

# METHODOLOGIES FOR THE DETECTION AND CHEMICAL CHARACTERIZATION OF PARTICLES RELEASED FROM CIGARETTE FILTERS

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## SUMMARY

It has been claimed previously that cellulose acetate (CA) fibres and carbon granules can be released from cigarette filters during smoking, thereby presenting a health risk to the smoker. The purpose of this study was to conduct an assessment of cigarette filters made of CA and containing activated carbon granules to gain a greater insight into the likely risks involved. The release of particles from cigarette filters containing activated carbon ("Dalmatian" type filters) under simulated smoking conditions was compared to the results of a standard CA filter using two different methodologies.

Unlit cigarettes were "sham smoked" in a vertical position on a sampling apparatus according to ISO conditions and released particles were collected on a gold-coated capillary pore membrane filter. The loaded filter was then analysed directly using a Raman microprobe: the individual particles were detected and counted in randomly selected areas of the filter and classified to three different types of particles/fibres based on their length/width ratio. Full characterisation could be achieved by microscopic examination combined with a Raman spectroscopic system which allows the assignment of chemical compositions of organic or polymeric particles.

Using an alternative methodology, the released particles were detected online and classified in eight size ranges between 0.3 and 15.0  $\mu\text{m}$  by means of a laser aerosol spectrometer.

Although different experimental approaches for particle detection were used, comparable results were obtained. The particles released from cigarette filters were mainly identified as typical cigarette and filter constituents. Compared to the dust exposure for a corresponding volume of the usual ambient air, the number of particles released from cigarette filters corresponds to clean-air room particle concentrations. Due to the low number of particles released, high variability was observed between single measurements.

## INTRODUCTION

It has previously been claimed that fibres and carbon granules can be released from cigarette filters during smoking, thereby presenting a health risk to the smoker [1, 2].

There is currently no standard methodology for characterising the incidence of particle release from filters. As described in the literature [3-5], procedures were developed based on current best practices within the tobacco industry. These covered 'tap tests' in which the filter end is placed on an agar gel surface to simulate mouth contact and 'sham smoking' tests (defined as puffing unlit cigarettes). In the latter case, filters are subjected to a standard puffing cycle and any matter released into the air stream is collected. In all cases, particles were counted and characterised using image analysis systems.

In 2007, the DIN (German Institute for Standardization) ad-hoc group "Particles from Filter" published a report which summarises the results of investigations to determine the number and size/dimension of respirable particles released under simulated smoking conditions (more

intense sham smoking than ISO 4387) from three leading CA filter cigarette brands on the German market, and also to characterize the dust generated by cutting cigarette filters during the cigarette manufacturing process [6]. The DIN report concludes that the particle load, based on the smoking volume, is much lower compared to the dust exposure of the usual ambient air. From the toxicological perspective it is concluded that the release of particles from acetate filters does not constitute a particular health risk.

The purpose of this study was to conduct an assessment of cigarette filters containing cellulose acetate and carbon granules and to gain a greater insight into the likely risks involved.

## EXPERIMENTAL

### 1. ITG Methodology

#### 1.1 Principle

Unlit cigarettes are “sham smoked” in a vertical position on a sampling apparatus according to ISO conditions. A sample of particles released from the mouth end of the cigarette filters is collected on a commercially produced gold-coated capillary pore membrane filter (pore size 0.8  $\mu\text{m}$ ). The loaded filter is then analysed directly using a Raman microprobe without any alteration of the particulate deposit: the individual particles are measured and counted in randomly selected areas of the filter according to defined particle counting rules, and classified to three different types of particles/fibres based on their length/width ratio. The chemical characterisation of particles collected is performed by Raman spectroscopy.

#### 1.2 Sampling Apparatus (“Sham Smoking”)

A rotary sampling apparatus (“Particle Sampler Burghart”, *Heinrich Burghart GmbH, Wedel, Germany*) was designed which allows “sham smoking” according to different smoking regimes. For this experiment, cigarettes were sham smoked in a vertical position according to ISO conditions: each sham smoking run was performed with 20 cigarettes using a 35 mL puff volume, puff duration 2 seconds, puff frequency 60 seconds. Ten puffs were taken per cigarette. Ten sham smoking runs were performed with each membrane. Following this protocol, particles from 2000 puffs delivered by 200 cigarettes were collected and a total volume of 70 litres was drawn through each membrane.

During sampling, the cigarettes were covered by a Plexiglas hood equipped with four air filters (pore size 0.2  $\mu\text{m}$ ). Prior to sampling, the hood was flushed with filtered air for 5 minutes to generate a defined environment within the hood and to afford a constant “background noise” during sampling. A constant air flow was ensured during sampling. Every working day, the sampling system was checked for leak tightness and the puff volume was controlled.

#### 1.3 Cigarette Holder, Particle Collecting System

The cigarette holder and membrane housing were constructed as follows: In the cigarette holders, typical labyrinth seals were used covered with metal plates to assure vertical positioning of the cigarettes. No washer supports were used in the cigarette holders but the cigarettes were retained in the holder by metal rings equipped with small pins. This assembly allowed a tight seal and an even hold of the cigarette with minimal pressure on the filter and minimal coverage of its mouth end area.

The membrane housing was made of stainless steel and designed in a way that the membrane was inserted between two components and sandwiched by two “O”-ring seals. The housing was firmly closed by pushing both elements together and interconnecting them using a

bayonet connector. With this arrangement, the membrane was directly exposed to the mouth end of the cigarettes within a distance of 10 mm when mounted on the sampling apparatus. Any particles released from the cigarettes would fall directly onto the membrane without any obstruction.

Particles were collected on Raman-inactive gold-coated polycarbonate membrane filters with defined pore size (0.8  $\mu\text{m}$ ) enclosed by an aluminium ring. The membrane was mounted into a membrane holder with an effective diameter of 2.2 cm (diameter of “O”-ring seals). In order to avoid contamination, the membrane holder was rinsed with isopropanol and dried under clean room conditions before the membrane was inserted and the holder was assembled and sealed with a cap in a clean room bench. Immediately after sampling, the membrane holder was capped again for particle counting and characterisation.

#### 1.4 Particle Counting and Characterisation

Image analysis was performed using optical microscopy to enumerate and analyse the size and shape of the particles. All steps were performed automatically by the Single Particle Explorer® (*rap.ID Particle Systems GmbH*, Berlin, Germany) which randomly generates a specific number of microscopic images in 500  $\mu\text{m}$  x 500  $\mu\text{m}$  fields of the membrane surface. Enumeration and identification of particles was performed in six specific size ranges: particles from 2-5  $\mu\text{m}$ , 5-10  $\mu\text{m}$ , 10-25  $\mu\text{m}$ , 25-50  $\mu\text{m}$ , 50-100  $\mu\text{m}$  and > 100  $\mu\text{m}$ .

It has been demonstrated in this study that an equal distribution of particles was given which allows a statistical evaluation of the counted particles over the effective membrane diameter.

In combination with the image analysis, an automated Raman microprobe measurement was performed [7, 8]. Full characterisation could be achieved by microscopic examination combined with a Raman spectroscopic system which allows the assignment of chemical compositions of organic or polymeric particles [9, 10]. The resulting Raman spectra were compared with a database and the best match was automatically written into a report.

The Raman microprobe allows investigation of molecular vibrations associated with specific organic functional groups and yields structural information. It can provide limited information on non-metallic inorganics. A limitation of the Raman technique is the competitive fluorescence that always accompanies Raman spectroscopy. If strong fluorescence occurs, no information on chemical constitution can be collected [9, 10].

The numerical concentration of the particles was determined by the particle count result, the examined filter area, and the effective filter area.

The detected particles were classified in three categories [11, 12]:

- “Spherical particles” – particles without a specific orientation and elongated particles with a length/width ratio  $L : D < 3 : 1$ .
- “Respirable fibre-shaped particulates” (RFP) – elongated particles with a length > 5  $\mu\text{m}$ , a width < 3  $\mu\text{m}$  and a length/width ratio  $L : D > 3 : 1$ .
- “Non-respirable fibre-shape particulates” (non-RFP) – elongated particles with a length/width ratio  $L : D > 3 : 1$  which are not considered RFPs.

Particle number and size accuracy testing demonstrated a qualified performance of 15 %: The system for particle counting and characterisation is considered to be within specification when the counting accuracy determines a mean particle size with less than 15 % deviation from the true size of the spheres. Failure is usually attributed to generation of a poorly focused image [5, 13].

## **1.5 Blank Test**

For the blank test, the system was operated without any cigarettes inserted in the holders and in exactly the same way as for sampling from cigarettes. Thus, the blank test describes the number of particles detected in the filtered ambient air inside the Plexiglas hood.

## **2. Online Methodology (ITCF Denkendorf) [14]**

### **2.1 Principle**

Unlit cigarettes were “sham smoked” in a horizontal position using a Borgwaldt single-port smoking machine under clean room conditions (ISO 14644-1 clean room classification 5). Sham smoking was performed under more intense conditions than those of ISO 4387: puff volume 55 mL, puff duration 2 seconds, puff interval 15 seconds. For each sample, 20 cigarettes were sham smoked separately with 10 puffs per cigarette. The released particles were detected online and classified in eight size ranges between 0.3 and 15.0 µm using a laser aerosol spectrometer.

### **2.2 Clean Room, Background Contamination**

Sampling and particle counting was performed in a clean room at ITCF Denkendorf, Germany. The clean room was equipped with airlocks and kept at a positive pressure. Sampling was performed in a clean bench with a laminar air flow of 0.45 m/sec to assure ISO 14644-1 clean room classification 5 (equivalent to class 100 Fed. Std. 209E). During sampling, protective clothing such as hats, face masks, gloves, boots and coveralls were worn. The effective background contamination inside the clean bench was determined prior, after and in between the individual measurements. The specified ISO clean room classification 5 was achieved at any time; in most cases ISO class 4 was met.

### **2.3 Blank Sample**

A blank test was performed to describe the background “noise” of the particle collecting system. For the blank test, the system was operated in exactly the same way as during sampling from cigarettes – but without any cigarette inserted in the holder. Blank tests were performed prior and after each sample series and prior to a new sample. The total amount of particles detected for the blank test (0.3 µm to 15.0 µm) was ranging between 0.1 and 0.6 particles/puff. Considering this very low number of particles, the blank concentration was not taken into account.

### **2.4 Particle Counting**

Particle counting was performed using a laser aerosol spectrometer 1.108 (supplier: Grimm GmbH, Ainring, Germany) at a frequency of 1 measurement per second. The detected particles are classified in eight size ranges: 0.3 - 0.4 µm, - 0.5 µm, - 0.65 µm, - 0.8 µm, - 1.0 µm, - 1.6 µm, - 2.0 µm, - 15 µm. The laser aerosol spectrometer was calibrated by the supplier using dolomite dust standards. The intake volume was 1.2 L/min.

### **2.5 Cigarette Holder, Smoking Machine**

Standard cigarette holders with typical labyrinth seals were used as described in ISO 3308. Cigarette and holder were in a horizontal position during “sham smoking”. A Borgwaldt single-port smoking machine was used for sampling under more intense conditions than those defined in ISO 4387: puff volume 55 mL, puff duration 2 seconds, puff interval 15 seconds. For each sample, 20 cigarettes were separately sham smoked with 10 puffs each.

## 2.6 Particle Collecting System and Sampling

Figure 1 schematically describes the main components of the particle collecting system. The cigarette holder is connected with a pump (smoking machine) using silicone tubing. The pump outlet is linked with the aerosol spectrometer using a branch connexion which allows the supply of air through a HEPA (High Efficiency Particulate Air) filter.

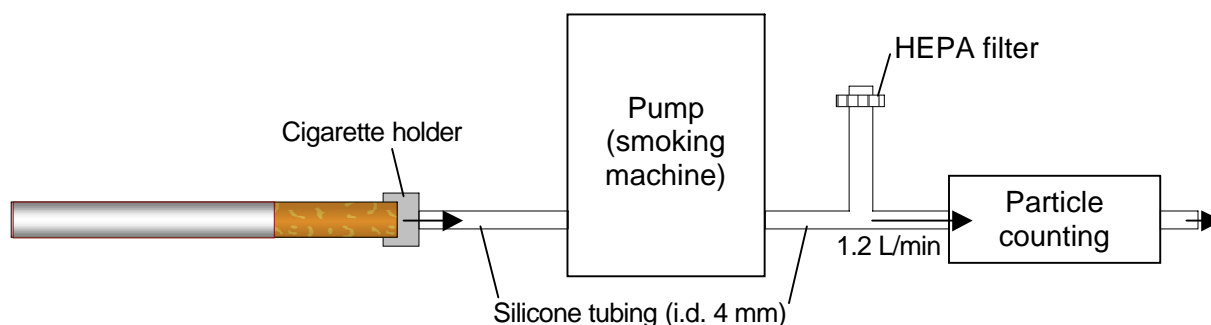


Figure 1: Particle collecting system for on-line methodology.

Using this experimental setup, the particle counter was operated at a constant flow of 1.2 L/min with filtered air provided in between the puffs. Cigarette holder and sample cigarettes should be aligned horizontally. Samples should be handled with care when inserted into the cigarette holder to prevent mechanical strain on the filter.

## 2.7 Calculation of Results

For each sample, twenty replicate measurements were performed and it was noted that the amount of particles released from a specific sample varied considerably between single measurements. For a better interpretation of data, the diagrams below contain the following figures: arithmetic mean, median (50 % percentile), and 95 % confidence interval. For the classification of particles into different size ranges, the amount of particles released per cigarette (10 puffs) is given for each size range. For the analysis on a puff-by-puff basis, the total amount of particles in all size ranges (0.3 – 15.0  $\mu\text{m}$ ) is given for each puff.

## STUDIED MATERIAL

The following American blended test cigarettes have been investigated in this study:

- 'Dalmatian' – filter cigarette with active carbon granules; carbon loading 85 mg/cig (commercial 30/70 mesh grade, 10 % moisture content); 0 % filter ventilation.
- CA control – filter cigarette with mono-acetate filter; 0 % filter ventilation.

Cigarette dimensions: 93 mm length (25 mm filter, 68 mm rod length).

## RESULTS AND DISCUSSION

### 1. ITG Methodology

#### CA control and ‘Dalmatian’ samples

Table 1 summarises the mean figures of three replicate measurements for the CA control and the ‘Dalmatian’ sample after extrapolation to the effective membrane area and expressed as “particles per 20 cigarettes”.

For the CA control cigarettes, the particles found were mainly identified as typical cigarette and filter constituents. Possible contaminants observed for the CA control sample were polyamide and carbon. Some particles could only be counted, but not be identified unambiguously (Fluorescence, Manifold). Most particles had a spherical shape, only a few “non-respirable fibre-shape particulates” (non-RFP) could be detected – but no RFPs.

For the ‘Dalmatian’ type cigarettes, a smaller amount of particles was detected. Five RFP were found and identified as Carbonate (2) and Fluorescence/Manifold (3).

Table 1: Number of particles and size distribution for CA control and ‘Dalmatian’ filters (mean figures of three replicate measurements, shown without decimal places).

| CA control<br>Particles per 20 cigarettes (10 puffs each) |           |                        |           |          |          |          |          | 'Dalmatian' filter<br>Particles per 20 cigarettes (10 puffs each) |                        |           |          |          |          |          |          |          |
|---|-----------|------------------------|-----------|----------|----------|----------|----------|---|------------------------|-----------|----------|----------|----------|----------|----------|----------|
| Substance/Type  | Σ         | Size Distribution [µm] |           |          |          |          |          | Σ   | Size Distribution [µm] |           |          |          |          |          |          |          |
|   |           | 2-5                    | 5-10      | 10-25    | 25-50    | 50-100   | > 100    |   | 2-5                    | 5-10      | 10-25    | 25-50    | 50-100   | > 100    |          |          |
| Polyamide   | 12        | 7                      | 2         | 0        | 3        | 0        | 0        | 0   | 0                      | 0         | 0        | 0        | 0        | 0        | 0        | 0        |
| Titanium(IV)oxid (Anatase)                                | 12        | 9                      | 3         | 0        | 0        | 0        | 0        | 0   | 0                      | 0         | 0        | 0        | 0        | 0        | 0        | 0        |
| Keratin (Protein)   | 5         | 3                      | 0         | 2        | 0        | 0        | 0        | 1   | 1                      | 1         | 0        | 0        | 0        | 0        | 0        | 0        |
| Long chain hydrocarbon                                    | 2         | 0                      | 0         | 2        | 0        | 0        | 0        | 2   | 1                      | 1         | 1        | 0        | 0        | 0        | 0        | 0        |
| Nitrate   | 3         | 3                      | 0         | 0        | 0        | 0        | 0        | 0   | 0                      | 0         | 0        | 0        | 0        | 0        | 0        | 0        |
| Celluloseacetat   | 0         | 0                      | 0         | 0        | 0        | 0        | 0        | 0   | 0                      | 0         | 0        | 0        | 0        | 0        | 0        | 0        |
| (Calcium)carbonate  | 22        | 17                     | 5         | 0        | 0        | 0        | 0        | 23  | 11                     | 7         | 5        | 1        | 0        | 0        | 0        | 0        |
| Cellulose   | 3         | 0                      | 3         | 0        | 0        | 0        | 0        | 0   | 0                      | 0         | 0        | 0        | 0        | 0        | 0        | 0        |
| Carbon  | 10        | 7                      | 2         | 2        | 0        | 0        | 0        | 9   | 3                      | 3         | 3        | 0        | 0        | 0        | 0        | 0        |
| Fluorescence  | 15        | 5                      | 5         | 3        | 2        | 0        | 0        | 3   | 0                      | 1         | 1        | 1        | 1        | 1        | 0        | 0        |
| Manifold  | 0         | 0                      | 0         | 0        | 0        | 0        | 0        | 1   | 0                      | 1         | 0        | 0        | 0        | 0        | 0        | 0        |
| <b>Total</b>  | <b>85</b> | <b>51</b>              | <b>20</b> | <b>9</b> | <b>5</b> | <b>0</b> | <b>0</b> | <b>39</b>   | <b>15</b>              | <b>12</b> | <b>9</b> | <b>2</b> | <b>1</b> | <b>1</b> | <b>0</b> | <b>0</b> |
| <b>RFP</b>  | 0         | 0                      | 0         | 0        | 0        | 0        | 0        | 5   | 0                      | 3         | 2        | 0        | 0        | 0        | 0        | 0        |
| <b>Fiber not RFP</b>                                      | 2         | 0                      | 2         | 0        | 0        | 0        | 0        | 4   | 1                      | 0         | 2        | 1        | 0        | 0        | 0        | 0        |
| <b>Spherical</b>  | 85        | 53                     | 19        | 9        | 5        | 0        | 0        | 30  | 15                     | 9         | 5        | 1        | 1        | 1        | 0        | 0        |

#### Blank test

For the blank test, the system was operated without any cigarettes inserted in the holders and in exactly the same way as for sampling from cigarettes. Thus, the blank test (Table 2) describes the number of particles detected in the filtered ambient air inside the Plexiglas hood (background “noise”).

Table 2: Number of particles and size distribution for blank test (mean figures of three replicates, without decimal places). Yields calculated for 200 puffs ( $\approx$  20 cigarettes, 10 puffs each).

| Blank test (background "noise") - particles per 200 puffs |          |                                     |      |       |       |        |            |
|---|----------|-------------------------------------|------|-------|-------|--------|------------|
| Substance/Type  | $\Sigma$ | Size Distribution [ $\mu\text{m}$ ] |      |       |       |        |            |
|   |          | 2-5                                 | 5-10 | 10-25 | 25-50 | 50-100 | $\geq 100$ |
| Polyamide   | 9        | 5                                   | 0    | 2     | 2     | 0      | 0          |
| Carbon  | 24       | 19                                  | 3    | 2     | 0     | 0      | 0          |
| Keratin (Protein)   | 2        | 0                                   | 0    | 0     | 2     | 0      | 0          |
| Celluloseacetat   | 0        | 0                                   | 0    | 0     | 0     | 0      | 0          |
| (Calcium)carbonate  | 27       | 24                                  | 3    | 0     | 0     | 0      | 0          |
| Fluorescence  | 15       | 7                                   | 3    | 2     | 2     | 2      | 0          |
| Titanium(IV)oxid (Anatase)                                | 3        | 2                                   | 0    | 2     | 0     | 0      | 0          |
| Cellulose   | 0        | 0                                   | 0    | 0     | 0     | 0      | 0          |
| Manifold  | 2        | 2                                   | 0    | 0     | 0     | 0      | 0          |
| Long chain hydrocarbon                                    | 2        | 0                                   | 0    | 2     | 0     | 0      | 0          |
| Nitrate   | 0        | 0                                   | 0    | 0     | 0     | 0      | 0          |
| Total Blank test  | 84       | 58                                  | 10   | 9     | 5     | 2      | 0          |
| RFP   | 2        | 0                                   | 2    | 0     | 0     | 0      | 0          |
| Fiber not RFP   | 0        | 0                                   | 0    | 0     | 0     | 0      | 0          |
| Spherical   | 82       | 58                                  | 9    | 9     | 5     | 2      | 0          |

For the blank test, most particles detected were characterised as calcium carbonate, fluorescence, polyamide and carbon. The latter must be considered a contaminant during sampling/sample handling. The total number of particles detected is comparable to that of the CA control sample. Two fluorescent particles were classified as RFP; all the other particles had a spherical shape.

## 2. Online Methodology (ITCF)

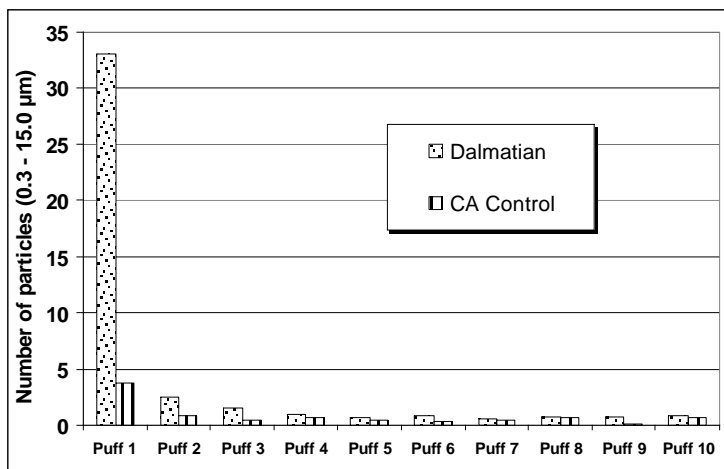
Table 3 shows the number of particles detected for the different samples investigated in this study using the online methodology (twenty replicate measurements each). The blank concentration was not taken into account due to the very low number of particles detected under clean room conditions.

Table 3: Released particles per cigarette and size distribution (mean figures, 10 puffs).

| Number of particles released per sample (mean figures, 10 puffs) |                                 |             |              |              |             |             |             |       |                           |
|--|---------------------------------|-------------|--------------|--------------|-------------|-------------|-------------|-------|---------------------------|
|  | Particle size [ $\mu\text{m}$ ] |             |              |              |             |             |             |       | Total amount of particles |
|  | 0.3 - 0.4                       | > 0.4 - 0.5 | > 0.5 - 0.65 | > 0.65 - 0.8 | > 0.8 - 1.0 | > 1.0 - 1.6 | > 1.6 - 2.0 | > 2.0 |                           |
| Dalmatian  | 19.05                           | 11.30       | 7.50         | 1.75         | 1.50        | 0.80        | 0.25        | 0.45  | 42.60                     |
| CA Control   | 3.60                            | 2.30        | 1.35         | 0.30         | 0.40        | 0.20        | 0.05        | 0.15  | 8.35                      |

It was noted that the amount of particles released from a sample considerably varied between twenty single measurements. The number of particles detected per puff within the total size range 0.3 - 15.0  $\mu\text{m}$  is shown in Figure 2.

Figure 2: Number of particles counted on a per-puff basis (0.3 - 15.0  $\mu\text{m}$ ).



During sham-smoking of 10 puffs, the main amount of particles was released during the first two puffs and seemed to stay on a constant level from the third puff on. Again, it was noted that the amount of particles released from a sample varied considerably between twenty single measurements.

## CONCLUSION

In the present study, the majority of the identified particles were organics such as polyamide, cellulose, cellulose acetate, proteins and activated carbon. Particles containing titanium dioxide and calcium carbonate were also observed. Due to fluorescence, some particles could not be assigned unambiguously. The number of particles detected in the blank test was comparable to the figures of the CA control and ‘Dalmatian’ type filter sample. No “respirable fibre-shaped particulates” (RFP) were found for the CA control sample. For the ‘Dalmatian’ type cigarettes, five RFP were found and identified as Carbonate (2) and Fluorescence/Manifold (3).

The examinations of the release of particulates from cigarette filters should be performed under clean-air room conditions in order to be quantifiable above the ubiquitous background dust concentration in the ambient air. The result achieved in this study show that the particle load released from cigarette filters, based on the smoking volume, is much lower compared to the dust exposure of the usual ambient air and corresponds to clean-air room particle concentrations.



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