

Systematic ranking of filaments regarding their particulate emissions during fused filament fabrication 3D printing by means of a proposed standard test method

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Abstract

The diversity of fused filament fabrication (FFF) filaments continues to grow rapidly as the popularity of FFF-3D desktop printers for the use as home fabrication devices has been greatly increased in the past decade. Potential harmful emissions and associated health risks when operating indoors have induced many emission studies. However, the lack of standardization of measurements impeded an objectifiable comparison of research findings. Therefore, we designed a chamber-based standard method, i.e., the strand printing method (SPM), which provides a standardized printing procedure and quantifies systematically the particle emission released from individual FFF-3D filaments under controlled conditions. Forty-four marketable filament products were tested. The total number of emitted particles (TP) varied by approximately four orders of magnitude ($10^9 \leq TP \leq 10^{13}$), indicating that origin of polymers, manufacturer-specific additives, and undeclared impurities have a strong influence. Our results suggest that TP characterizes an individual filament product and particle emissions cannot be categorized by the polymer type (e.g., PLA or ABS) alone. The user's choice of a filament product is therefore decisive for the exposure to released particles during operation. Thus, choosing a filament product awarded for low emissions seems to be an easily achievable preemptive measure to prevent health hazards.

KEYWORDS

3D printer, emission test chamber, fused filament fabrication, indoor emission, standard test method, ultrafine particle

1 | INTRODUCTION

Fused filament fabrication (FFF) is a 3D printing technology, which was already utilized for rapid prototyping in the industry in the early 1990s.¹ Particularly in the 2010s, this technology has progressively matured and has simplified handling for non-professionals in the form of desktop models. In conjunction with the declining

acquisition prices, desktop FFF-3D printers have obtained a great popularity among users in educational institutions or private households.^{1,2} Following the trend, the diversity of printing materials (filaments) continues to grow rapidly. In addition, the emergence of online 3D printing platforms enables the exchange of knowledge and the share of digital design files, which encourages the application as home fabrication.

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FFF is based on material extrusion where thermoplastic filaments are melted, extruded through a heated nozzle, and then deposited layer by layer onto the printing bed to progressively build a 3D object. In the molten filament, evaporation and thermal decomposition of the thermoplastic occurs. Thus, volatile organic compounds (VOCs) and particulate matter, mainly as ultrafine particles (UFP, particle diameter $d_p \leq 100$ nm), are formed during the printing process, as described in many studies.^{3–34} These air pollutants are often released uncontrolled and unfiltered into indoor spaces where desktop FFF-3D printers normally are operated. Especially low-cost models are often neither equipped with an enclosure nor filter systems. Users are therefore directly exposed, which poses a potential health hazard. The focus of this study is the systematic quantification of particle emissions, especially ultrafine particles (UFP), released from FFF-3D printing. It has been shown that UFP such as ambient air pollution reach alveoli and have the potential to enter the bloodstream and reach other organs.^{35,36} Furthermore, carbonaceous UFP have been detected in the central nervous system of rats that possibly results from bypassing the blood–brain barrier.³⁷ Concerns have been increasing regarding potential health risks associated with prolonged exposition in indoor environment during a printing process.

Summarizing from previous emission studies,^{3,4,6–14,16,17,19,22,24,25,29,30,32,34} the particle emission from FFF-3D printing mainly varies as a function of: (1) the filament type, i.e., the basic polymer and fillers/dyes/additives, (2) the printer model, which comprises hardware and software, and (3) the individual printer settings, depending on the options of the printer model and the filament's specific instructions for use. The choice of a filament has been pointed out to influence the particle emission by up to several orders of magnitude.^{3,4,6–14,16,17,22,24,25,29,30,32,34} Further investigations suggested that color,¹⁰ metal^{14,16,25,34} and wood^{14,16,25} additives affect the particle emission level. Seeger et al.²² assumed that the particle emission is specific for an individual filament product rather than for its basic polymer and a ranking of emissions by the filament's basic polymer types seems not expedient. In the following, we therefore use the terms “filament product” as synonym for an individual compound comprising a basic polymer with stabilizers and usually unknown impurities, a selection of additives and dyes. The term therefore describes individual feedstock for purchase from a known marketplace or supplier rather than a class of material. Ideally, a filament product comes with a declaration of ingredients, instructions for use and a safety leaflet. In practice, this is not a standard.

Azimi et al.⁶ and Zhang et al.¹⁷ found printer hardware and firmware to be also influential for the emissions. A plausible explanation is that the firmware pre-defines the “object building strategy”, which comprises for instance the control and movements of extruder and printing bed, the details of the time-dependent deposition of material and the design of support structures such as rafts. Additionally, the heating power and rates as well as the accuracy of the temperature control should be considered as hardware-related emission factors. Printer equipment, e.g., fans, dust protection and built-in filters, may also have an influence on the exposure to released

Practical implications

- Nowadays, FFF-3D printers are used more and more frequently in private households.
- It is known that harmful emissions such as ultrafine particles are released indoors during operation.
- A proposed standard test method for rapid screening and identification of low particle emitting filament products is presented in this study.
- Selecting a filament product awarded for low emissions seems to be an easily achievable preemptive measure to prevent health hazards.

particles.^{10,12} Relevant customizable parameters are inter alia the operating temperatures of extruder^{6,7,12–14,17,22,24,25,30} and printing bed,^{6,22} the printing speed,^{7,19} and the object infill density and structure.¹⁹ Overall, it is evident that a multitude of influencing factors could greatly change the emission of particles and gases during a 3D printing process. In addition, the lack of standardization in terms of experimental conditions, measurement sites, instruments, metrics, and printing procedure details complicate an objectifiable comparison of research findings.

Many studies have emphasized the necessity of a standard test method.^{15,17,24,34} Zhang et al.¹⁷ and Stefaniak et al.¹⁵ made a first approach based on the application of the German Blue Angel ecolabel test guideline RAL-UZ 171 (latest revised version: DE-UZ-219:2021, Appendix S-M “Testing method for the determination of emissions from hardcopy devices”),³⁸ which was initially designed by the Federal Institute for Materials Research and Testing (BAM) for test chamber-based measurements of laser printer emissions.³⁹ BAM applied this test protocol in an early attempt to measure emissions from FFF-3D printers as well.²² An adaption to the special features of FFF-3D printing is required, not least because FFF-procedures involved materials and hardware settings are more versatile compared to those of laser printers. Several studies^{6,16,23,24} followed the test design of the National Institute of Standards and Technology (NIST)⁴⁰ and observed a variation of the print job duration between 2.5 and 4 h depending on the variety of printer hardware and filament-specific printer settings.⁶ Similar findings were described by Kim et al.⁴ while printing an identical object using different printer models and filament types. A standard method ANSI/CAN/UL 2904 for testing and assessing particle and chemical emissions from 3D printers was published in 2019.⁴¹ The experimental setup and the metrics for quantifying particle emission of that standard are based on the abovementioned Blue Angel test guideline. ANSI/CAN/UL 2904 proposes to print a cube with a fixed printing time of 4 h regardless of the printer model and the filament type. Albeit ANSI/CAN/UL 2904 tries to establish a comparability by normalizing the quantified particle emission to either mass of filament used or printing time, both will not adequately compensate the

bias resulting from varying filament properties. Firstly, the filament density varies to a large extent, for instance, metal additives could increase the weight of a given object by a factor of 2 or 3 and hence would cause an undervaluation of the related particle emission. Secondly, a normalization based on a fixed printing time would favor filaments with lower required printing speed. Last but not least it is questionable, if a long test duration is required to characterize material-related differences in the emissions as the highest particle emission rates are often obtained in the first minutes of extrusion (<15–20 min).^{4,6,11,12,17,30}

Summarizing, for FFF-3D printing, a standardized laboratory-based test procedure is still missing, which minimizes printer-specific biases and allows the measurement of filament product-related emissions for a better comparability of laboratory-based and in situ studies. Currently, the results from many in situ studies cannot be compared easily as the reported experimental conditions vary to a large extent. Therefore, we have worked on a standard printing procedure for a systematic filament product ranking with respect to particle emissions, assuming that even emissions from filament products with same color, additives and basic polymer may be grossly different. The procedure enables identifying and assigning low-emitting filament products, which could definitively raise the consumer's awareness and encourage them to use distinguished feedstock with proven low environmental impact. This represents an important step towards preventive consumer protection especially when health, safety, and environment requirements (HS&E) are not mandatory or available (see Table S1, for list of abbreviations and acronyms).

2 | MATERIALS AND METHODS

2.1 | FFF-3D printer and filaments

From the BAM's stock of printers, the Craftbot 2 (CRAFTUNIQUE Kft., Hungary) with proven lowest susceptibility to malfunctions was selected as reference for our measurements. The Craftbot 2 has a non-airtight enclosure and a cover hood with a built-in HEPA filter. Cover and HEPA filter were removed during the measurements to prevent a potential particle emission reduction effect. The printer utilizes 1.75-mm-diameter feedstocks and extrudes through a 0.4-mm brass nozzle. Extruder and printing bed operate within the temperature ranges of 180–260°C and 50–110°C, respectively, and enable the use of many popular filaments with a variety of basic polymers, colors, and additives for home and professional fabrication. Furthermore, the printer software, Craftprint (version 1.14), allowed the remote start of print jobs from outside the test chamber. This feature is expedient because opening the door after reaching a low background particle concentration in the chamber could be avoided. With this printer, 44 filament products, which are described in the supplementary data (Table S2), were systematically screened using the developed standard test method.

2.2 | Standard test method

2.2.1 | Measurement setup

Emission test chamber

The measurements were conducted in a 1-m³ emission test chamber, which complies with the standard ISO 16000-9:2006 and the basic criteria of DE-UZ-219. The emission chamber maintains standard climate conditions ($T = 23^{\circ}\text{C} \pm 2^{\circ}\text{C}$, $\text{RH} = 50\% \pm 5\%$) and is supplied with clean air at an exchange rate of 1 h^{-1} . All measuring instruments were operated outside the chamber and connected via antistatic ducts. The chamber is run with a slight overpressure to prevent contamination from outside.

Particle measurement

Particle emissions were continuously measured by means of the following instruments: A condensation particle counter (CPC, model 3775, TSI Inc., USA) measures the total particle number concentration (TPNC). The CPC's calibrated minimum detectable particle size, specified as diameter of sucrose particles with 50% detection efficiency (D50), is 4 nm while the detectable particle size maximum is $>3 \mu\text{m}$. It is worth mentioning that particles from 2.5 nm on are also measurable but with efficiencies considerably below 50%. The lower limit of detection (LLOD) of the TPNC is approx. 0.1 cm^{-3} ; hence, the CPC was also used to verify a negligible background TPNC of less than 100 cm^{-3} in the chamber. Particle number size distributions (PNSD) were determined by an aerosol spectrometer (EEPS™, model 3090, TSI Inc., USA) in the size range of 5.6–560 nm and additionally by an optical aerosol spectrometer (OPSS, model 1.108, GRIMM Aerosol Technik Pouch GmbH, Germany) in the size range of 0.3–20 μm . EEPS™ and CPC record the TPNC at 1 Hz.

2.2.2 | Data evaluation

Following the definition in DE-UZ-219, in this study, the total number of emitted particles (TP) during a print job is the evaluation parameter for particle emission. The calculation of TP considers particle losses in the chamber due to air exchange, wall adhesion, and other mechanisms. Under the given conditions, agglomeration and coagulation have been neglected, and this simplification can be justified by the observed PNSD time series but also from theoretical assumptions.⁴² The PNSDs revealed no indication for coagulation processes during a short print job. The TPNC rises rapidly with the start of extrusion and decreases exponentially after printing. The decrease of the TPNC curve represents in a simplified form the total particle loss constant, β , in the absence of an active emission source as defined by Equation (1):

$$\beta = \frac{\ln(c_1/c_2)}{t_2 - t_1} \quad (1)$$

C_1 and C_2 are the corresponding TPNCs at times t_1 and t_2 , respectively, after printing. The calculation of TP is described in Equation (2):

$$TP = V_C \left(\frac{\Delta C_p}{t_{stop} - t_{start}} + \beta \cdot C_{AV} \right) (t_{stop} - t_{start}) \quad (2)$$

with the chamber volume V_C and $t_{stop} - t_{start}$ assigning the duration of emission that starts from heating up and ends with stop of printer activities. ΔC_p is the difference in particle number concentration and C_{AV} is the arithmetic mean, both values are calculated between t_{stop} and t_{start} . For the calculations, CPC datasets were used due to the higher detection sensitivity.

The geometric mean diameter (GMD) and the corresponding geometric standard deviation (GSD) were determined from the EEPSTM data for describing the particle number size distributions.⁴³ The contribution to the TPNC in the size range of the OPSS was in all measurements negligibly low and hence was not considered for the TP calculations and the evaluation of PNSDs.

Other relevant descriptive measures are the arithmetic mean and the relative standard deviation (RSD) for a sample size (N) of at least three to describe the dispersion of data. The population RSD was calculated for comparison of the arithmetic mean values from very different sample sizes (N between 3 and 10). Grubbs outlier tests at a significance level of 0.05 were performed for $N > 6$. Detected outliers were excluded from evaluation. Linear regression was used for correlation analysis.

2.2.3 | Printer preparation

Prior to filament emission testing, the printer was subjected to a comprehensive preparation procedure to minimize residues from prior filament test runs. Especially when changing from a low-temperature to a high-temperature filament, the emission could significantly increase by a carry-over effect from the high heat stress and thermal decomposition of polymer residues.

Therefore, filament residues were manually removed from the hotend, and a new nozzle was used before each filament product exchange. Several meters of the filament under test were extruded by printing small cubes. This further reduced remaining residues, and at the same time, we used this procedure to verify for an individual filament product the suitable temperature setting for extruder and printing bed. All extruder temperatures were generally set approximately in the middle of the manufacturer's recommend ranges and adjusted only within this range if a cube printing failed. A successful print job was characterized by the non-appearance of warping or object detachment, and an apparently good object finish. Finally, residues on the printing bed were removed with a spatula or, if necessary, wiped off with alcohol before placing the printer into the chamber. It should be noted that the plentitude of available adhesives may be another source of particle or gaseous emissions, overlapping with those from the filaments. A systematic investigation was beyond the

scope of this study, and therefore, adhesives were not applied during all experiments.

2.2.4 | Strand printing method (SPM)

Our goal was to develop a robust standard printing procedure, which allows a non-biased comparison of filament product-specific emissions. Using a real 3D test object—even a simple cube—seemed not ideal for this purpose for the following reasons: (a) The printer's firmware may apply specific building strategies, i.e., it may vary slicing, extrusion speed, and lateral material dispense.⁶ These factors cannot be controlled by non-experts but could have an impact on emissions as well as the repeatability of the printed object's mass. (b) The cost and time effectiveness: Printing of large objects requires several hours of operation. (c) The production error rate: It may increase significantly if—as intended here—adhesives are not applied. Therefore, we designed a printing procedure, which solely extrudes a fixed length (i.e., a fixed volume at a given filament diameter) of a filament as a strand without building a 3D object. For each filament product under test, the filament length (or volume) was the same. A user normally intends to print an object of given dimensions and design, which uses a certain volume or length of feedstock. If he wishes to estimate the particle emission levels to be expected from a collection of filaments with highly different densities, he may use the normalized metric TP per extruded filament length or volume. Since a standard filament length is chosen for all tested filaments, we simplified the comparative parameter to TP. Designing the printing procedure with strands of constant mass, on the other hand, would entail different lengths, i.e., the smaller the density the longer the strand. Consequently, the extruding times would differ considerably and may impair the comparability. While the particle emission rate is used for comparison in several studies, it seems not well suited for a systematic comparison because it is a time-varying metric. The normalized TP, on the other hand, as an integral parameter seems to better compensate for small temporal variations and therefore was considered as an appropriate metric, which makes particle emissions for a given filament length and period of time comparable.

This minimized and standardized printing procedure is called "strand printing method" or SPM in this study. Figure 1 illustrates how the strand is extruded and laid down on the bed.

The SPM makes use of a special G-code, i.e., the printer's control code, which was initially developed for the Craftbot 2 with the software Repetier-Host (Version 2.0.5, Hot-World GmbH & Co. KG, Germany) and was exclusively tested for this model. A reference G-code is given in Figure S1. Some relevant printer settings must be specified: the temperature settings of extruder and printing bed, the extrusion rate, i.e., the extruded filament volume or length per time, and the total extrusion duration.

The temperatures were set for each filament product under test based on manufacturer's recommendations, and the settings were

examined in pretests (cube printing) as mentioned before. In general, the printing bed was also heated up to properly mimic the condition of a real 3D object printing and to include potential emissions.

A constant extrusion rate of approximately 68 mm/min (millimeters of filament per minute) was chosen, which represents an average printing speed for FFF-3D printing. The highest particle emission rate is usually observed within the first minutes (<15–20 min) after the extrusion starts.^{4,6,11,12,17,30} Therefore, we decided to extrude filaments with a fixed length of 800 mm to reach a total extrusion duration of about 12 min. This turned out to be sufficient for recording rise and characteristic maximum or plateau of the particle number concentration. Moreover, the short printing procedure reduces the required total testing time and allows up to two repetitions per day for an improved statistical quality.

It is apparent that the SPM omits an essential component of FFF-3D printing, namely the layer-by-layer construction. On the other hand, it determines the actual emission resulting mainly from the heat stress of the filament material (during heating up, melting, extruding, and cooling) while minimizing the abovementioned factors that are hard to control experimentally.

As reported in several emission studies,^{13,15,21,22} a loss in bed adhesion forces an abort of the measurement. Especially materials prone to warping tend to fail quite often even if adhesives are

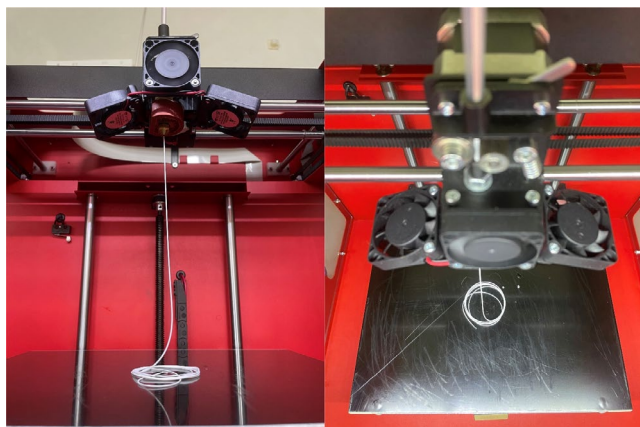


FIGURE 1 Strand printing method (SPM)

TABLE 1 Details of printing procedures

Parameter	15.8-mm cube	SPM
Extruder temperature	Filament-specific	Filament-specific
Printing bed temperature	Filament-specific	Filament-specific
Printing speed	Filament-specific	Constant
Filament consumption	Approx. ^a 805 mm	800 mm
Extrusion duration	Approx. ^b 10 min 15 s	Approx. ^b 11 min 47 s
Layer height setting	0.2 mm	–
Infill density/structure	15%/parallel lines	–
Raft	Yes	–

^aBased on Repetier-Host software calculation.

^bBased on own measurement.

applied. In emission tests without adhesives, this issue would not only increase the error rate but would also limit the choice of filament test candidates. SPM avoids this as adhesives are not used and hence play no role as potential emission sources. Therefore, the benefits of the SPM seem to outweigh the disadvantages. Material-driven emissions from filaments can thus be clearly quantified and ranked based on their emission level with a reasonable cost- and time-effective test effort.

2.2.5 | Cube printing

A direct comparison of particle emissions from SPM and object printing is discussed in Section 3.3. A cube model was printed with a raft, i.e., a foundation underneath the 3D object for a better bed adhesion. The filament consumption of the whole object printing was adjusted to that of SPM. The printer's slicing software Craftware (Version 1.21.1) was utilized for generating the G-Code for cube printing. Table 1 summarizes all details of both printing procedures in this study.

3 | RESULTS AND DISCUSSIONS

3.1 | Filament screening

3.1.1 | Reproducibility of the extruded filament mass

Within the SPM, the appropriate measurand for a check of reproducibility of the material throughput is the mass ratio of extruded strand to filament input. The input is via the G-code set for feeding 800 mm of filament at a fixed rate, and it was validated how accurate the filament feeder could reproduce this. For each filament product, 800 mm of unprocessed filament was manually cut and weighed. The masses were compared with the respective masses of the extruded strands. These could easily be taken out of the printer without any loss of material. All masses were determined with a semi-micro balance (Model MSE225P, Sartorius

Lab Instruments GmbH & Co. KG, Germany) in an air-conditioned laboratory.

Figure 2 shows a linear correlation through the origin ($R^2 = 0.998$) between the strand masses and the masses of the cut filaments. The scatter of datapoints is very low. From repeat printing, the relative standard deviation of the strand masses was found to be typically less than 1%. Uncertainties from cutting and weighing the filaments seem negligible for the purpose of comparison. The diagram indicates that for a variety of filaments with different densities and extruder temperatures, the G-Code provides filament feeding with very high reproducibility. The slope of 0.965 points at a small mass loss after extrusion which, after correction for the densities, turns out to be constant in volume for all filaments. A plausible explanation would be evaporation of volatile organic compounds (VOCs) during thermal processing of the filament.⁴⁴

The SPM G-code and the filament feeder of the Craftbot 2 provide reproducible filament extrusion for a wide range of investigated filament products, except for one tested TPE (thermoplastic elastomer) filament, whose strand mass was considerably lower than the filament mass. Probably, the softness and elasticity of TPE compromises the feeding by the extruder. It may require a rather slower extrusion rate than the applied one for screening.

3.1.2 | Emission from printer hardware

Due to the relatively high operating temperatures, it can be expected that heated or heat-exposed printer parts (extruder, hotend, printing bed, mechanical parts with lubricant such as gears and slide rails) have the potential to emit particles. To investigate this, the printer was thoroughly cleaned as described in Section 2.2.3 to avoid influences from filament residues. The extruder temperature was set between 190°C and 260°C, and the printing bed was constantly kept at 110°C while executing all SPM steps but without actually feeding a filament. From the empty extruder, a quantifiable onset of particle emission was observed at temperatures of 250°C ($TP = 3 \cdot 10^9$) or higher (260°C: $TP = 2 \cdot 10^{10}$). There is still quantifiable hardware emission observable for extruder temperatures $\geq 250^\circ\text{C}$, which has to be considered for the interpretation of TP values for filaments with extruder temperatures in this range. It can be assumed that a printer's hardware contribution to emissions may vary depending on the model.

3.1.3 | Quantification of TP

A total number of 44 filament products were screened for their particle emissions by means of the above-described SPM. Filament products can roughly be divided into low emitters and high emitters separated by the TP median (approx. 10^{11}) of all filaments investigated. The TP values in Figure 3 were calculated from CPC data. In DE-UZ-219, the lower limit of quantification (LLOQ) for TP of

approximately 10^9 is connected to a minimum required increase in particle number concentration of 1000 cm^{-3} . Below, a proper calculation of TP is not possible, and in these cases, the measurement will be considered as valid but "not quantifiable." All experiments with quantifiable TPs were repeated at least twice, and the relative standard deviations (RSDs) were determined. Overall, only one outlier was detected for the filament HP-01 according to Grubbs tests at a significance level of 0.05 (see Figure S2). A detailed data set is provided in Table S2. Figure 3 reveals that among the 44 filament products, TP varies by approximately four orders of magnitude ($10^9 \leq TP \leq 10^{13}$). The range would have been even greater if filaments with TP below the LLOQ ($TP \approx 10^9$) were quantifiable. Grouping filament products by their basic polymers reveals that within each of the groups, a notable variation in TP is evident and, in some cases, even larger than between the groups. It seems therefore not appropriate to classify or estimate a filament product's particle emission level solely by its basic polymer. Instead, a proper ranking of individual filament products seems necessary and is feasible by the SPM approach.

PLA or PLA-based filaments

PLA is by far the most common utilized basic polymer in FFF due to its high variety of products filled for example with wood, stone, or metal additives. PLA filaments with and without fillings could in this study mostly be classified as low emitters. Values for TP were either not quantifiable or below $<10^{10}$. However, two copper-filled PLA filaments (MF-CO-01 and MF-CO-02) showed substantial higher emissions ($TP_{\text{Mean}} > 10^{11}$) and hence are rather high emitters, and the impact of (copper) additives is apparent. TP_{Mean} therefore varied within the PLA filament group by more than two orders of magnitude ($TP_{\text{Mean}}: 10^9 - 10^{11}$).

PETG or PETG-based filaments

PETG or PETG-based filaments such as HDglass (HD-K-01) or carbon fiber-filled PETG (CA-01) are low emitters. Overall, TP_{Mean} stayed at the level of 10^{10} or below for this polymer type, even though high extruding temperatures were applied for PETG-T-02 ($T_E = 255^\circ\text{C}$) and CA-01 ($T_E = 260^\circ\text{C}$), which implies a substantial contribution from printer hardware (see Section 3.1.2).

PVA filament

One PVA filament was screened, and TP was below the LLOQ.

PP filament

$TP_{\text{Mean}} \approx 10^9$ identifies the PP filament as low emitter.

TPE filaments

Two tested TPE filaments showed low TP_{Mean} of approx. 10^9 , hence both were classified as low-emitting filaments. However, the filament extrusion was not very reproducible for TPE-W-01 as mentioned in Section 3.1.1. Nevertheless, based on the TP level, it can be assumed that TPE-W-01 is a low emitter.

ABS filaments

Five ABS filament products were tested. The range in TP was three orders of magnitude between the lowest (ABS-T-01, $TP_{\text{Mean}} = 4 \cdot 10^9$) and the highest emitter (ABS-G-02, $TP_{\text{Mean}} = 4 \cdot 10^{12}$). The comparison of two ABS filaments, ABS-T-01 (transparent) and ABS-W-01 (white), from the same manufacturer hints to the influence of color additives, in this case presumably Ti pigments. Both filaments were operated with identical settings, and the difference in TP was about two orders of magnitude ($TP_{\text{Mean}}: 10^9$ – 10^{11}). Moreover, two black ABS filaments (ABS-S-04, rTi-01) originating from different manufacturers but extruded at the same temperature differed by one order of magnitude ($TP_{\text{Mean}}: 10^{10}$ – 10^{11}). These examples are underpinning our general finding that filament products—even those with

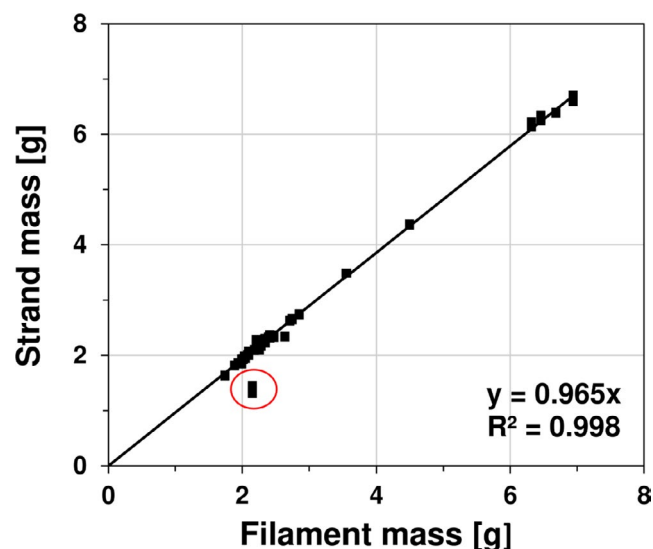
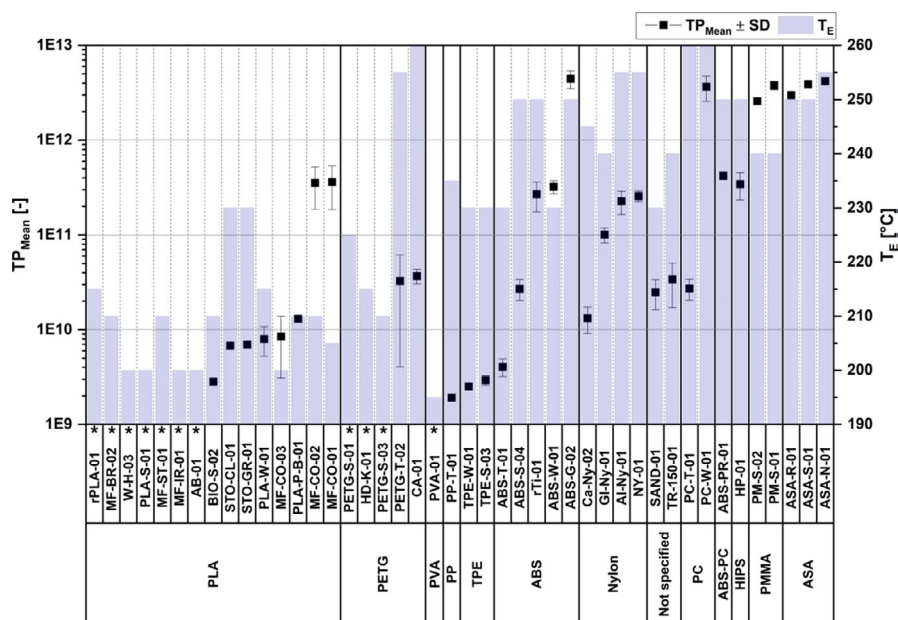


FIGURE 2 Linear correlation between filament and strand mass. Red circle marks the filament TPE-W-01

FIGURE 3 Particle emissions (TP_{Mean}) from 44 investigated FFF-3D filament products and corresponding extruder temperatures (T_E). Black squares: arithmetic mean, TP_{Mean} , of multiple measurements. Error bars: standard deviation of the mean values (SD). Filaments with asterisk: TP below the lower limit of quantification (LLOQ). Filament products are grouped by their respective basic polymers. CPC data were used



the same basic polymer—may have distinct and individual particle emission levels.

Nylon filaments

Four different nylon filaments showed unequal particle emissions ($TP_{\text{Mean}}: 10^{10}$ – 10^{11}), which possibly resulted from their heterogeneous composition by additives (e.g., carbon and glass fiber) or the applied extruder temperatures.

PC filaments

The TPs of a white and a transparent PC filament from two different manufacturers varied by approximately two orders of magnitude ($TP_{\text{Mean}}: 10^{10}$ – 10^{12}). The particle emission of the transparent product was significantly lower than that of the white filament. As it was also observed for ABS, the effect of color additives could be the same. Considering the fact that PC-T-01 was extruded at 260°C, the emission is expected to originate from the printer hardware, and the filament itself is assumed to only have a negligible contribution.

ABS-PC filament

The investigated filament product consists of ABS and PC and has a flame retardant property. With a TP_{Mean} greater than 10^{11} , this filament belongs to the high emitters.

HIPS filament. One filament product was tested with a TP_{Mean} above 10^{11} ; hence it is a high emitter.

PMMA filaments. Two black PMMA filament products showed the highest emission level of $TP_{\text{Mean}} > 10^{12}$ among the tested products and are therefore classified as high emitters.

ASA filaments. Three differently colored (red, black, white) ASA filaments from the same manufacturer were all at almost the same

emission level and comparable with PMMA and hence are high emitters as well. Seemingly, the colors had no influence on TP.

Filaments with unspecified basic polymer. SAND-01 and TR-150-01 showed a moderate TP level of 10^{10} .

The normalized TP (number of emitted particles per extruded filament length or volume) may be used as a comparative metric in future emission studies. For comparison with previous studies, the particle emission rate or the particle emission per extruded mass can theoretically be converted to normalized TP values via time integration or by multiplying the filament density, respectively, if the filament consumption is known. It should be noted that assumptions and calculations for emission rates or emission per extruded mass may differ from study to study, and hence, a direct comparison is not applicable.

Particle emissions are overall expected to be temperature-driven, but several exceptions are observable: for example, the PETG filaments (PETG-T-02, CA-01) emitted a relatively low number of particles at high extruder temperatures which mainly result from the printer hardware as described above. On the other hand, two copper-filled PLA filaments (MF-CO-01, MF-CO-02) show a contrary trend; TPs are clearly above 10^{11} while the extruder is at rather low temperatures ($\leq 210^\circ\text{C}$). Moreover, TPs for the two PC filament products differ by approximately two orders of magnitude while the extruder temperature is the same. In Figure 4, no proportionality between extruder temperature (T_E) and TP was found. It can be observed that the upper emission level ($TP_{\text{Mean}} > 10^{12}$) was only achieved at relatively high $T_E \geq 240^\circ\text{C}$. At the same extrusion temperatures, however, a much lower emission level of $TP_{\text{Mean}} = 10^{10}$ could also occur. Based on the extruder temperature alone, the particle emission from a filament product cannot be estimated.

3.1.4 | Particle number size distribution

The temporal course of the number-based particle size distribution (PNSD) during SPM is shown in Figure 5. In general, the particle number concentration increases rapidly with the start of extrusion and declines after stop. Two emission patterns were typically observed:

1. The particle number concentration quickly reaches a peak as the extrusion starts and remains constant or is slightly decreasing afterwards, e.g., ASA-N-01. With the stop of extrusion, the concentration declines exponentially.
2. The particle number concentration rises continuously from start to the end of the extrusion, e.g., HP-01. After the extrusion is finished, the concentration level drops.

It is noticeable that no significant shift in PNSD maxima can be observed within the short period of extrusion of around 12 min indicating that there is practically no coagulation/agglomeration. The geometric mean diameter (GMD) and the corresponding geometric standard deviation (GSD) were determined at the end of the printing process as the spectrum at that time characterizes the expected size distribution most closely if the printing process is continued.

The typical PNSD for SPM are in the UFP range with GMDs between 20 and 90 nm and are rather polydisperse (with GSDs between 1.37 and 2.01). Hence, they are comparable to those from 3D object printing as reported in the literature.^{4,9-11,16,17,21,22,24,34} Some filaments revealed a relatively high variation of GMDs in repeat measurements (see Table S2). This can be seen as rather filament-specific as there is no obvious correlation with printing parameters, e.g., extruder temperature or TP levels.

It should be noted that particulate emission from FFF-3D printing may contain a substantial amount of sub-4-nm particles,^{13,25} which could not be detected with our instrumentation.

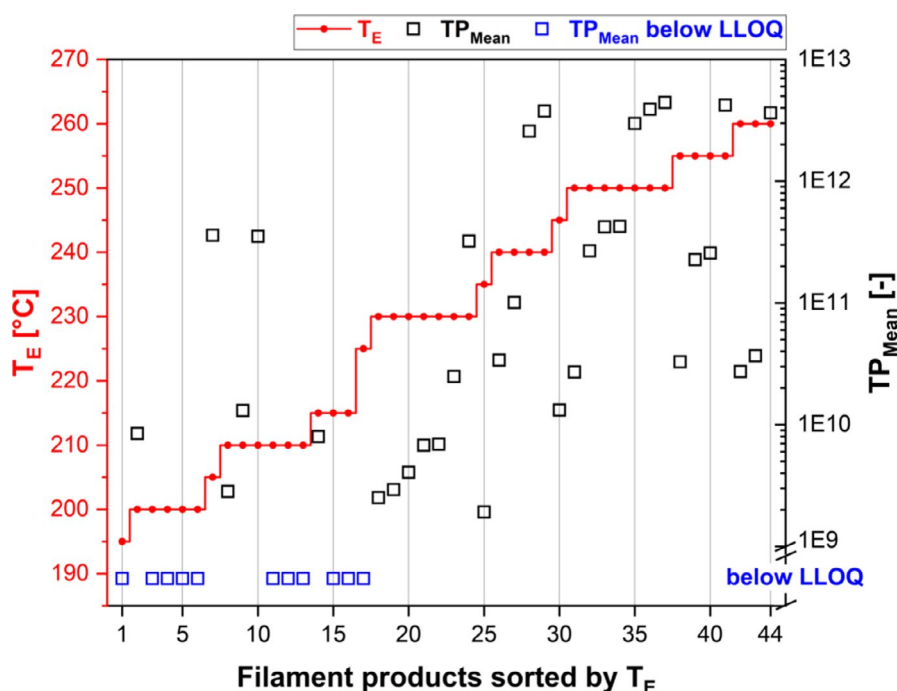


FIGURE 4 Correlation between extruder temperature T_E (red line) and TP_{Mean} (black squares). TP_{Mean} below the lower limit of quantification (LLOQ) is marked in blue. Filament products are sorted by T_E . TP_{Mean} are shown on a logarithmic scale

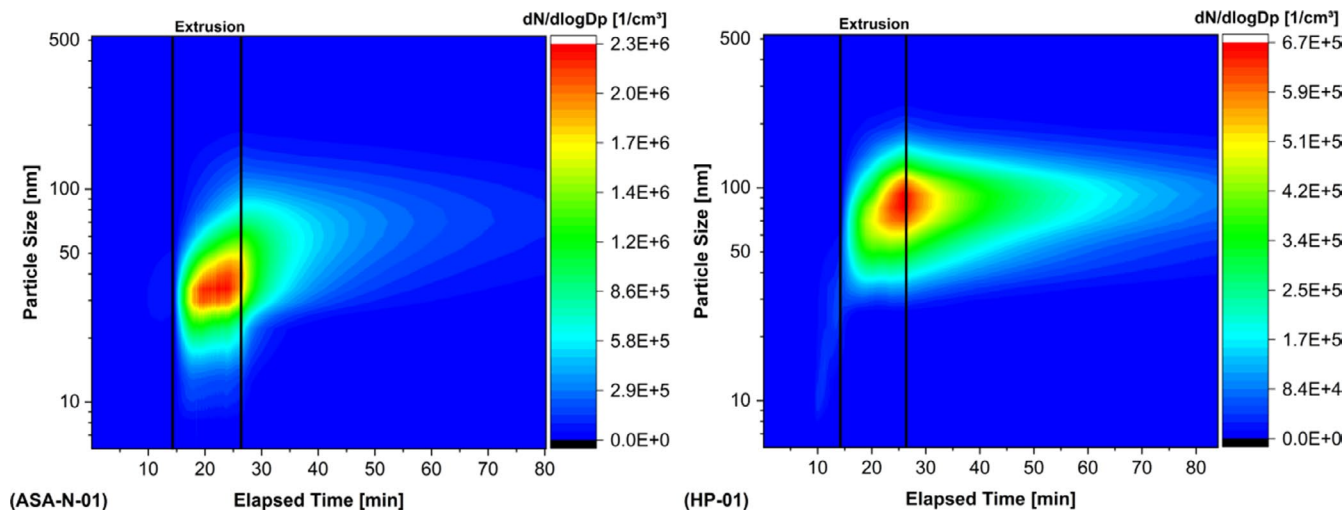


FIGURE 5 Time-dependent PNSD for ASA-N-01 and HP-01. The period of extrusion is marked. The data were obtained from the EEPSTM

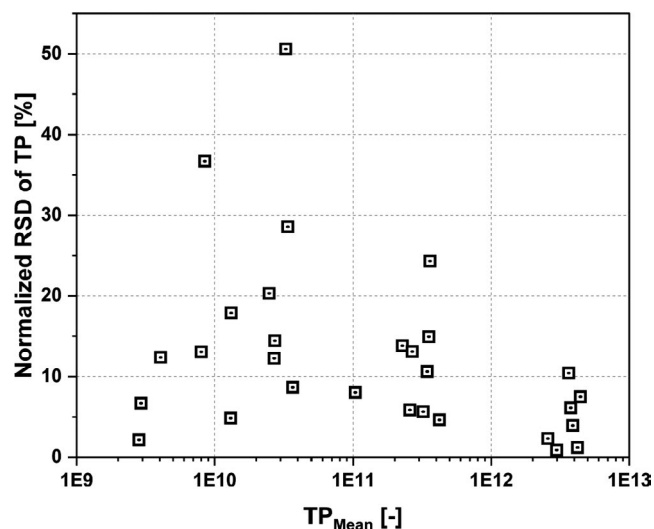


FIGURE 6 Reproducibility of TP. Black squares show the normalized relative standard deviation of TP over different TP levels

Our working hypothesis assumed sub-4-nm particles to be rather the small tails of monomodal size distributions with maxima clearly above several 10 nm. Poikkimäki et al.²⁵ stated that the sub-3-nm fraction could account for up to 48% and may occur as a separate mode in the PNSD. Therefore, our TP values could probably underestimate the true total number of emitted particles but even when a factor of two is considered, the ranking among filaments with TP varying by up to four orders of magnitude is not significantly affected. Nevertheless, the quantification of the sub-4-nm particle size fraction will be included in our ongoing FFF-3D printer emission studies, not least in order to decide if particle enhancers or magnifiers such as used by Poikkimäki et al. are indispensable for a standardized testing setup.

3.1.5 | Reproducibility of TP

The reproducibility of TP within repeat measurements was quantified by the relative standard deviation (RSD) considering numbers of repetition between 2 and 9 (see Table S2). RSDs were normalized to better reflect the effect of different sample sizes. As shown in Figure 6, there is no proportionality to TP over several orders of magnitude. The majority of normalized RSDs remain below 20%, and hence, SPM can be considered as sufficiently selective at the same level as for laser printer particle emission testing.³⁹ Some filaments, e.g., MF-CO-01, MF-CO-03, PETG-T-02, and TR-150-01, show normalized RSDs clearly above 20% (Figure S3). This is considered as characteristic for a filament material rather than typical for the SPM testing method. The phenomenon of fluctuating levels of emissions was also observed for some laser printer models in comparability tests.³⁹

3.2 | Influence of the extruder temperature

As reported in previous studies, the applied extruder temperature could have a major influence on the level of particle emission, especially when exceeding the manufacturer-recommended range.^{12-14,30} The effect of an increase in extruder temperatures on filament emissions was measured by applying the respective recommended maximum extruder temperatures ($T_{E,Max}$) (Figure S4). The TP values at $T_{E,Max}$ were compared to those from the filament screening at T_E , listed in Table S2.

In our experiments, the extruder temperature increase ΔT_E from T_E to $T_{E,Max}$ varied between 5 and 25°C. Figure 7 shows that an increase of extruder temperatures elevated the TP levels consistently for all investigated filament products. While the effect is observable already at $\Delta T_E = 5^\circ\text{C}$, it reaches up to one order of magnitude or higher at $\Delta T_E = 10^\circ\text{C}$ (MF-CO-03, SAND-01, and ABS-T-01). $\Delta T_E = 25^\circ\text{C}$ caused an increase in TP for the filament PP-T-01 by

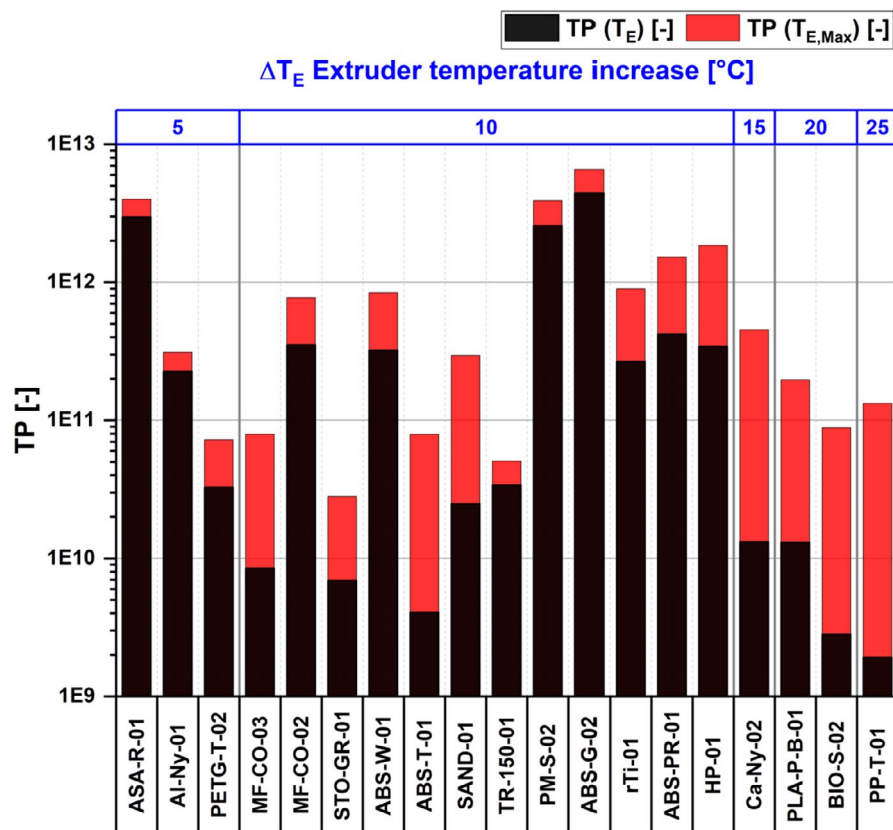


FIGURE 7 Effect of extruder temperature increase ΔT_E on TP. Black bars: TP at T_E ; red bars: TP at $T_{E,Max}$

nearly two orders of magnitude. It is therefore evident that generally any increase in extruder temperature—even within the recommended range—leads to a substantial increase in particle emission for a given filament product. This result strongly emphasizes the importance of a deliberated temperature setting to avoid unnecessarily elevated exposures.

At a given ΔT_E , the corresponding increase in TP is disproportionate even for filaments of the same basic polymer. This can be illustrated by the examples of ABS-W-01 and ABS-T-01, which both originated from the same manufacturer. While for $\Delta T_E = 10^\circ\text{C}$ the particle emission from ABS-W-01 increased moderately, an increase by approximately one order of magnitude was observed for ABS-T-01 under the same experimental conditions. The example emphasizes again that emissions from filaments are highly specific for a product, i.e., a compound from a basic polymer with additives and probably impurities, rather than for the basic polymer alone. This finding supports our intention to identify and assign low-emitting filament products.

It is worth mentioning that the PNSD generally remained unchanged when the extruder temperature was increased (see Table S3). Only 1 out of 14 investigated filament products revealed a significant change in PNSD.

3.3 | SPM compared to 3D object printing

The strand printing method intentionally omits the “object building process” for simplification and standardization purposes as described before. Generally, SPM can only anticipate the filament

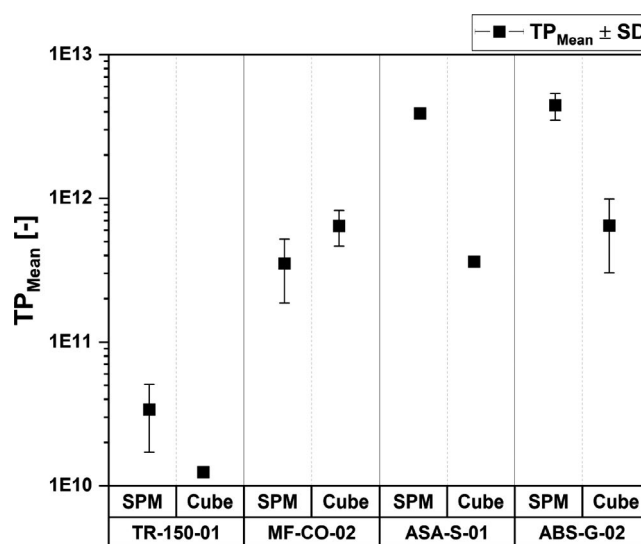


FIGURE 8 Comparison of TP_{Mean} from repeated strand printing and object printing (cube). Error bars represent standard deviation of the mean values (SD)

material-related effect on particle emission during real object printing. It must be said that in reality, this effect can be overshadowed by several abovementioned usercontrolled factors and printer hardware. A filament characterized by SPM as low-emitting may therefore show higher emissions when the settings for object printings are not optimal. Our working hypothesis postulates therefore that the use of a low-emitting filament is—besides optimizing the printer

setting—a measure to reduce the emission from object printing. To validate this, TP values from strand printing were compared to those from cube printing with identical filament consumption (see Table 1). The comparison of four filaments shows that the SPM-based ranking is not exactly reproduced by the cube printing results but confirmed as a trend over almost two orders of magnitude in TP (see Figure 8). The ASA-S-01 and ABS-G-02 filaments show larger discrepancies between SPM and cube printing. We like to point out that these results are based on only few measurements and hence should be considered as preliminary. Additional experiments will be conducted in an ongoing research project in order to support the concept of using SPM to classify filaments by their particle emissions.

4 | CONCLUSION AND OUTLOOK

In this study, a new approach for testing particle emissions during the processing of filament in an FFF-3D printer—the strand printing method (SPM)—was presented. SPM is emission chamber-based and enables the characterization of filament products by their individual emission strengths. The advantages of SPM are (1) conclusive comparative analysis of a wide range of filament products at fixed filament consumption, extrusion rate and duration, and under controlled conditions, (2) minimized printer software and hardware bias, (3) time and cost efficiency due to short duration testing with very low failure rate and good repeatability, (4) parallel testing of gaseous emissions based on Tenax[®] TA sampling and TD-GC/MS analysis was successfully included into the SPM scheme and results will be published in a future paper.

Our comparative study revealed that the particle emission varies within each group of most popular polymers (namely PLA and ABS) by at least two orders of magnitude. Based on the results from 44 filament products tested, a general categorization of the particle emission just from the filament's basic polymer seems not plausible. Even though the basic polymer has an impact, the origin of a polymer, its potential (unidentified) impurities, and the compounding of additives may influence the emission on a large scale. Last but not least, the increase of thermal stress on filaments during printing is an important emission factor. Further investigations should corroborate our preliminary finding that emission from strand printing seems to correlate with real printing scenarios. It also seems desirable to apply SPM to other FFF-3D printer hardware for a determination of printer-specific emission factors. Hence, SPM seems suited for awarding an ecolabel, such as the German ecolabel "Blue Angel", to low-emitting filament products. This has two intended effects: (a) stimulus to manufacturers to reduce the emissions of their products and (b) information of users (by the ecolabel tag) to make an informed choice. Reducing the health risk from air pollution due to emitted particles seems feasible if proven low-emitting filament products are available.

Such an application requires that filament products can be tested and ranked according to at least one property of concern (particle emission) so that a selective pass-fail criterion can be established.

We have demonstrated that such a ranking is possible and that a pass-fail criterion for particle emissions can be established based on the TP parameter. The presented overview on the particle emission levels from 44 marketable filaments may serve as a data basis to quantify a particle emission limit.

CONFLICT OF INTEREST

The authors declare no conflict of interests.

AUTHOR CONTRIBUTIONS

Conceptualization, data curation, formal analysis, investigation, methodology, project administration, software, supervision, validation, visualization, writing—original draft, and writing—review and editing: Chi-Long Tang. *Conceptualization, data curation, funding acquisition, methodology, project administration, resources, software, supervision, writing—original draft, and writing—review and editing:* Stefan Seeger.

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SUPPORTING INFORMATION

Additional supporting information may be found in the online version of the article at the publisher's website.

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