothing itself (larger emission area) and larger contact area between skin and clothing, which promotes detachment of human skin flakes and clothing material via friction.[54,55,59] Particle emissions from clothing can originate from intrinsic sources such as clothing fiber and fabric fragments from manufacturing, and extrinsic sources through environmental particle uptake (Table S5).[59] Since the clothing used in the experiments was laundered with non-whitening detergent (to limit the effect of whitening agents on fluorescence) and sealed prior to use, we hypothesize that particles with extrinsic origin do not play a major role compared to intrinsic emissions.

Despite the lower emission rates of FAPs when the volunteers wore short clothing, the fraction of fluorescent particles in the total particles was higher with short clothing in the absence of ozone, particularly for 1-4 μm particles (p=0.02). This may suggest that a larger proportion of non-fluorescent particles was released in the smaller particle size range with long clothing compared to short clothing. The fluorescent fractions were similar for the two clothing types when ozone was present, although it was still slightly higher with short clothing for 1-2 μm particles.



320

321 **Figure 3**. Influence of clothing type and ozone on size-resolved per person number emission rates
322 of FAPs and total particles, and fluorescent fraction during periods when the volunteers were sitting.
323 The lines represent averages of all available data within each size bin for the given experimental
324 conditions (N=6 (sitting periods per condition); see Table 1). The shaded areas represent standard
325 deviations. "Long" and "short" clothing refer to long-sleeve shirts and pants, and T-shirt and shorts,
326 respectively.

327 Ozone concentrations in the chamber decreased rapidly from ~100 ppb to ~40 ppb after
328 human volunteers entered the chamber (Figure S4), reflecting reactions of ozone with
329 constituents of skin oils, such as squalene. These reactions generate a number of new
330 volatile organic compounds (VOCs).[67,87–90] Ozone reactions with used clothing have
331 been reported to generate ultrafine particles (10-100 nm).[91] However, the presence of
332 ozone did not influence human emissions of 1-10 μm fluorescent and total particles
333 significantly (Table 1 and Figure S5). Although the differences were small, emission
334 rates were consistently slightly higher with ozone absent. This finding was confirmed
335 by the supplementary experiments with teenagers and seniors (Table S8). Existing

literature on the relationship between ozone and human-associated FAPs is limited and merits further attention, particularly with longer term exposures.

Influence of age. Figure 4 compares the size-resolved number emission rates of fluorescent and total particles for the three age groups (young adults, teenagers and seniors) during periods of sitting, with "long" clothing and ozone absent. Teenagers contributed to indoor FAP levels with similar average emission rates as young adults (13 ± 3 vs. $12 \pm 3 \times 10^6$ per person-h). While teenagers emitted slightly more particles in the smaller size fraction, young adults emanated more particles in the larger size fraction, and had thus slightly higher FAP emission rates by mass compared to teenagers (1.00 ± 0.50 vs. 0.82 ± 0.31 mg per person-h). Senior volunteers emitted as much as ~50% fewer FAPs and total particles than teenagers and young adults (see also Table 1). While particle production by seniors within the 1-2 μm size range was similar to that of teenagers and adults, large differences were observed for particles larger than 2 μm. FAP emission rates by mass were thus significantly lower for seniors (0.30 ± 0.16 mg per person-h, $p<0.01$). The fraction of fluorescent particles in total particles was lower for seniors than for the other two age groups (Figure 4). The fluorescence type of particles emitted from seniors also differed from that from teenagers and adults. Seniors emanated significantly larger proportion of type "A" and smaller proportion of type "ABC" FAPs, particularly in the large size-ranges (Figure S6 and Table S9). During the experiments, seniors had similar activity levels compared to teenagers and adults, as evidenced by the comparable steady-state CO_2 concentrations in the chamber (Table S1), whereas their microscale movements may have differed and thus led to varied emissions. In addition, we suspect that the observed disparity in particle emissions could be attributed to differences in skin condition.[60,92,93] Seniors replace their stratum corneum slower than teenagers and young adults,[94] and may thus have a lower rate of

skin shedding (desquamation). In addition, larger wrinkle areas[95] and increased skin roughness[96] among seniors could lead to reduced physical contact between clothing and skin. This could be responsible for trapping particles and eventually lowering particle emissions. Age-dependent skin microbial diversity may also play a role in the disparities between FAP emissions across the three age groups.[97,98] Further research is warranted to investigate the underlying emission mechanisms associated with different age.

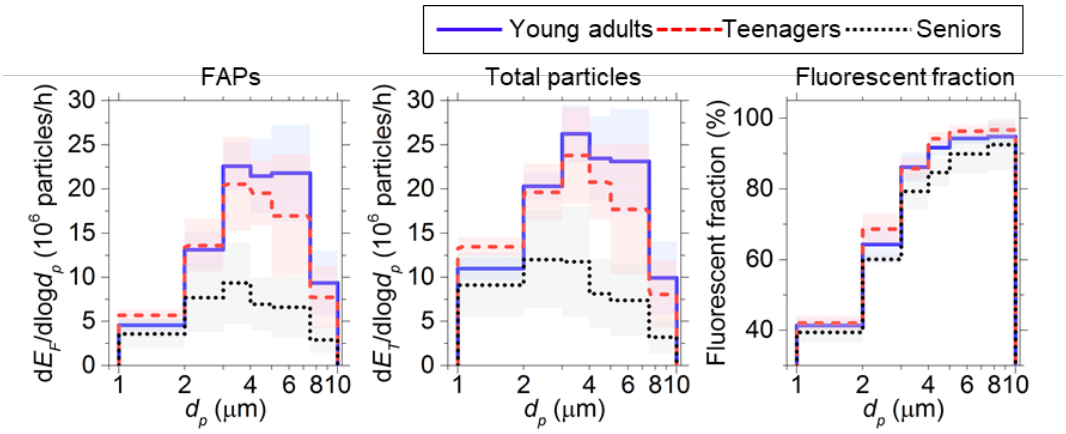
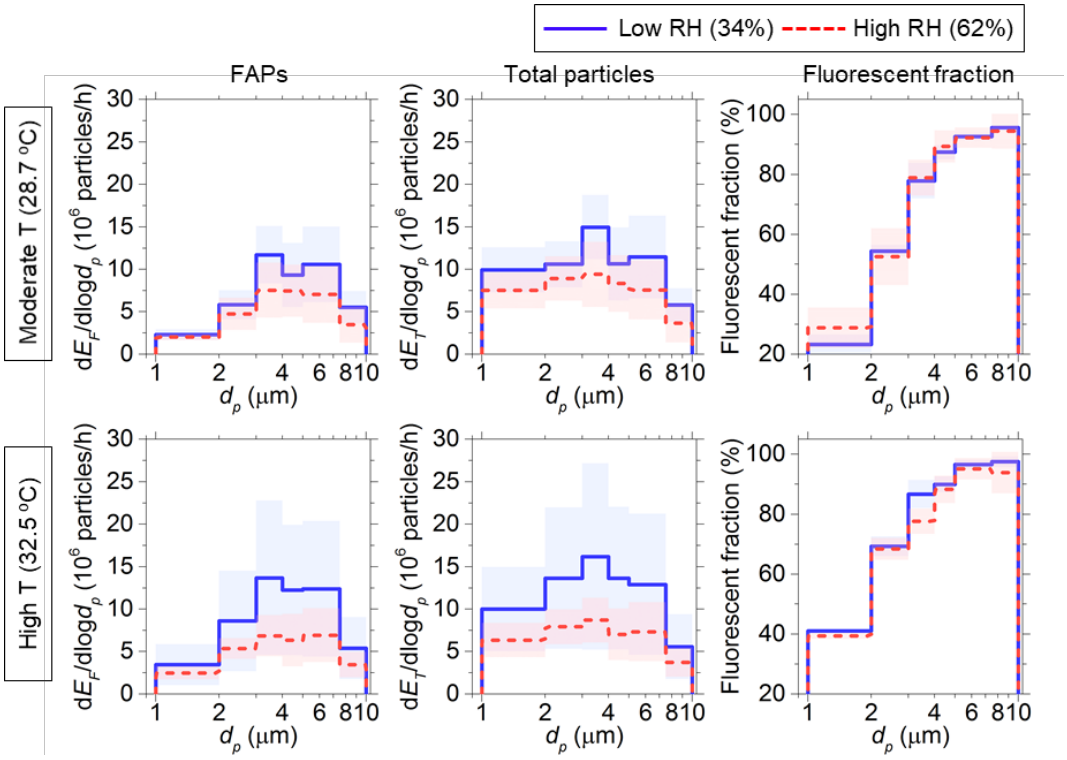


Figure 4. Influence of age on per person size-resolved number emission rates of FAPs and total particles, and fluorescent fraction during periods when the volunteers were sitting. The lines represent averages of all available data within each size bin for the given experimental conditions (*N*=6 (sitting periods per condition); see Table 1). The shaded areas represent standard deviations.

Influence of T and RH. Figure 5 compares the size-resolved number emission rates of fluorescent and total particles from seated young adults in 2×2 experiments with two different temperature (moderate and high) and RH levels (low and high), "long" clothing and no ozone in the chamber. Increased RH from 34 to 62% was associated with 30-60% lower FAP and total particle emissions from human volunteers at both 28.7 and 32.5 ºC (see also Table 1). Elevated RH results in increased capillary adhesion forces acting on particles,[99,100] which could lead to decreased clothing-associated

particle emissions.[101] However, elevated RH is also associated with decreased electrostatic adhesion of particles.[102] There is inconsistent evidence for the influence of RH on particle emissions reported in the literature. Zhou et al.,[46] found an insignificant influence of humidity on human-body particle emissions. In our study, humid conditions were associated with higher skin water content at both moderate and high T (Figure S7). Elevated skin wetness may enhance the adhesion of particles onto the skin and reduce the rate of shedding. We observed a negative correlation between skin water content and emission rates of total particles (r=-0.589, p=0.02) and FAPs (r=-0.393, p=0.13), as shown in Figure S8.

The influence of T on human emissions of fluorescent and total particles changed with RH (Table 1 and Figure S9). At low RH, high T (32.5 ºC) was associated with 50% higher FAP emissions compared to moderate T (28.7 ºC). At high RH, the difference between the two temperatures was insignificant. High air temperature was associated with an increased proportion of fluorescent particles in total particles regardless of RH. Due to the lack of replicate experiments for these conditions (except for the moderate T and high RH condition), these results should be interpreted with caution.



396

397 **Figure 5.** Influence of relative humidity (RH) on per person size-resolved number emission rates of
398 FAPs and total particles, and fluorescent fraction during periods when the volunteers were sitting.
399 The lines represent averages of all available data within each size bin for the given experimental
400 conditions (N=6 (sitting periods per condition; see Table 1) for the moderate T & high RH condition,
401 and N=3 for the other conditions). The shaded areas represent standard deviations.

402 ***Comparison with Related Studies***

403 Table 2 compares the human emission rates of total particles and FAPs in this study
404 with those from three similar chamber-related studies.[25,45,78] The emission rates
405 reported here were generally higher, but within the same order of magnitude. The
406 disparities can be attributed to differences in study conditions (skin coverage with
407 clothing, clothing material and cleanliness, subject characteristics, vigor of body
408 movement and particle resuspension from the flooring), and in the instrumentation used.

409

410 **Table 2**. Comparison of per person number emission rates of total particles within 1-10 μm range

411 ($E_{T,\ 1\text{-}10}$), total particles within 5-10 μm range ($E_{T,\ 5\text{-}10}$) and FAPs within 2.5-10 μm range ($E_{F,\ 2.5\text{-}10}$)[a]

412 with three other studies.

	$E_{T,\ 1\text{-}10}$ (10^6/h)	$E_{T,\ 5\text{-}10}$ (10^6/h)	$E_{F,\ 2.5\text{-}10}$ (10^6/h)	Remarks on study conditions and instrumentation
This study [b]	15±4	6.2±1.4	9.2±2.8	• Four seated young adults, freshly laundered long clothing and socks, no ozone, regular activity, reduced resuspension from the floor (stainless steel open mesh flooring) • WIBS NEO (Optical diameter sizing, three fluorescent channels)
Licina et al.[78]	8±3	--	--	• One seated adult, freshly laundered long clothing and shoes, ozone not specified, moderate movement, minimized resuspension from the floor (sticky doormat and floor cleaning with a sticky roller) • Grimm 11-A (Optical diameter sizing, no fluorescent channel)
	12±2	--	--	• Same as above, but intensive movement
Bhangar et al.[45]	--	1.6±0.7	0.9±0.3	• One to eight seated adults, clothing not specified but in shoes, ozone not specified, regular activity, resuspension from the floor unaltered • Ultraviolet Aerodynamic Particle Sizer (UV-APS, aerodynamic diameter sizing, one fluorescent channel)
	--	0.7±0.4	0.4±0.2	• Same as above, but minimized resuspension from the floor (floor covered with clean antistatic strips)
	--	4.6	2.4	• One seated adult, clothing not specified but in shoes, ozone not specified, intensive movement, resuspension from the floor unaltered
Li et al.[25]	32±32	1.9±1.8	8±8	• One adult, normal living conditions in a bedroom, using a laptop most of the awake time • WIBS NEO (Optical diameter sizing, three fluorescent channels)

413 [a] Size range adapted to match data from the other studies.

414 [b] Refers to emissions during sitting periods in the benchmark case, first row in Table 1.

415 ***Study Limitations***

416 Several study limitations need to be considered. The activities of the human volunteers
417 in the chamber were not strictly regulated. Variation in body movements and
418 manipulating desk items can influence particle emissions.[78] Another limitation was the
419 poor control of the air temperature in the chamber. This was caused by the relatively

high outdoor air temperatures, heat generated by the volunteers, low ventilation rates with 100% outdoor air and an undersized cooling system.[67]

Particles detected in the chamber may also originate from human expiratory emissions. Our measurements would likely underestimate the contribution of exhaled droplets due to their drying in the instrument. However, as the volunteers were all healthy (not sneezing or coughing inside the chamber) and we observed limited speech between them, the exhaled particles were likely of primarily submicron size.[102] In addition, a previous study demonstrated that relative to typical office work, human emissions of supermicron particles are considerably (~27 fold) lower during a sitting-still scenario, when particles are mainly released from breathing.[78] Hence, the expiratory particle emissions are not expected to play an important role compared to the coarse-particle emissions from skin and clothing in this study.

Although the WIBS enables the study of dynamic bioaerosol processes, it lacks information on biological specificity, and thus the reported FAPs may include fluorescent particles originating from abiotic sources, as detailed in Table S5. Further research is warranted to elucidate the contribution of skin- and clothing-associated non-biological fluorescent particles to FAP emissions.

Implications and Future Outlook

Our results contribute to the scarce body of literature on human emissions of particles including FAPs. Our results can improve coarse-mode particle modelling and exposure estimation indoors, and our understanding of the mechanisms of particle release associated with human bodies. The results indicate that human skin and clothing can be a potent source of particles, including FAPs. The 1-10 μm particle and FAP emission rates of human skin and clothing (5.3-16×10^6 FAPs/h, and 7.5-20×10^6 particles/h) are

444 generally 1 to 3 orders of magnitude lower than, or comparable to (depending on
445 activities), other major indoor sources, such as cooking (24-45 mg/h total particles, and
446 7.5-19.2 mg/h FAPs),[103] vacuuming (~50×10^6 FAPs/h) and making the bed (~150×10^6
447 FAPs/h),[24] and walk-induced resuspension (10^{-4}-10^2 mg/h total particles,[66,81,104] and
448 120-950×10^6 FAPs/h).[79] Nonetheless, human emissions are continuous, releasing
449 coarse particles and FAPs directly into the perihuman space, which magnifies the
450 relative effect that human skin and clothing exert on daily exposures.

451 When considering human emissions of particles, the influence of source proximity on
452 exposure should be taken into account. Enhancement of particle concentrations in the
453 breathing zone compared to room-average levels, termed "personal cloud" effect,[79,105]
454 should be considered when estimating human exposure to particles, particularly when
455 it comes to emissions from humans themselves. This is especially the case in actual
456 indoor environments, which often manifest strong air pollution gradients. The
457 characterization of the "personal cloud" effect of particles, including bioaerosols
458 associated with human emissions, is an important research avenue for further
459 exploration. Additionally, the health implications of human-associated bioaerosols
460 remain unknown.

461 Since the human body is almost continuously in close contact with clothing, the effect
462 of clothing on human emissions and exposures to particles deserves attention. Clothing
463 fit, texture, material, thickness, weave, dye and permeability, as well as wear, care, and
464 storage practices are expected to influence the acquisition, retention, and transmission
465 of particles.[59] Further research on the mechanistic processes driving FAP emissions
466 from skin and clothing is warranted. As shown, skin coverage with clothing, skin
467 properties (e.g. skin moisture) and age further influence emissions of FAPs. The impact

of the dynamic nature of various indoor environmental parameters on particle uptake and release from skin and clothing, and consequent exposures, deserves attention.

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Supporting Information Available

Details of experimental settings, quality control, and supplemental results can be seen in the Supporting Information.

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