Particle size in the smoke produced by six different types of cigarette tobacco

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Summary

Introduction For several decades a more peripheral distribution of the broncho-pulmonary tobacco-related pathologies has been observed.

Methods The aim of this study is to examine whether changes in the particle size of cigarette smoke resulting from new manufacturing technologies could play a part in the observed epidemiological changes through a more distal deposition of smoke particles in the airways. Using a smoking machine and a low pressure electrostatic impactor we measured the particle size of the smoke from six different types of cigarette, representing the older and more modern manufacturing techniques. The effectiveness of filter tips was assessed using a size analyser measuring the electrical mobility of the particles.

Results The results show a difference in particle size between the primary smoke stream inhaled by the smoker, S1 (0.27 +/- 0.03 micrometers) and the secondary stream, S2 inhaled by passive smokers (0.09 +/- 0.01 micrometers). There is no difference in particle size between the 6 different types of cigarette. Filters dilute the smoke without altering particle size.

Conclusion The recent alterations in the distribution of tobacco related pathologies cannot be explained by changes in particle size in cigarette smoke. The explanation has to be sought elsewhere.

Key-words: Tobacco smoke • Aerosol particle sizing • Lung deposition.
Introduction

The manufacturing process and the composition of cigarette tobacco have substantially changed over the last fifty years. In Europe and the United States for example, there has been a threefold decrease in the nicotine and tar content. Over 400 chemical additives have been authorised in commercial cigarettes and filters have been added.

Although there has been no significant change in the mortality rate [1, 2, 3], tobacco-related diseases of the lungs and bronchi are now observed in more distal sites [4, 5]. However, the facts show that:

– the relative incidence of cancer of the small calibre airways has increased,
– the relative incidence of emphysematous disorders has increased.

We therefore put forward the hypothesis that smaller sized particles in cigarette smoke, may play a role in these anatomico-pathological changes because they are deposited in the more distal parts of the airways.

There are 3 air streams in cigarette smoke:

– the primary stream, S1, inhaled by the smoker when the smoke is breathed in;
– the secondary stream or side stream, S2, produced by slow combustion of the cigarette between the active puffs taken in by the smoker; S2 is released into the ambient air then inhaled by the smoker or any passive smokers in the vicinity;
– the tertiary stream, S3, exhaled by the smoker; this stream mixes with the previous one, but is not the subject of this study.

We used a smoking machine to study the distribution of particle sizes in cigarette smoke and for this purpose we took S1 and S2 measurements for six different types of cigarettes: dark or light tobacco, with or without filters, manufactured using the older and more modern technologies.

According to the studies in the literature, cigarette smoke has a particle size spectrum ranging from a nanometer to a few micrometers [6, 7, 8, 9, 10]. For this reason we used a 13 plate electrostatic impactor, which enabled us to measure the particle sizes below one micron that reach the distal parts of the airways.

Materials and Methods

The characteristics of the six types of cigarettes studied are as follows:

RB: Dark tobacco with a high tar content, above the European standards: Russian Dark (Prima®). Tar: 13 mg/cig; Nicotine: 0.7 mg/cig. No filter tip.

G: Mild, dark tobacco (Gauloises®): Tar: 10 mg/cig; Nicotine: 0.6 mg/cig. No filter tip.

MRF: Light tobacco (Marlboro® red) with filter tips. tars: 10 mg/cig; Nicotine: 0.8 mg/cig.

MRNF: Light tobacco (Marlboro® red) no filter tip. tars: 10 mg/cig; Nicotine: 0.8 mg/cig.

MLF: Mild, light tobacco (Marlboro® white) with filter tips. tars: 8 mg/cig; Nicotine: 0.6 mg/cig.

MLNF: Mild, light tobacco (Marlboro® white) no filter tip. tars: 8 mg/cig; Nicotine: 0.6 mg/cig.

Smoking machine [10]

The smoking machine (fig. 1) is a lung model with a 6 litre cylindrical reservoir in transparent plastic half filled...
with water at 37°C controlled by a thermostat and agitated to ensure that the air above the water level is saturated. A variable capacity and frequency cylinder and piston system is used to simulate breathing in and out through two apertures equipped with one-way valves located in the upper part of the reservoir [11]. Using this machine, we were able to study the primary or main smoke stream, S1, inhaled lengthways through the cigarette when the subject breathes in.

**Electrostatic low pressure impactor (ELPI)**

The size and concentration of the particles were measured with a 13 plate cascade impactor capable of measuring a very wide range of particles sizes [12]. The apparatus operates at low air-flow pressures and is equipped with an electrostatic detector to trap ultra-fine loaded particles; it will thus detect particle sizes ranging from 0.028 to 10 µm. The samples are taken at a flowrate of 10 l.min⁻¹.

The ELPI impactor (Dekati Ltd, Tampere, Finland) gives an almost instantaneous particle size reading (<5 seconds); its performances and applications have been published in the literature [13-14].

**Measurement of particle sizes**

The smoking machine was set at a frequency of 12 cycles per minute and a volume of 500 ml BTPS (Body Temperature Pressure Saturated) to simulate a human adult subject’s mean resting ventilation rate. During the combustion of one cigarette (around one minute):

- stream S1 was sampled by placing a T-tube between the cigarette and the smoking machine.
- stream S2 was sampled directly above the incandescent end of the cigarette.

The reproducibility of the measurements was checked by repeating them three times for each type of cigarette in each of the conditions of the study.

**Processing the data supplied by ELPI**

The particles trapped on the 13 collection plates of the impactor were counted on 12 digital channels using the ELPI XLS software package (sizes between 0.028 and 10 µm). The size of a particle, measured by impaction, is equivalent to its aerodynamic diameter. The diameters and number of particles are presented in cumulative frequency of distribution and in total particle numbers for each measurement.

The following standard calculations are usually made from the cumulative distributions of frequency: D50 (median diameter), D84.3 (84.3% of the particles are smaller than this diameter), standard geometric deviation (σg), D84.3/ D50 ratio. The particles are then classified in three size categories:

- Below 0.3 µm (microparticles)
  - From 0.3 to 2 µm, (particles that penetrate deeply into the lungs)
  - Above 2 µm.

This classification was selected in reference to the "Particulate Matter Sizes" used to measure atmospheric pollution so that it was compatible with the values supplied by the digital channels. We have allocated a percentage of the total number to each category.

**Statistical analysis of the results**

The mean values and the standard geometric variations (σg) in particle size were calculated for each series of measurements and used to compare the relative value (in percentiles) of each of the three categories. We chose to use the British Standards Institution reproducibility coefficient (two standard deviations). To compare the different brands we used the Student t test, when possible. The ranked Mann and Whitney U tests were used when the respective data numbers were too limited to make the standard calculation of statistical significance [15].

**Measuring the efficiency of a cigarette filter**

The efficiency of a cigarette filter is defined as a ratio between the number of particles in the primary air stream inhaled and the number of particles that are sent into the filter. To make this evaluation we used diethylsebacate (DES) particles forming a poly-disperse, calibrated aerosol produced by a Collison atomiser then sent through an MRF and MLF cigarette. A krypton 85 radioactive source was used to neutralise the loaded particles, then their size distribution was measured in a TSI SMPS (Scanning mobility particle sizer) before and after the filter tip. In the SMPS [16], a differential mobility analyser or DMA (TSI 3081©) that sizes particles according to their electrical mobility was added to a nucleus condensation counter (NCC) in which the micro-particles are proportionately increased in size by condensing them with alcohol fumes until they can be measured (TSI 3022©). The SMPS detects particles in a range between 0.01 et 1 µm. The flow-rate of the smoke through the filter was 25 ml per second.

**Results**

**Particle diameters (table I)**

1. The results show a significant difference between the D50 values for air streams S1 and S2 whatever the type of cigarette.
   - For S1, the D50 values range between 0.21 and 0.29 µm (mean = 0.27 µm)
For S2, the D50 values range between 0.08 and 0.11 µm (mean = 0.09 µm).

However, no difference was found between the different types of cigarette when the D50 values were compared for S1 and S2.

2- Particulate distribution in the size categories (table I and figure 2).

The number of particles in the size category above 2µm was close to zero for both air streams. 69.6 % ± 13.8 of the S1 particles and 95.8 % ± 4.4 of those in S2 are below 0.3 µm. 30.4% ± 14.8 of the S1 particles and 4.2% ± 4.1 of those in S2 fell into the intermediate category between 0.3 and 2 µm.

Filtering efficiency of a cigarette filter tip

The diethylsebacate, (DES) calibrated particle concentration measurements made upstream and downstream from the filter tip showed that all the particles under 0.25 µm pass through the filter tip (fig. 3).

The measurements made with the ELPI counter confirmed these results: there is no difference in the distribution of the particle size in S1 for MR and ML cigarettes whether they have filter tips or not.

Filter tips on cigarettes dilute the smoke with the air that is let in through the sides of the filter tip and through the pores of the paper casing, but do not trap these particles because they are too fine. A small proportion of the tar particles are deposited on them, so they do have some protective effect, although it is limited.

Discussion

This study was undertaken to ascertain whether the changes in cigarette manufacturing technology had led to any change in the particle size of the aerosol in the smoke they emit. Such changes could be the cause of the differences observed in tobacco-related diseases. For this reason, we selected a Russian brand of cigarettes (Prima) that is representative of the “older”, dark type of cigarette tobacco (high tar content, no filter tip, very few additives), an American brand of cigarettes (Marlboro “normal” and “light”) representing the more modern cigarette manufacturing technology (intermediate tar content, with or without filter tips and with many additives: 9%) and a French brand of cigarettes (Gauloises) representing the milder, intermediate type of dark tobacco (intermediate tar content, no filter tip and few additives: 4.5%).

In our study, the particle diameters (D50) measured in the main smoke stream S1, (0.21 to 0.29 µm, with almost 70% of the particles below 0.3 µm in diameter) agree with those in the literature: 0.18 to 0.34 µm in Bernstein’s review [6], 0.45 µm for Chen et al.[7] with the research cigarette, Kentucky 2R1; 0.54 µm for Morawska et al. [8], 0.2 and 0.5 µm for Hiller et al.[9] ,0.6 µm for Mc Cusker et al. [10] and 0.38 µm for Anderson et al. [17].

The mean size of the S2 particles are much smaller than those in S1, with a D50 of 0.09 µm, and almost 96% of them are under 0.3 µm in size. These results are similar to those obtained by Esquier [18] and Morawska and Phillips. [19].

Table I

<table>
<thead>
<tr>
<th>Brand of cigarette</th>
<th>D50 (µm)</th>
<th>σg (µm)</th>
<th>Nb of particles</th>
<th>D50 (µm)</th>
<th>σg (µm)</th>
<th>Nb of particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>MLF</td>
<td>0.29±0.04</td>
<td>2.26±0.49</td>
<td>68±13</td>
<td>0.11±0.04</td>
<td>2.67±1.23</td>
<td>95±6</td>
</tr>
<tr>
<td>MLNF</td>
<td>0.21±0.05</td>
<td>2.55±0.29</td>
<td>73±12</td>
<td>0.10±0.03</td>
<td>3.7±0.7</td>
<td>97±4</td>
</tr>
<tr>
<td>MRF</td>
<td>0.29±0.08</td>
<td>2.37±0.57</td>
<td>65±16</td>
<td>0.09±0.02</td>
<td>4.81±1.29</td>
<td>92±14</td>
</tr>
<tr>
<td>MRNF</td>
<td>0.25±0.09</td>
<td>2.67±0.40</td>
<td>77±17</td>
<td>0.09±0.02</td>
<td>3.58±1.66</td>
<td>96±1</td>
</tr>
<tr>
<td>G</td>
<td>0.29±0.06</td>
<td>2.87±0.61</td>
<td>75±14</td>
<td>0.08±0.02</td>
<td>4.56±1.44</td>
<td>97±2</td>
</tr>
<tr>
<td>RB</td>
<td>0.25±0.06</td>
<td>2.74±0.45</td>
<td>75±11</td>
<td>0.08±0.02</td>
<td>3.48±0.99</td>
<td>98±1</td>
</tr>
<tr>
<td>mean for the 6 brands</td>
<td>0.27±0.03</td>
<td>2.46±0.23</td>
<td>69.6±13.8</td>
<td>0.09±0.01</td>
<td>3.05±0.78</td>
<td>95.8±4.4</td>
</tr>
</tbody>
</table>

(a) no significant difference between the types of cigarette (Mann et Whitney U test)
According to the models developed by the International Commission on Radiological Protection (ICRP) [20], the probability of 0.3 μm S1 particles breathed in through the mouth depositing in an adult is close to 25% for all the airways, including 15% in the alveoli.

Experimentally, the percentage of the aerosol of cigarette smoke that is deposited in the airways varies substantially according to the authors: 47% (22-75) in 11 smokers for Hinds et al. [21] and 70 to 90% in 5 smokers for Hiller et al. [8]. For Martonen [22], these values could be even...
higher if the puff of smoke is not immediately diluted when inhaled, increasing the particle size by hygroscopic effect and aggregation; in that case the particles would be deposited in the proximal bronchi.

The probability of the 0.1 µm particles contained in the S2 stream that cause passive smoking being deposited in an adult breathing through the nose is 35% for all the airways and 22% of this amount would be deposited in the alveoli; whereas in a child aged one, these values would be 41 and 25%.

Hiller et al. [23] assessed the total value of the S2 stream deposited in the airways of adults at 11%; the value was 7 to 20% for Muller et al. [24].

These differences in the experimental values show that individual variability in the deposition of these particles is probably quite high for both S1 and S2 using a model based on particle size.

If one accepts that the different types of cigarettes tested are representative of the older, more modern or intermediate tobacco types, it is obvious that the measurements made on S1 and S2 do not show any major difference in the particle size and therefore the more distal locations found in broncho-pulmonary disorders cannot be explained by a finer particle size in cigarette smoke.

The more distal deposition of S1, may also depend on the way smoke is inhaled by the smoker: when breathing volume increases, for the same particle size the aerosol is deposited deeper in the distal parts of the bronchi and the alveoli rather than in the proximal bronchi as Heyder et al demonstrated [25]; a slight pause in the breathing pattern also enhances particle deposition.

Modern tobaccos are less strong and less irritating for the bronchi, mainly because they have a high pH (>6) and contain additives. This may have changed smoking habits [6]. Apart from the smokers who only draw the smoke into their mouths, without inhaling it, there are two different ways of inhaling [26, 27]:

Most smokers draw on their cigarette and take a puff of smoke into their mouth, then after a pause of 1 to 4 seconds they inhale it quickly into their lungs. The others inhale the smoke directly into the depths of their lungs.

With the older, high tar and nicotine content cigarettes, one can assume that most smokers used the first method, since inhaling the smoke directly would have had a highly irritant effect; the holding phase in the mouth allowed the particles to increase in size through hydration and aggregation before they were drawn down into the airways per se [28].

Cigarettes with a lower tar and nicotine content (until recently known as "mild") are less irritant and easier to inhale directly into the lungs, deeper and longer, thus enhancing the access and deposition of particles into the distal airways [20]. Medici et al. [28] observed that direct inhalation seemed to be common in emphysema patients and that the duration of the intra-buccal phase was close to zero (0.08 seconds) in these subjects.

When we compared the S1 streams of cigarettes with and without filters we did not find any difference in particle size (D50, and distribution in both size categories); this agrees with the findings of Anderson et al. [17]. It would seem that the smoke is not filtered, but rather diluted by the ambient air let in by the pores located around the filter and at its base [9]. This induces a specific compensatory mechanism in smokers who tend to increase the volume of smoke they draw in at each puff, breathing in more deeply to inhale the same amount of smoke [5]; this manoeuvre also tends to send the smoke farther down into the lungs. While the mean volume of a puff varies from 35 to 53 ml according to the authors [21], the increase of this volume due to the filter tip is probably 20 to 50% and can peak at 75 ml. However, this increase is not enough to correct the dilution of the smoke in a volume of air taken into the lungs that is around the mean tidal volume (500 ml) and this could explain why we observed smokers blocking the pores of the filter with their fingers while they inhaled, thus increasing the nicotine and tar concentrations in the air they inhale.

**Conclusion**

The particle sizes in the S1 air stream are larger than those in S2, which is the main cause of passive smoking; however, there is no difference between these air streams in six types of commercial cigarettes whether they have filter tips or not, although the tar, nicotine and additive contents are different. The filter probably dilutes the smoke when it is inhaled by the smoker, but has no effect in retaining the particles contained in the smoke because they are too fine. Thus, our study does not enable us to incriminate the
change in particle size in the smoke produced by different types of cigarettes in the evolution of tobacco-related diseases. This evolution must therefore be explained by a change in the chemical characteristics of the aerosol (changes in the tar and nicotine content, addition of numerous chemical substances) or by a change in the smokers’ behaviour (inhaling larger puffs that penetrate deeper and longer into the airways) which has been made possible by the less irritating smoke of modern cigarettes.

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References