ScienceDirect

Building and Environment

Volume 282, 15 August 2025, 113324

Impact of 3D printing on indoor particulate matter and volatile organic compounds in educational environments

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Highlights

- 3D printing temporarily increased ultrafine particles in classrooms during printing.
- 3D printing elevated concentrations of specific VOCs near printer locations.
- Up to 80 % of the hazardous chemicals detected in classrooms could be associated with 3D printers during use.
- Use of one or two low-emitting 3D printers in a classroom tended not to pose severe health risks.

Abstract

3D printing has been widely used in K-12 schools and universities as a tool to facilitate education. However, studies have shown that material extrusion 3D printing is a source of airborne particulate matter (PM) and volatile organic compounds (VOCs) that could pose health concerns to the occupants indoors, especially to vulnerable populations like children. This study evaluated the impact of low-emitting 3D printers and materials on indoor air quality in various educational settings that were not initially designed or operated for emission sources like 3D printers. The results showed minimal to moderate levels of ultrafine particles in printer rooms, but they were transient based on printing schedules. 3D printing associated VOCs such as aldehydes, hydrocarbons, aromatics and siloxanes were detected, and many showed elevated concentrations in printer rooms. 3D printing could be a source of hazardous chemicals in classrooms, in addition to other common indoor sources like cleaning and housekeeping products and personal care products. Overall, normal use of one or two validated low emitting 3D printers and materials tended not to pose significant health risks in classrooms, since measured PM and hazardous chemical levels were below indoor recommended levels for most scenarios. However, room conditions, ventilation settings, and occupant activities could also be important factors affecting indoor PM and VOC levels and should be monitored for safe environments.

Previous Next

Keywords

Fused filament fabrication; Particle; VOC; Indoor air quality; Exposure; School

1. Introduction

Three-dimensional (3D) printing has been widely used in educational settings such as K-12 schools, universities, makerspaces, libraries, and special education settings, which is used to teach students and educators about 3D printing (3DP), facilitate design and creativity skills and methodologies, produce artefacts that aid learning, and create assistive technologies [[1], [2], [3], [4], [5]]. Specifically in schools, 3DP is typically used to support science, technology, engineering, and mathematics (STEM) education with low-cost and consumer-level desktop 3D printers [1,[6], [7], [8]]. The most commonly applied 3DP technology in education settings is material extrusion, also called fused filament fabrication (FFF) or fused deposition modeling (FDM™) 3DP [4,9,10] which heats and extrudes a filament shaped material, typically made of thermoplastics, to build a 3D object. However, during printing when the filament is heated, airborne contaminants such as particulate matter (PM) and volatile organic compounds (VOCs) are released [[11], [12], [13], [14]]. Due to the discrepancy between raw material chemical compositions and observed emissions, it is important to characterize emissions and exposures in real printing scenarios [12,15,16]. Previous

studies have reported high levels of ultrafine particles (UFPs, smaller than 100 nm in size) and numerous VOCs emitted from FFF 3DP with various filament materials and different printing conditions [11,12,14,[17], [18], [19], [20], [21], [22], [23], [24]]. Among the most abundantly used thermoplastic materials in education settings [2,4,10,25] acrylonitrile butadiene styrene (ABS) materials tends to have higher PM and VOC emissions than polylactic acid (PLA), likely associated with the properties of the bulk filament materials, additives, and operating temperatures [11,12,17, [19], [20], [21], [22], [23],26,27].

Exposure to PM is associated with short-term and long-term health effects, particularly on respiratory and cardiovascular systems, which could induce or aggravate allergic reactions, asthma, and other associated diseases [[28], [29], [30], [31]]. Specifically for UFPs, due to the small size and high mobility, they can penetrate cells and reach multiple organs, deposit in the alveoli and cause chronic bronchitis, reach the blood, and affect central nervous system [[28], [29], [30], [31], [32], [33], [34], [35]] especially affecting vulnerable population like children [29,30,36]. In addition, among the most detected VOCs from FFF 3DP, numerous odorants, irritants, carcinogens, and reproductive and developmental toxins have been identified, such as styrene, formaldehyde, benzene, ethylbenzene, and acetaldehyde [12,14,15,24,37,38]. Therefore, exposure to the mixture of 3DP emissions could potentially induce adverse health impacts. Laboratory in vivo and in vitro studies have shown exposure to 3DP emissions could induce various effects, from minimal pulmonary or systemic toxicity to inflammatory responses, cytotoxicity, metabolomic changes, and oxidative stress, depending on printing materials and toxicological assessment methods [16,[39], [40], [41], [42]]. Human studies showed that the use of 3D printers may be associated with headaches, irritation and respiratory symptoms [[43], [44], [45], [46]].

Previous studies monitoring exposure levels in university makerspaces have shown the operation of 3D printers resulted in two to ten times higher levels of particle concentrations compared to background levels, with the majority attributed to UFPs [4,47,48]. Exposure to elevated levels of particles could result in increase of deposited dose in lungs [4]. The monitored VOC concentrations were generally low in makerspaces and university laboratories, including aldehydes, acrylates, and other VOCs [25,49]. However, makerspaces, including engineering labs that are designed for such activities are typically equipped with enhanced ventilation systems and sometimes dedicated local exhaust that could effectively reduce exposure levels [25,38,50]. While in K-12 schools, the rooms and buildings where 3D printers are located may only be designed for normal class use, and some may lack sufficient ventilation or natural ventilation options. Good indoor air quality in classrooms is important for the health, performance, and comfort of students and teachers [51]. Although many schools have switched to PLA materials with consideration of emission levels, other materials are also in use [2,4,10,25,47,48]. In addition, emissions from printing with PLA may still introduce or elevate indoor pollutant levels, depending on operating conditions and environmental settings. There are limited literature assessing biological effects in real world exposure to 3DP emissions, our pilot study showed that after exposed to particles collected in classroom during 3D printing, cells showed altered metabolic pathways for both PLA and ABS materials [52]. These pathways implicate

oxidative stress, DNA damage, and inflammation associated with exposure to airborne particles emitted from 3DP [52]. Therefore, assessing exposure hazards in schools is essential for occupants, especially children. A summary of discussed studies above is listed in Table A.1.

This study conducted air measurements in several K-12 schools and one university to monitor indoor air quality for PM and VOC levels associated with 3D printing. Specifically, this study identified several FFF 3D printers and filaments that had been validated as low emitting conditions based on standardized chamber testing [53]. These printers were deployed in different school sites that were not originally designed to manage the emissions from 3D printers. Indoor airborne PM concentration and size distribution were monitored online with particle sizers; VOC samples were collected and quantified using analytical methods. The impact of 3DP was evaluated by comparing to background levels when no printers were operating; control locations where no 3D printers existed were used as references. Chamber characterizations provided baseline information for field measurements and both results were compared to understand the agreement and discrepancy between the two methods. In addition, measured indoor VOCs were classified into source categories and the impacts of 3DP and other common indoor sources were assessed. Finally, the observed PM and VOC levels were compared to indoor air quality reference levels for health implications.

2. Methods and materials

2.1. Monitoring sites and conditions

Indoor air quality was monitored in various sites representing different educational settings, which included a STEM classroom in a high school (HS), a STEM classroom in an elementary school (ES), and a conference room in a university (UNI). The buildings where these rooms located were not originally designed for equipping emission sources like 3D printers, however, the HS STEM classroom was recently renovated with upgraded ventilation systems, and both HS and ES STEM classrooms had a portable air cleaner running continuously. In this study, the room conditions and class schedules remained as normal during indoor air quality monitoring. Windows and doors of the classrooms were closed when possible, to minimize impact from outside of the rooms. Within each monitoring site, two locations were studied; one was close to the 3DP activity and the other was far away from the printer(s). Background (BG) levels were also monitored in the printer room before the 3D printer(s) started to operate. In addition, hallway or a classroom without 3D printers (control) was also monitored as a reference when possible.

Three models of desktop FFF 3D printers (*P1, P2* and *P3*) and two types of commonly used materials (PLA and ABS) were studied. Each printing condition was setup according to the manufacturer's instruction (Table 1). *P11* and *P12* were two printers with the same model deployed in different sites. One ABS filament formula (*ABS1*) and three PLA filament formulas (*PLA1, PLA2* and *PLA3*) were studied. Printers with normal setups were evaluated to represent normal use of printers in schools and other activities in the studied rooms remained unchanged.

Table 1. Site and printing conditions for each visit.

Visit	Site	Printer	Material	Print temperature	Print duration
1	UNI	*P11*	*PLA11* black color	210 °C	3 h
2	UNI	*P11*	*ABS11* black color	245 °C	2.5 h
		P31	*PLA31* natural color	215 °C	2.5 h
3	UNI	*P11*	*ABS11* black color	245 °C	2 h
4	HS	*P12*	*ABS12* orange color	245 °C	2.5 h
5	ES	*P23*	*PLA23* black color	215 °C	3.5 h

Note: two-digit label of printer and material differentiate for manufacturers (first digit) and sites (second digit). For visit #2, two printers were operating at the same time.

2.2. Sampling and analysis methods

Airborne particles were monitored in real time using a NanoScan scanning mobility particle sizer (SMPS, TSI model 3910) and an optical particle sizer (OPS, TSI model 3330). NanoScan measures particles from 10 to 420 nm in size and OPS measures particles from 0.3 to 10 μm. Particle number distributions were reported from NanoScan and OPS with a sampling interval of 1 min for each scan. $PM_{2.5}$ (PM smaller than 2.5 μm) and PM_{10} (PM smaller than 10 μm) concentrations were calculated from the measured number concentrations assuming particles were spherical with unit density (1 g/cm^3) [11,16].

VOCs were collected onto Tenax® TA (60/80 mesh) sorption tubes and then thermally desorbed (Perkin Elmer TurboMatrix 650) into the gas chromatograph-mass spectrometry (GC–MS, Agilent 8890 GC, Agilent 5977B mass selective detector). Generally, individual VOCs with boiling points ranging from 35–250 °C were identified and quantitated according to US EPA Method TO-17 [54]. Total VOC (TVOC) was the sum of individual VOC responses obtained by the mass spectrometer that were calibrated relative to toluene for C_6 to C_{16} range. Sorbent cartridges with DNPH (2,4-dinitrophenylhydrazine) were used to collect low-molecular-weight carbonyl compounds and were analyzed by high performance liquid chromatography (HPLC, Agilent 1260 Infinity) following US EPA Method TO-11A [55]. Details of analysis methods have been described previously [12,53,56]. The limit of quantification (LOQ) is 0.5 μg/m^3. Target collection volume for VOCs was 18 L (0.2 L/min for 90 min) and 45 L for aldehydes (0.5 L/min for 90 min) using pre-calibrated portable vacuum pumps (Sensidyne, models Gilian LFS-113DC and GilAir5). Proximately 10 % of duplicates were taken and analyzed with the same methods for the purpose of QA/QC. A flow chart of site monitoring methods is shown in Fig. 1A.

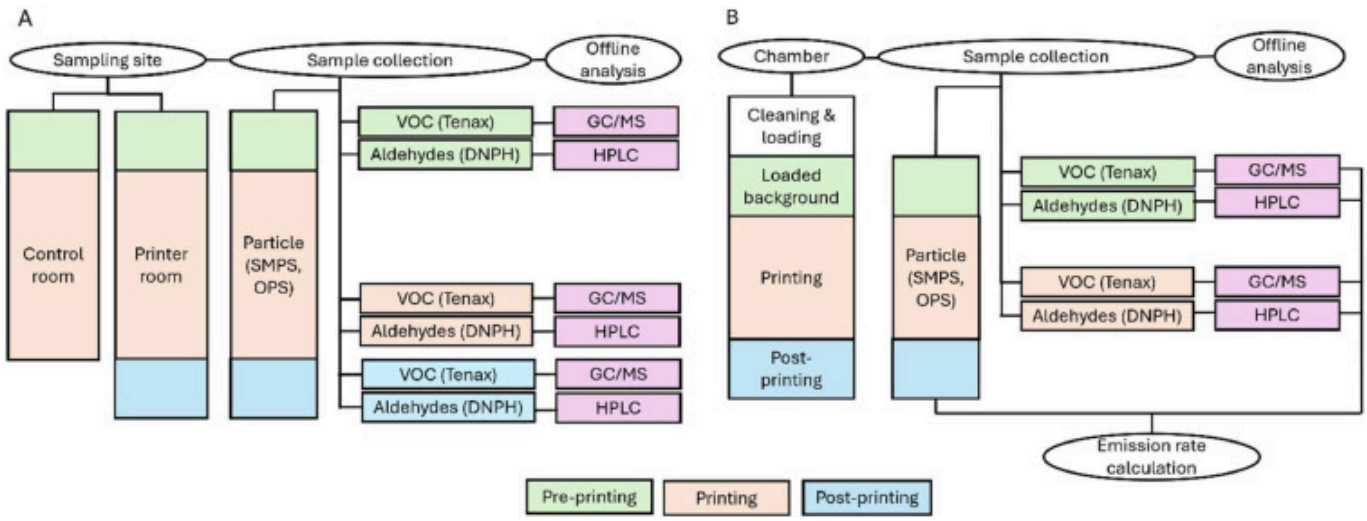

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Fig. 1. Summary of sampling methods used in field study (A) and chamber study (B). Colors indicate different printer operation stages.

2.3. Chamber emission characterizations

Baseline emission characterization of particles and VOCs was conducted using stainless steel exposure chambers based on a standard testing method, ANSI/CAN/UL 2904 [53]. The chamber specifications and setups, validation protocols, and sampling procedures have been described previously [11,12,53,56]. Particle number distribution inside the chamber was measured online with an SMPS spectrometer (TSI model 3938) for 7 to 300 nm size range and an OPS (TSI model 3330) for 0.3 to 10 μm size range. Printing duration for each emission test was 4 h. Particle emission rate (ER, #/h or μg/h) was calculated based on ANSI/CAN/UL 2904 [11,53,57]. VOC and aldehyde samples were collected and analyzed using the same method as in the monitoring sites. A background (BG) sample was taken before the printer started printing and another sample was taken at the last hour of the printing duration. The sampling duration for chamber characterization was 1 hour with the target sample volume of 12 L for VOCs and 30 L for aldehydes. Further, emission rates (μg/h) for individual VOCs and TVOC were calculated based on ANSI/CAN/UL 2904 [11,53,57]. A flow chart of chamber characterization is shown in Fig. 1B

2.4. Statistical analysis

Mann-Whitney U test was used to compare if two groups are different. *P* value smaller than 0.05 was considered statistically significant.

3. Results and discussion

3.1. 3D printing impact on indoor particulate matter

3.1.1. Particulate matter monitoring over time

PM concentrations (both number and mass) were monitored before and during the printing duration. Fig. 2 shows the PM levels in the university conference room with two printers running PLA and ABS materials for 2.5 h. After the printing started, particle number concentration (total N) showed an increasing trend while this trend was not seen for $PM_{2.5}$ or PM_{10}; the highest particle number concentration was observed during printing, which reached over 5000 #/cm^3 (Fig. 2). This indicated that 3DP emitted particles, which are mainly UFPs, did not notably contribute to mass concentrations [11,14,17,19,24]. In fact, the particle size distribution showed the average geometric mean diameter (GMD) for the period when total N concentration was elevated (65.7 nm) was smaller than that for the rest of the printing duration (78.4 nm), and they were statistically significantly different ($p < 0.0001$). In addition, near or far from printer monitoring locations seemed not affecting the observed particle total N or $PM_{2.5}$ levels, which followed the same trend (Fig. 2). Statistical analysis also showed near printer and far from printer observations were not significantly different ($p > 0.5$) for total N and $PM_{2.5}$. One thing to note is after printing stopped, there seemed to be a trend that PM_{10} increased near printer (Fig. 2), which could be associated with the operators near the printer and emissions from human activities [58,59]. As a reference, the concentration in a nearby classroom without 3D printers was 4021±224 #/cm^3, comparable to the background levels in the conference room. Similar trend was found when printing with PLA material at this site (Figure A.1), other than that visit had lower background levels, likely associated with the ventilation was not fully operating early in the morning before class started for visit #2 (Fig. 2). Previous study reported higher particle concentrations during non-working hours, likely due to the ventilation rate being reduced during after working hours [49]. The elevation in particle number concentration showed higher increase but lasted for shorter time (Figure A.1). In addition, the average GMD during the peak of total N was 46.6 nm while that for the rest of the print duration was 76.0 nm. The trend for near and far printer locations was also similar while PM_{10} trend possibly followed the activities of occupants. A previous study measured PM_{10} in elderly care centers and found the average PM_{10} levels in bedrooms were 8–16 μg/m^3 and in living room were 13–24 μg/m^3. The significantly higher PM_{10} levels in living rooms was likely associated with particle resuspension since more individuals were standing and walking in living rooms than in bedrooms [60]. The average PM_{10} levels in this study ranged from 2 to 15 μg/m^3, which was comparable or lower than previously reported in schools (4–8 μg/m^3)[61] and elderly care centers [60] while there were reported cases with PM_{10} above 100 μg/m^3 in schools [62].

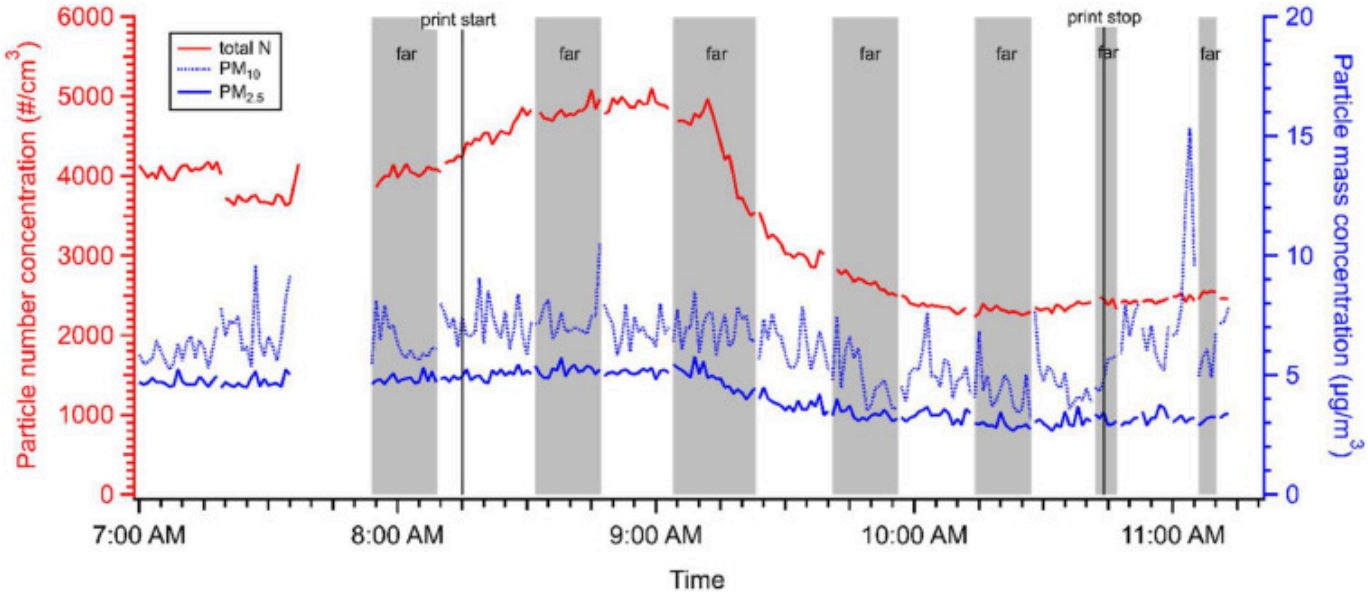

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Fig. 2. Particle number concentrations (total N) and mass concentrations ($PM_{2.5}$ and PM_{10}) as a function of time measured at UNI site with PLA and ABS materials. Monitoring was at near printer (clear area) and far from printer (shaded area) locations. Vertical lines indicate the printing duration.

The results showed 3DP could introduce UFPs in indoor environments during operation, which elevated particle number concentrations while seemed not impacting $PM_{2.5}$ or PM_{10} levels. As a reference, the total N concentration during weekend when no activity was expected was 1077±809 #/cm^3. During printing, the maximum concentration was 3158 (visit #1) and 5104 #/cm^3 (visit #2) for the two visits, which were elevated by 4.04 and 1.25 folds compared to the average background levels respectively. However, the impact of 3DP tended to be temporary in the studied sites, which decreased after a certain time into the printing. This was likely due to a combined effect of emission sources and environmental conditions of the sites. The deployed printers and materials were tested to be low-emitting and were observed with similar trends in particle number counts in chamber experiments. In addition, occupant activities may also contribute to the particle levels indoors, especially PM_{10}.

3.1.2. Difference of particulate matter for each scenario

Considering the entire printing duration, a comparison of particle number and mass concentrations for each scenario is shown in Fig. 3. Particle levels were specific to each site and print condition. Statistical analysis showed that there was significant difference between BG and during printing for $PM_{2.5}$ at all 5 sites, while not all of them showed elevated $PM_{2.5}$ during printing. On the other hand, only two visits showed significant difference in particle number concentrations, which could be driven by the high outliers. In general, printing with ABS material (*ABS1*) seemed to increase $PM_{2.5}$

median levels compared to BG as observed in UNI and HS sites, while printing with PLA material (*PLA1* and *PLA2*) seemed to generate high outliers in total N concentration, which was observed in UNI and ES sites (Fig. 3). The particle levels for printing with two materials showed larger variations. For proximity effects, the UNI far printer location tended to have higher median total N concentrations, while near printer locations tended have higher median total N concentrations at HS and ES sites. This inconsistency was likely associated with the room conditions such as ventilation and air mixing, printing conditions and target measurements, which was also observed previously in field studies [2,38]. However, the differences in particle concentration associated with proximity were only statistically significant for three out of 10 comparisons between near and far printer locations, which seemed to be driven by the high outliers. It should be noted that there were cases when BG levels were higher than those during printing, which indicated that there may be other conditions that affecting indoor PM levels, such as room ventilation and occupant activities. In addition, ES site showed statistically significant changes for all scenarios (Fig. 3), indicating the PM levels and spatial distributions in this site were more sensitive to the change of indoor activities.

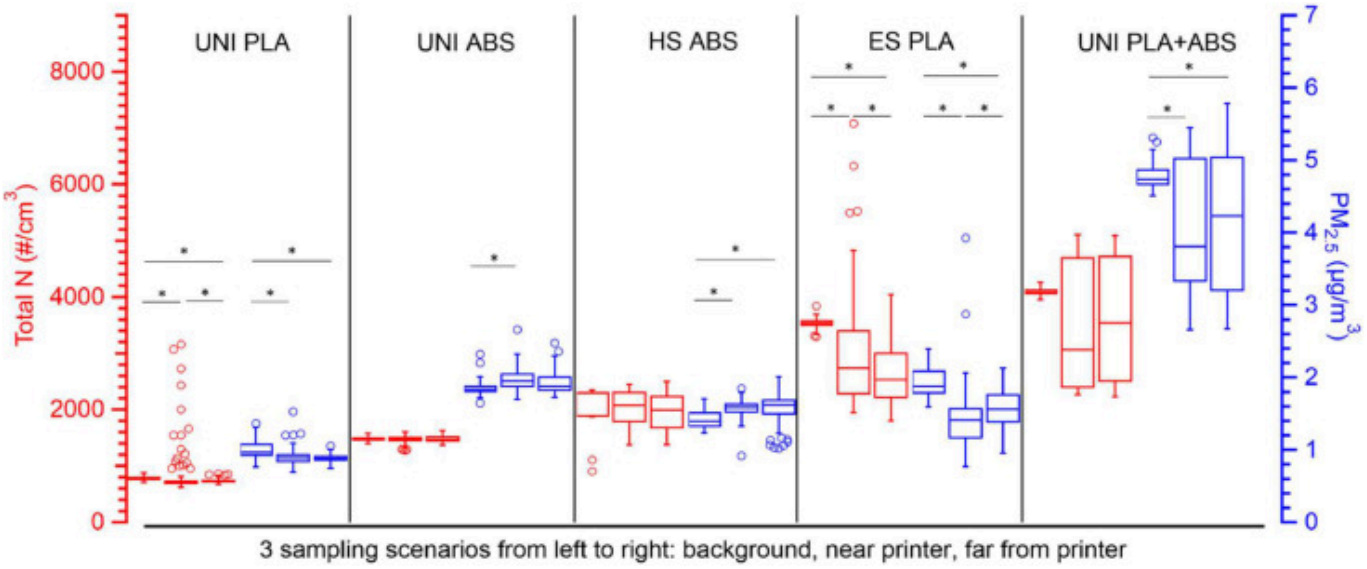

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Fig. 3. Particle number concentration (total N) and mass concentration ($PM_{2.5}$) for different sampling scenarios. Asterisk indicates the difference between two groups is statistically significant with $p < 0.05$.

There were also variations among the size distributions for different printing and site conditions (Fig. 4). The mode of particle size distribution during printing was comparable to that of BG, which indicated that pre-existing particles likely dominated the mode size. The HS and the ES sites had larger variations in size distribution, which pointed to the contribution of particle emission sources; higher variations were seen at ultrafine size range, which was consistent with that FFF 3DP generates mainly UFPs [11]. Specifically, there was an increase in particles smaller than 20 nm compared to BG distributions for both sites, which was likely attributed to 3DP emitted UFPs. The

UNI site consistently showed larger mode sizes, which were approximately 100 nm. For UNI site with PLA and two materials, there was an increase in particle distribution at approximately 10 – 50 nm size range, while ABS material showed an increase at over 70 nm size range. Overall, considering the entire printing duration, the impact of 3DP emitted UFPs tended to increase the proportion of smaller particles (< 50 nm), while the overall particle size distribution may be dominated by the pre-existing particles and site conditions.

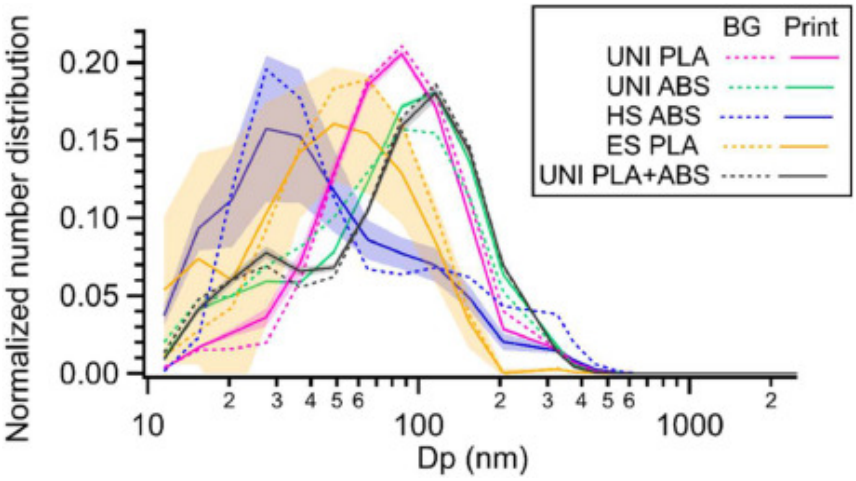

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Fig. 4. Normalized particle number distribution for each sampling site. Data are averaged including both near and far printer locations, shaded area indicates standard error, averaged background (BG) size distribution is as a reference (no standard error).

3.2. 3D printing impact on indoor volatile organic compounds

3.2.1. Total volatile organic compound

TVOC levels in different locations are shown in Table 2. TVOC levels were elevated comparing to BG for all UNI scenarios, with an increase of up to 2.6 folds. The background TVOC was higher for HS and ES sites, which could be associated with ventilation conditions, residual VOCs from cleaning activities, and occupancy activities in the room. For HS and ES sites, TVOC levels were higher at near printer locations, while at UNI site, TVOC could be higher at the far printer location. This inconsistency was also reported for TVOC measured using a photoionization detector (PID) at different locations in an engineering laboratory; average TVOC concentrations ranged from 40 to 300 μg/m^3, which is comparable to this study, however, the maximum TVOC levels could reach over 1000 μg/m^3 [38]. Similar to particle concentration trends, there was no consistent trend for printer proximity impact on TVOC concentration. Decay samples were taken after print stopped, based on the monitoring sites, TVOC concentrations during decay were 34 %–88 % and 25 %–93 % of those during printing for near and far printer locations separately. Specifically, over 50 % of the detected VOCs showed a decreasing trend compared to during printing (92 % for UNI PLA, 51 % for UNI

PLA+ABS, and 65 % for ES PLA). This could be associated with the cease of emission from 3DP and overall dilution of emissions. However, there were also VOCs that showed increasing trend during decay than printing, which accounted for 5 % for UNI PLA, 49 % for UNI PLA+ABS, and 29 % for ES PLA. This could be related to the release of VOCs when opening printer doors to remove printed parts. In addition, outdoor TVOC levels were measured as a reference, which was much less than indoor levels (below 10 – 30 $\mu g/m^3$).

Table 2. Total VOC (TVOC) levels at each sampling location. Blank indicates no samples taken.

TVOC ($\mu g/m^3$)	**BG**	**Printing near**	**Printing far**	**Decay near**	**Decay far**
UNI PLA	64.5	124	168	41.7	41.6
UNI PLA+ABS	31.5	47.0	32.4	25.9	30.0
UNI ABS	22.1	25.4	38.9		
HS ABS	358	313	311		
ES PLA	579	550	444	485	383

3.2.2. Most abundantly detected volatile organic compounds

There were over 200 individual VOCs detected from the samples that were above LOQ and there were more VOCs detected during printing than BG. Considering VOCs detected from both near and far printer locations, the top 10 VOCs with the highest concentrations are listed in Fig. 5. There were a few VOCs that were more likely to have higher BG levels than during printing (Figure A.2). For instance, decamethylcyclopentasiloxane (D5), formaldehyde, hexanal, α-pinene, and propylene glycol (1,2-propanediol), which were higher in BG than during printing for over 50 % of the detected cases, are typically associated with cleaning products and personal care products. VOCs with elevated concentrations during printing that could be attributed to 3DP included acetaldehyde, toluene, 1-butanol, octamethylcyclotetrasiloxane (D4), hexanoic acid, 2,2,4-trimethyl-1,3-pentanediol monoisobutyrate, nonanal (nonyl aldehyde), and propanal. The concentrations at near printer locations of these most abundantly detected VOCs are shown in Fig. 5, grouped according to the comparison of concentrations at near and far printer locations. Among the listed 27 VOCs, 10 of them were detected from all sites and 5 were unique to one specific site. The bottom five VOCs in Fig. 5 that were unique to a specific monitoring site while not necessarily detected in low concentrations, were less likely to be associated with 3DP, but the cleaning products and household care products such as cleaners, air fresheners, and laundry detergents used daily at each site. The coefficient of variation between near printer and far from printer locations were typically (75 percentile) within 30 %. For the most abundantly detected VOCs, there were 57 cases where VOC concentrations were elevated at near printer location and 32 cases were elevated at far printer

location, which indicated that 3DP tended to have larger impact on VOC levels at the near printer location.

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Fig. 5. The top 10 detected VOCs during printing from each site with their concentrations at near printer location. Red indicates the concentration at near printer location was higher than that at far from printer location; blue indicates the concentration at near printer location was lower than that at far from printer location; gray indicates not detected or concentration was below LOQ.

The comparison of VOCs in printer rooms with those in control rooms was more complex. Although there was no 3DP activity in control rooms, VOC levels could be elevated by other indoor sources. Therefore, the control room VOCs were only used as informative references. In general, control rooms had comparable or fewer individual VOCs detected. For the top 10 VOCs at each site, UNI site showed approximately 50 % of similarity in the printer and control rooms, which were next door to

each other; ES site shared only 3 VOCs in common between the two rooms, likely due to they were in different floors of the building; HS site had the same top 10 VOCs for the two rooms that were next door to each other, which may indicate the building recirculated air through the ventilation system and distributed airborne contaminants in different rooms.

3.2.3. 3D printing associated volatile organic compounds

VOCs detected from the chamber study were used as a baseline to evaluate the impact of 3DP on indoor VOCs, comparison was paired for the same printing conditions. In general, PLA materials emitted less VOCs; 47 % to 60 % of them were also found in the monitoring sites with the same printing conditions (Fig. 6). ABS materials generated more varieties of VOCs, while only 11 % to 25 % of them were found in field monitoring, which indicated that some low emitting compounds became below LOQ when emitted in larger and more diluted environments. There was 9 % to 41 % of the field detected VOCs that were potentially from 3DP, since they were also detected in the chamber study (Fig. 6). Specifically for UNI site with two materials, 18 out of 44 VOCs in the field were likely associated with 3DP emissions, with 10 compounds detected from both materials (Fig. 6E). However, this also indicated that there were other VOC sources at each site that could contribute to up to 90 % of the numbers of indoor VOCs detected. One thing to note is that the interaction among VOCs may generate new products or change the chemical compositions due to the complex conditions in real-world scenarios compared to well-controlled chamber environment.

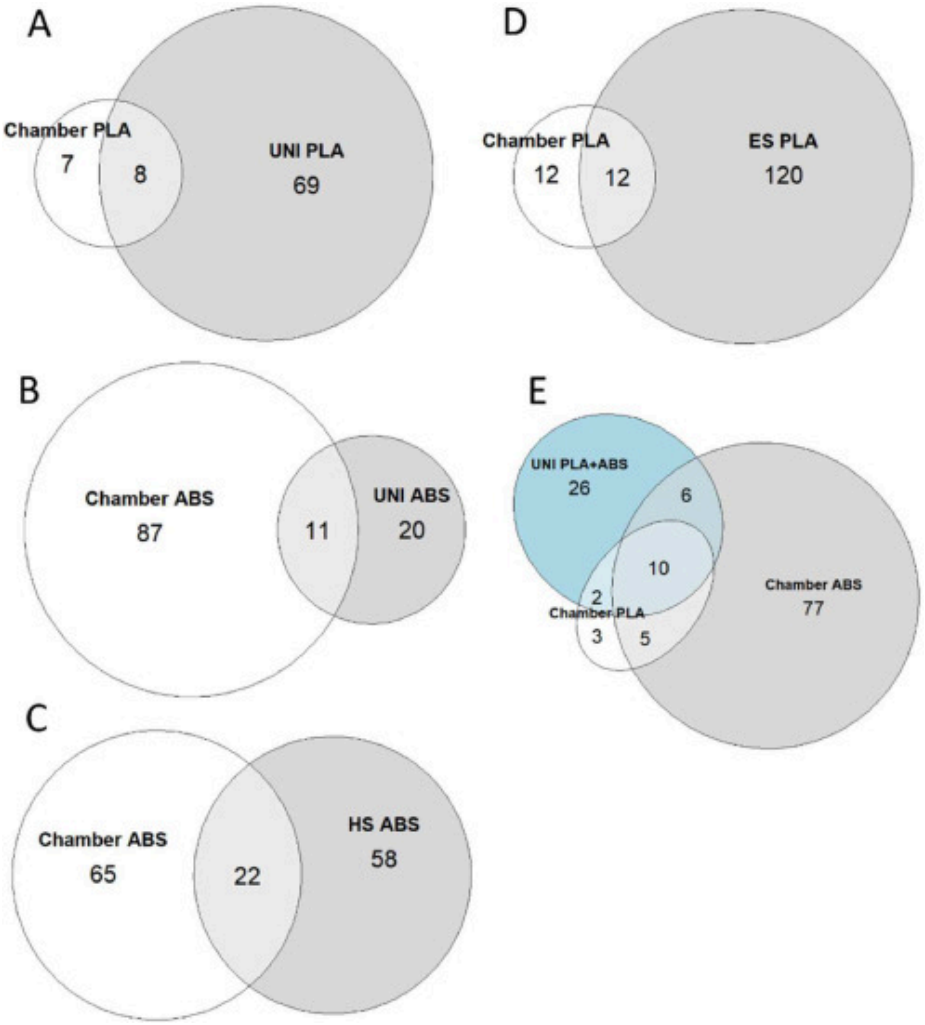

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Fig. 6. Euler plot for the numbers of VOCs detected in monitoring sites (labeled as site and material) overlapping with those from chamber evaluation (labeled as chamber and material) for the same printing conditions.

Fig. 7 summaries the common VOCs identified from baseline chamber evaluation, listed in descending order according to concentration level and detection frequency. In general, 70 % to 97 % of the common VOCs had higher concentrations during printing except for HS site, which could be linked to the emissions from 3DP. Specifically, acetaldehyde and D4 (octamethylcyclotetrasiloxane) were consistently higher during printing compared to BG in all five sites, increased for 4 % to 62 % and 20 % to 73 % separately. Other elevated VOCs associated with 3DP included nonanal, decanal, styrene, hexamethylcyclotrisiloxane (D3), tetradecane, and toluene. In addition, 2-ethylhexanol concentrations increased with printing PLA filaments and benzaldehyde with ABS filaments. Although less frequently detected, ABS was likely a source of 2,2,4,6,6-pentamethylheptane and 4-hydroxy-4-methyl-2-pentanone, and PLA a source of dodecamethylcyclohexasiloxane (D6), dimethylsilanediol, undecane, tetradecamethylcycloheptasiloxane (D7), and α,α-

dimethylbenzenemethanol (Fig. 7). Formaldehyde and D5 (decamethylcyclopentasiloxane) were detected with relatively higher concentrations, however, their concentrations during printing were lower than BG for some scenarios, which indicated that the contribution from 3DP may not be as significant as other sources in the background for specific scenarios. Formaldehyde has been reported previously being detected in high concentrations in schools [51,63]. For HS site with ABS material, there were 48 % of the common VOCs that had lower concentrations during printing, which was 6 %–30 % at other sites. This pointed to potential sink of 3DP emissions and emissions from other sources at HS site, which may align with that the HS control room showed similar VOC profiles as the printer room that could be associated with ventilation settings of the building.

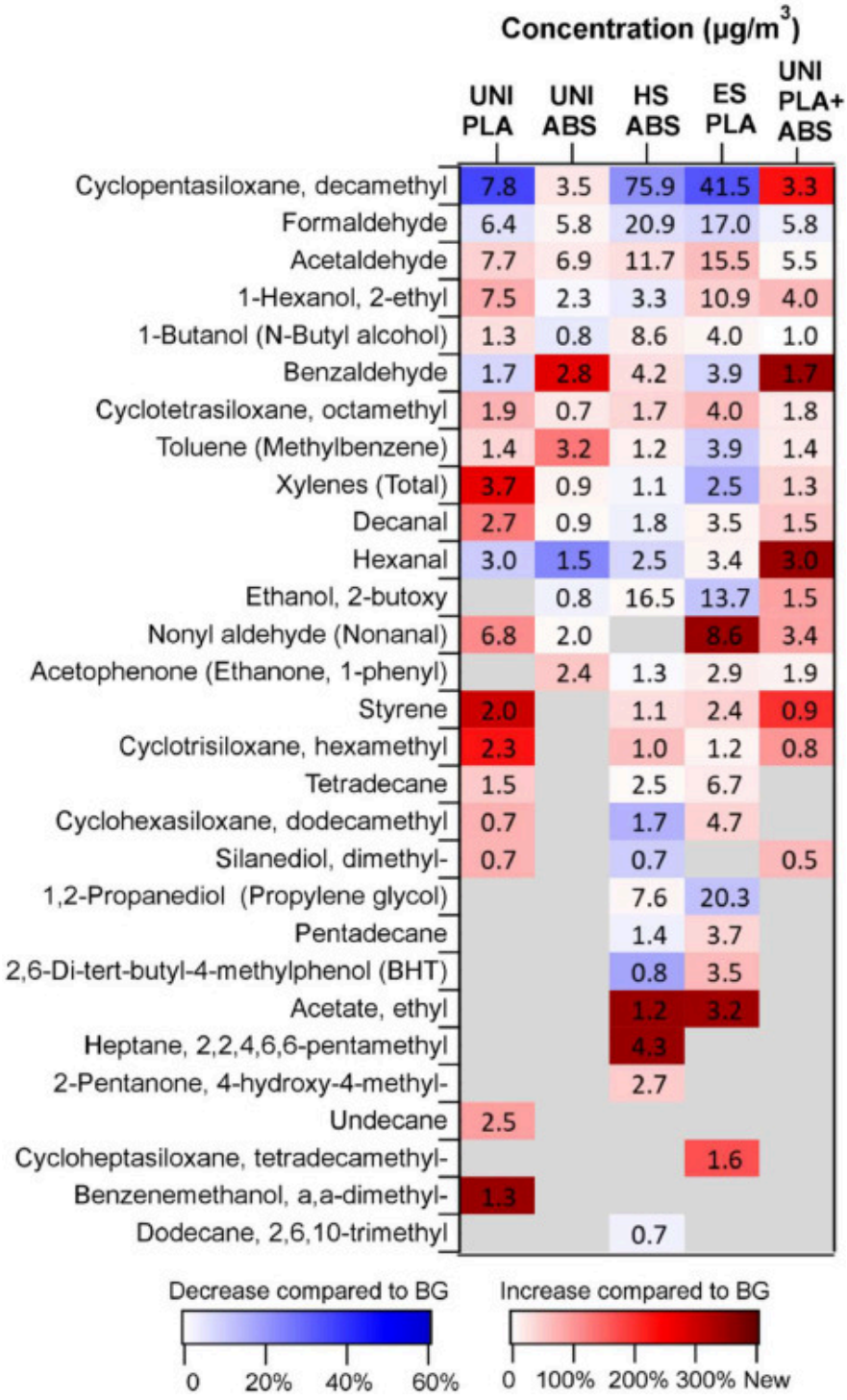

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Fig. 7. Common VOCs detected in chamber and field studies with their maximum concentrations during printing on site listed. Gray indicates below LOQ for the site; red indicates the percentage of concentration increased during printing compared to background (BG); “New” indicates below LOQ in BG; blue indicates the percentage of concentration decreased during printing compared to BG.

For 3DP associated VOCs listed in Fig. 7, 41 %–75 % of them showed decreased concentrations during decay when 3D printing stopped, while there were also VOCs with elevated concentrations during

decay. Acetaldehyde, styrene, benzaldehyde, and nonanal were likely continuously off-gassing from the printers during cooling down and opening printer doors to remove printed parts since their decay concentrations were the highest among all scenarios including in the control rooms. If VOCs had low decay rates they could linger for extended time and be mixed and spread throughout the room. On the other hand, formaldehyde, hexanal and tetradecane may be associated with other sources since the control rooms also had similar levels. A previous study reported that average formaldehyde concentrations were 19–55 μg/m^3 in primary schools without 3DP [63]. Overall, impact of 3DP declined as TVOC concentrations also decreased during the decay period (Table 2). In addition, the most abundantly detected VOCs in control rooms, such as 1-butoxy-2-propanol, propylene carbonate, 1-methoxy-2-propanol, acetic acid, 2-phenoxyethanol, 2-butoxyethanol, d-limonene, 2-(2-butoxyethoxy)ethanol, D5, 2-(2-ethoxyethoxy)ethanol, 2-(hexyloxy)ethanol, α-pinene, and hexanoic acid, were more likely to be associated with emission sources other than 3DP, except that D5 and 2-butoxyethanol were also detected from 3DP.

3.3. Health implications

In general, indoor $PM_{2.5}$ was below 5 μg/m^3 and PM_{10} below 10 μg/m^3 at each site and are below the US EPA National Ambient Air Quality Standards (NAAQS) daily criteria (35 μg/m^3 for $PM_{2.5}$ and 150 μg/m^3 for PM_{10}) and World Health Organization (WHO) daily air quality guideline criteria (15 μg/m^3 for $PM_{2.5}$ and 45 μg/m^3 for PM_{10}) [64,65]. The impact of emitted particles from 3DP on indoor air quality seemed limited due to the emissions were mostly UFPs that did not contribute significantly to PM mass concentrations. However, it should be noted that even the concentrations indoors are well below the ambient standard levels, people spend most of their time indoors. Study have found that exposure to particles from low emitting 3DP could induce negative responses in human airway epithelial cells [42,52].

Among the 29 VOCs that are associated with 3DP, 18 were chemicals of concern (CoC), which are listed in indoor air quality and health related guidance documents [[66], [67], [68], [69], [70]] as well as in ANSI/CAN/UL 2904 with emission criteria [53]. Exposure to high levels of CoCs could result in irritation, throat dryness, allergies, and respiratory health issues [51]. The deployed 3D printers and materials in this study were tested following the standard with all VOC emissions within the suggested criteria. These chemicals of concern were still detected in the field, with 16 out of the 29 3DP associated VOCs detected in over half of the visits. The most frequently detected chemicals of concern included formaldehyde, acetaldehyde, D5, 1-butanol, toluene, D4, and xylenes, which were detected in all field samples. Specifically, formaldehyde, acetaldehyde, and styrene are carcinogens and toluene is a developmental toxin [66,67] other compounds can be irritants and odorants [71]. There were no VOCs exceeding the recommended indoor levels except for formaldehyde, where HS and ES sites showed concentrations higher than recommended indoor level (9 μg/m^3) [53,69]. It should be noted that VOC results also depend on measurement methods. Zontek et al. reported all chemical measurements below LOQ using occupational sampling methods that mainly aim at screening for occupational references in ppm levels typically [25]. These

methods may not be applicable for measuring low concentrations in indoor environments like school classrooms with susceptible populations.

The real-world indoor VOC compositions are more complex than chamber testing and can be attributed to various sources. US EPA ChemExpo Knowledgebase (version 0.1.1, www.comptox.epa.gov/chemexpo/ ↗) collects chemical data in public documents that are relevant to exposure assessment and groups them into 34 main Product Use Categories (PUCs). In this study, to understand the potential sources of the detected VOCs specifically in schools, PUCs were reorganized into 6 source categories. Category 1 was specifically defined in this study for 3DP based on the discussion in Section 3.2.3. Category 0 indicated limited data or no information in the database. The rest categories were grouped by sources, which were category 2 for safety, cleaning and household care products, category 3 for personal care products, category 4 for home maintenance products, and category 5 for the rest of products, which included construction and building materials, electronics and appliances, and other consumer products and materials. Source apportionment was conducted for VOCs detected during printing, including 65 VOCs in category 0, 29 in category 1, 38 in category 2, 15 in category 3 and category 4 each, 22 in category 5, in total of 184 VOCs. Fig. 8 shows the distribution of potential VOC sources at each site, the percentage was calculated as the sum of VOC concentrations in a specific category over the sum of all VOC concentrations at a specific site. Detected VOCs were mainly in category 1 through 4, with the rest accounting for 4 % to 26 % of the VOC compositions. It is noted that 4 % to 18 % of the detected VOCs had limited information on ChemExpo database, which pointed to the potential knowledge gap that these compounds tended to be less frequently reported. Except for ES PLA site where personal care products (category 3) were the most abundant VOC source, 3DP associated VOCs accounted for the most portion of VOC compositions (33 % to 58 %). In addition, cleaning products (category 2) and personal care products (category 3) were also main sources of VOCs indoors. Considering only chemicals of concern, the proportion of category 1 (3D printing) increased for all sites, accounting for 28 % to 79 % of the overall chemicals of concern (Fig. 8). While 80 % of the cases associated with the rest categories showed decreasing trend when considering only chemicals of concern. This indicated that 3DP VOCs contained more health concerning compounds than those from other sources like cleaning and personal care products. For ES PLA, personal care products associated VOCs accounted for over 60 % of the chemicals of concern, while approximately 99 % of them was ethanol. It should be noted that the sources of indoor VOCs are complex and some 3DP VOCs were also associated with other source categories (Fig. 8) [72]. For instance, siloxanes (D5, D3, D4, and D6) are also likely from personal care products; nonanal, decanal, and benzaldehyde can be associated with cleaning products; formaldehyde, xylenes, and toluene can be associated with home maintenance products. Within category 5, most VOCs were listed as laboratory supplies, raw materials, and construction associated, which were industrial and occupational PUCs. In addition to emissions from materials and products, human metabolism can be a source of acetone, methanol, ethanol, and monoterpenes [72].

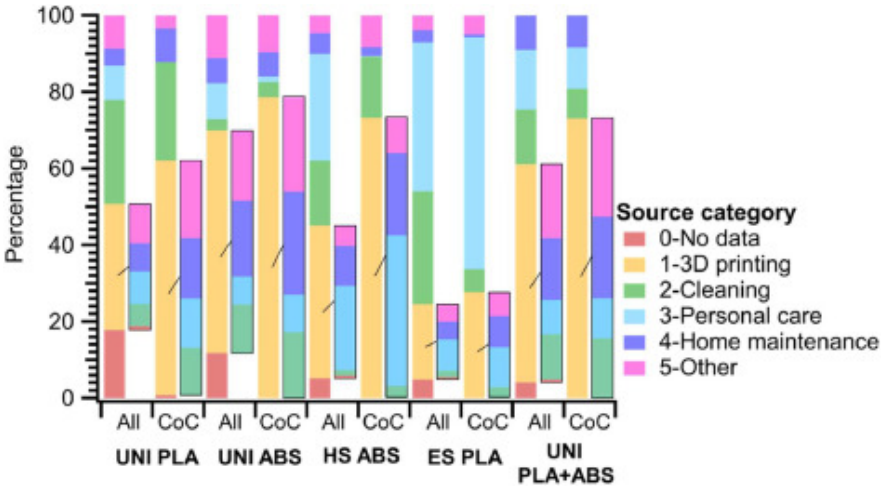

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Fig. 8. Source apportionment of detected VOCs at each site, grouped by all VOCs (All) vs. chemicals of concern (CoC). Inset next to category 1 indicates the breakdown of 3D printing associated compounds into other potential source categories.

3.4. Limitations

A recent study validated that the combined effect of mechanical ventilation and portable air cleaners complicated the air flow dynamics and impacted the distribution of airborne PM indoors [73]. In this study, there was limited control on the settings and activities on site during visits, which could contribute to the variations of measured particle and VOC levels. For instance, air cleaners could be a sink of particles and VOCs, and air fresheners and occupants could be sources of particles and VOCs. Classroom settings were not altered to maintain normal school operation scenarios while more detailed record of participant activities could be taken. The ventilation settings at each site were likely different and could be important parameters affecting the observed concentration levels and trends. However, no information of building dimensions or operations was available, which should have been measured on site. Since this information, such as HVAC system specifications and room volumes, can be important to interpret air distribution and pollutant transportation, as well as to conduct detailed airflow modeling. Source apportionment of particles is challenging without understanding the chemical composition of them. In this study, potential sources of particles were discussed regarding particle sizes. To better understand particle emissions from 3DP in real world scenarios, a complex fluid dynamic model may also be needed. Indoor VOC emission sources in schools can be complex while were not evaluated individually in this study. The source apportionment analysis only assigned one category (the most abundant category) for each VOC while some VOCs fall in multiple categories.

4. Conclusions

In this study, indoor PM and VOC concentrations were monitored in different educational settings to evaluate the potential impacts of 3DP on indoor air quality. Specifically, desktop 3D printers that were verified as low emitting were deployed and operated in the classrooms that were not designed or operated for emission sources like 3DP. PM monitoring results showed temporarily elevated UFPs in printer rooms. For the printing materials studied, ABS tended to be associated with increase in $PM_{2.5}$ while PLA with increase in particle number counts. The distance to the printer generally had a minimal impact on PM concentrations, probably due to the emitted particles being mixed by the room ventilation systems. Over 200 VOCs were detected in classrooms, among which were a complex mixture of chemicals of concern. Detected VOCs associated with 3DP included aldehydes, hydrocarbons, and siloxanes, such as acetaldehyde, decanal, nonanal, D4, toluene, and styrene. In addition, cleaning and housekeeping products and personal care products were also sources of classroom detected VOCs. It should be noted that indoor contaminant levels also depend on the room conditions, such as room size and ventilation, which impact observed concentrations at different site locations and how quickly they decay after printing ended. Overall, the low emitting 3D printers in this study seemed unlikely to pose significant health risks to occupants in the classrooms with measured concentrations of PM and chemicals of concern below indoor reference levels, except for formaldehyde in some scenarios. One thing noted during site visits was, although there were portable air cleaners operated at some school classrooms, there was no maintenance or schedule for changing the filters, which may reduce the efficacy of air cleaners. Therefore, more studies are needed to gain knowledge towards establishing appropriate practices to maintain good indoor air quality in classrooms.

CRediT authorship contribution statement

Qian Zhang: Writing – original draft, Visualization, Methodology, Investigation, Data curation, Conceptualization. **Mark Wilson:** Writing – review & editing, Conceptualization. **Marilyn S. Black:** Writing – review & editing, Resources, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Acknowledgement

This work was funded by the Chemical Insights Research Institute of UL Research Institutes. We appreciate the educational and facility staffs of Georgia State University and Coretta Scott King School of the City of Atlanta for their assistance in these studies.

Appendix. Supplementary materials

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Appendix A. Supplementary data: Table A.1.; Figure A.1.; Figure A.2.

Recommended articles

Data availability

Data will be made available on request.

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