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Particulate matter emissions of four different types of one cigarette brand with and without additives

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Abstract:

Objective: Inhaled Particulate Matter (PM) in second-hand smoke, also called environmental tobacco smoke (ETS), is deleterious for smokers and non-smokers. Different additives in cigarettes like aromatics and humectant agents might have influence on the amount of PM. To investigate an effect of additives on the concentration of PM (PM_{10}, $PM_{2.5}$ and PM_1) ETS of four different types of cigarettes of the brand Lucky Strike, each two with (Original Red, Original Blue) and without (Straight Red, Straight Blue) additives, were analysed in comparison to the 3R4F reference cigarette.

Design: Experimental

Setting: An automatic environmental tobacco smoke emitter (AETSE) generated ETS in an enclosed room with a volume of $2.88m^3$, followed by laser aerosol spectrometry.

Results: The Lucky Strike Straight Blue, a cigarette type without additives and lower tar amount, shows 10% to 25% lower $PM_{2.5}$ mean values compared to the other tested Lucky Strike products, but 27% higher mean values than the reference cigarette. The $PM_{2.5}$ mean of all measured smoke-free baseline values, that means clean air, was $1.6\mu g/m^3$. This increased up to about $1800\mu g/m^3$ for the reference cigarette and about $3070\mu g/m^3$ for the Lucky Strike Original Blue.

Conclusions: These results are possibly an indication that additives in tobacco products increase the PM amount in ETS. For validation, further comparative studies are necessary that are focusing on the comparison of the PM concentration of cigarettes with and without additives. In general, this study showed the massive increase of PM amount by smoking cigarettes in enclosed rooms.

Implications:

Due to the exposure to second-hand smoke 890,000 people die each year worldwide. Particulate matter (PM) in environmental tobacco smoke (ETS) endangers the health of both non-smokers and smokers. This study considers the effect of additives like aromatics and humectant agents in cigarettes on PM in ETS. Do additives increase the amount of PM?

Keywords:

Environmental tobacco smoke; particulate matter; additives; aromatics; humectant agents

Strength and limitations of this study:

- We are using an automatic environmental tobacco smoke emitter (AETSE) with a standardised smoking protocol that enables us to generate reproducible and reliable particulate matter (PM) levels.
- Passive smoking endangers the health of non-smokers worldwide. Herewith is the PM in cigarette smoke an important factor. The application of the applied methodology allows the comparison of different types of tobacco products, in this case cigarettes of the brand Lucky Strike with and without additives. The aim is to measure the influence of additives in the amount of PM.

- Hence, the developed technique applied in this study, is not able to imitate the human smoking behavior accurate in every detail.
- Nevertheless, its mechanism simulates reliable and comparable conditions, by avoiding subjective deviations. Do that the findings are absolute trustworthy without endangering test-persons.

Introduction:

Airborne particulate matter (PM) as a part of air pollution causes tremendous adverse health effects especially cardiovascular and respiratory diseases (1) and aggravates airway inflammation and hyperresponsiveness in asthmatic patients (2). The exposure to PM is also associated with increased risk of ischemic stroke (3) and breast cancer mortality (4). Several studies have shown that morbidity and mortality increase in relation to PM exposure (5). PM is a differently sized mixture of solid and liquid particles that varies in composition and origin (6). One option to classify PM is by the size of the particles, that determines how deep they penetrate the respiratory system. The smaller the particles the deeper they penetrate (7, 8). Indeed, the size-related definition of PM is the most relevant one. The U.S. Environmental Protection Agency (EPA) differentiates between $PM_{10,}$ inhalable coarse particles equal or smaller than 10µm, and $PM_{2.5,}$ fine inhalable particles equal or smaller than 2.5µm (9). In addition, PM_1 is the fraction of particles equal or smaller than 1µm. The majority of PM derives from tobacco smoke (10). Worldwide approximately one billion adults smoke (11). Each year more than 7 million people are killed due to tobacco use, and 890,000 of those are non-smokers being exposed to secondhand smoke (12) that is also called environmental tobacco smoke (ETS). ETS is a mixture of mainly side-stream smoke emitted from the smoldering tobacco product and the exhaled mainstream smoke from the smoker (13, 14) and is considered to be the major risk factor for air pollution in indoor spaces (15).

Previous studies showed variations of PM levels within different brands and types of cigarettes (16-18). Different additives, e.g. aromatics, humectant agents and the content of tar and nicotine might affect the amount of PM (19). Based on these conclusions it seems to be reasonable and necessary to compare different cigarette types of a special brand with and without additives. Therefore, the particle size fractions of PM_{10}, $PM_{2.5}$ and PM_1 of four specific types of the popular cigarette brand Lucky Strike (20) as well as the ones of the reference cigarette 3R4F developed by the Kentucky Tobacco Research and Development

Center (University of Kentucky, USA) (21) were analysed. The four cigarette types of Lucky Strike were Original Red and Original Blue (with additives) (22, 23), and Straight Red and Straight Blue (without additives) (24, 25). They differ among others in their content of tar, nicotine, carbon monoxide and additives as shown in table 1.

Methods:

The tested tobacco products from Lucky Strike were Original Red, Original Blue, Straight Red and Straight Blue. Straight Red and Straight Blue contain no additives like aromatics and humectant agents. For more extensive information, it is referred to the Federal Ministry of Food and Agriculture of Germany (*Bundesministerium für Ernährung und Landwirtschaft*) (22-25)

Nineteen cigarettes of four tobacco products from Lucky Strike, each with and without additives, and one reference cigarette were smoked using an automatic environmental tobacco smoke emitter (AETSE). The various PM-levels (PM_{10}, $PM_{2.5}$, PM_1) were recorded and evaluated. A modified smoking protocol according to the Tobacco Smoke Particles and Indoor Air Quality (ToPIQ) studies (16, 26) was used.

In a glass chamber with a volume of 2.88m^3 the AETSE was located. This smoke pump for medical research, designed and engineered by Schimpf-Ing, Trondheim, Norway (27), allows the smoking of tobacco products in a reproducible way without exposing test persons and the investigator. A stepper motor moved via a linear actuator a 200ml glass syringe for imitating the smoking process. The puff volume (40ml), puff flow rate (13ml/sec), puff frequency (2/min), inter puff interval (22sec) and the amount of 9 puffs were adjusted by a microcontroller. The smoking protocol was departed in four different phases of five minutes each and started with the pre-ignition phase and measurement of the baseline values. After the cigarette was lighted and smoked in the combustion phase, followed by the post-combustion phase, that started with the extinguishing of the cigarette. In the final suction phase, the indoor air was cleaned by using an industrial suction device, before the next cycle started. So, each cycle lasted 20min. The PM concentrations in the chamber were measured in a dilution of 1:10 with compressed air by a Grimm Portable Laser Aerosol Spectrometer and Dust Monitor model 1.109 (28, 29). The dilution with air was necessary to protect the spectrometer against blockage of the laser measuring chamber by high particle concentrations and subsequently, in the data processing the dilution ratio was considered. The spectrometer detected via light scattering airborne particles with a size range from 0.25µm to 32µm. It displayed the output of measurement data as particle count [l^{-1}], detailed dust mass fractions in 31 channels [$\mu g/m^3$], output of the measurement results important for occupational health according to European standard EN 481, inhalable, thoracic and alveolic [$\mu g/m^3$] (30) and the PM_{10}, $PM_{2.5}$ and PM_1 values according to EPA (9) in real-time. The received data was recorded every six seconds. The PM values were analysed using the area under the curve (AUC) and the mean concentration (C_{mean}) referring to the mean value of 19 cigarettes of each type. In this study, the AUC describes the area under a concentration-time curve in the five minutes lasting interval from ignition to extinction of a cigarette. A test for Gaussian normality was performed and all cigarette type samples passed the test. Afterwards the two sample t-test was performed to assess the differences between respectively two cigarette brands.

Results:

All results of the AUC-PM values and the C_{mean} values allow a direct comparison of the PM emission differences among the cigarette brands, because all tobacco products were tested exactly the same way with identical suction volume, speed and puff amount. The results of the AUC-PM values are shown in figure 1, and the results of the C_{mean} values are shown in table 2. The data of the AUC-PM of all types of the Lucky Strike cigarettes were 21-71% higher than the data of the reference cigarette 3R4F. The values of C_{mean}-PM of all types of Lucky Strike cigarettes were 22-71% higher than the values of C_{mean}-PM of the reference cigarette 3R4F as well. Furthermore, the AUC and the C_{mean} of PM_1, $PM_{2.5}$ and PM_{10} of the Lucky Strike brands Original Red, Original Blue, Straight Red and Straight Blue showed a high statistical significance compared to the AUC-PM and C_{mean} of the reference cigarette 3R4F.

More specifically, the Straight Blue, a tobacco product without additives and lower tar amount (see table 1), shows 10 to 25% lower PM mean values compared to the other tested Lucky Strike products. All differences are highly significant ($p<0.05$). The Straight Blue showed also less PM values than the Original Red (with additives and higher tar amount) but no statistical significance ($p>0.05$). Details are shown in table 3. In comparison to the reference cigarette, the AUC-PM_1 mean values were only 21%, AUC-$PM_{2.5}$ and AUC-PM_{10} 27% higher. Accordingly, the C_{mean} values of PM_1, $PM_{2.5}$ and PM_{10} of the Straight Blue were 22%, 27% and 28% higher than the values of the reference cigarette.

In contrast, the PM mean values as well as the C_{mean} values of the Lucky Strike Original Blue, a cigarette with additives, but the same tar amount as the Straight Blue, were substantial higher with 63%, 70% and 71% higher values for C_{mean} and AUC of PM_1, $PM_{2.5}$ and PM_{10} compared to the reference cigarette. The measuring data of the Lucky Strike Straight Red and Original Red were in between with 44% higher C_{mean} values of PM_1 and 50% higher C_{mean} values of $PM_{2.5}$ and PM_{10} of the Straight Red, and 37% higher C_{mean} values of PM_1 and 42%

higher C_{mean} values of $PM_{2.5}$ and PM_{10} of the Original Red compared to the reference cigarette. According to this, the AUC mean values of the Straight Red were 43% (PM_1) and 50% ($PM_{2.5}$ and PM_{10}), and of the Original Red 36% (PM_1), 41% ($PM_{2.5}$) and 42% (PM_{10}) higher.

The fact, that the additive-free Straight Blue with a lower tar amount showed the lowest measurement values of all four Lucky Strike cigarette products, suggests that additives in cigarettes in combination with a higher tar amount increase the PM emissions.

The PM mean of all measured baseline values, that means clean air, was 1.6µg/m³. The measured C_{mean} of PM_{10} increased up to 1803µg/m³ (± 320µg/m³) for the reference cigarette and 3076µg/m³ (± 321µg/m³) for the Lucky Strike Original Blue and in case of $PM_{2.5}$ up to 1801µg/m³ (± 320µg/m³) respectively 3068µg/m³ (± 319µg/m³). For PM_1 the values raised up to 1762 µg/m³ (± 302µg/m³) respectively 2865µg/m³ (± 282µg/m³).

Discussion:

Different studies show a hazardous increase of PM levels in smoking rooms and households (31) (32) (33) (34). The results of our study show, that tobacco smoke in an enclosed space of $2.88m^3$ (capacity of the measuring cabin) increased the particulate matter emissions compared to smoke free air (baseline values) more than a 1000-fold. The measured $PM_{2.5}$ values exceeded the daily average concentration of at most $25µg/m^3$ according to the *WHO Air quality guidelines* (35) approximately 70- to 120-fold, depending on the cigarette brand. For example, a compact car, classified by the EPA with a total passenger and cargo volume of $2.832m^3$ to $3.087m^3$ (36), has a comparable indoor volume. This is a fundamentally important aspect of our study design, because many people smoke in cars. The passive smoke with the contained particulate matter is not only hazardous to the health of smokers but also of passengers, which are often children.

In addition, the tested tobacco product without additives and lower tar amount (Lucky Strike Straight Blue) emitted less particulate matter than the cigarette with no additives but higher tar amount (Straight Red), respectively the cigarette with additives but lower tar amount (Original Blue).

The results lead to the assumption that cigarettes without additives emit less particulate matter. In contrast, the Straight Red and the Original Red had similar PM values. Hence, it could not be ascertained beyond doubt that additive free cigarettes emit less PM. So, further studies are necessary to prove the assumption that cigarettes without additives and lower tar amount emit less PM.

Only a few studies are published with respect to an effect of additives on PM with contradictory conclusions. In 2002, Rustemeier et al. (19) performed a study, in which 333 commonly used ingredients were added in three different groups to the 1R4F reference cigarette. The results showed an increase of 13-28% of PM relative to the cigarettes without added additives. In 2011 Wertz et al. (37) analysed previously secret tobacco industry documents and found among others four peer reviewed publications which concluded no evidence of substantial toxicity as well as total particulate matter (TPM). They ascertained that internal documents of the tobacco industry showed post hoc changes in protocols after primary statistical findings of an additive-associated increase in toxicity and TPM concentrations in cigarette smoke with additives. No significant difference of PM of cigarettes with and without the additive menthol showed the studies of Gaworski et al. (38) and Gerharz et al. (39). Similar results were described by Wasel et al. (17). They found no significant differences in the PM amount of L&M cigarettes with and without additives. But higher PM values of the L&M filtered cigarillo could be shown.

In this study, by far the largest part of PM is represented by particles smaller or equal than 1µm and larger than 0.25µm. Already in 1960, Keith and Derrick (40) published similar results that most of the particles in tobacco smoke has a size between 0.1µm and 1µm peaked between 0.2µm and 0.25µm. Nazaroff and Klepeis (41) described ETS with a particle size between 0.02µm and 2µm in diameter. There is no common agreement on the peak size. On the one hand side-stream smoke particles were described with geometric mean diameters of 0.1µm (42, 43). As opposed to that, Haustein and Groneberg (44) reported mean diameters of 0.5µm. The technical limitation of the used aerosol spectrometer Grimm model 1.109 to detect particles with a minimum size of 0.25µm resulted in a nonconformity with the EPA definition, where particles down to 0.1µm are included. Here it must be mentioned that the used laser aerosolsprectrometer, built for continuous measurement of PM and also common used in monitoring networks (45), has a measuring range from 0.25µm to 30µm. This limitation effected that a proportion of PM_1 could not be measured and for particles smaller than 0.25µm a new measurement system is essential. In addition, the AETSE is not able to imitate exactly ETS and the smoking behavior of humans, because in the respiratory tract the inhaled mainstream smoke is humidified and due to hygroscopic growth the exhaled smoke particles are approximately 1.5-fold larger (46, 47). By using the AETSE it is not possible to differentiate between inhaled and exhaled mainstream smoke, but it should be enhanced that ETS consists only of approx. 15% mainstream smoke and approx. 85% side-stream smoke (48, 49). However, the AETSE is able to exactly imitate side-stream smoke and, hence, the measured PM emissions are very similar to ETS. It is worth pointing out that reproducible results have been ensured by avoiding inter-individual deviations without the exposure of a test person to any health risk. The used modified smoking regime differed from the already existing protocols like, e.g. ISO/TR 17219 (50) or the *Standard operating procedure for intense smoking of cigarettes* by the WHO (51). On that point it must be said that no "gold standard" exists for smoking regimes (52-55). Moreover, this study as well as all previous ToPIQ-studies focuses on data comparison to the 3R4F reference cigarette and not on absolute numbers. Therefore, the use of the modified protocol and the application of AETSE can be considered as valid.

Author Contributions: This article is part of the thesis of Elsa-Luise Fromm, whereas Markus Braun, Alexander Gerber, Ruth Müller and David A. Groneberg contributed significantly to the conception and design of the study. Moreover, they prepared the experiments, which were performed by Elsa-Luise Fromm. Elsa-Luise Fromm and Alexander Gerber analysed the data. The manuscript was written by Markus Braun and critically reviewed by all authors. All authors have participated sufficiently in the work to take public responsibility for appropriate portions of the content and agreed to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved. All authors have read and approved the final manuscript.

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Patient involvement: Patients were not involved

Ethics approval: Not required

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Figure legends:

Figure 1: Comparison of AUC (PM_{10}, $PM_{2.5}$ and PM_1) for all tested tobacco products.

Table 1: List of the cigarette ingredients with reference to tar, nicotine, carbon monoxide and additives (21-25). H.A. = Humectant Agent. A. = Aromatic

	3R4F Reference cigarette	Lucky Strike Original Red	Lucky Strike Original Blue	Lucky Strike Straight Red	Lucky Strike Straight Blue
Tar [mg]	9.4	10	7	10	7
Nicotine [mg]	0.73	0.8	0.6	0.9	0.6
Carbon monoxide [mg]	12	10	8	10	8
Glycerin (H.A.)	yes	yes	yes	no	no
Sugar (A.)	yes	yes	yes	no	no
Cocoa Powder (A.)	n/s	yes	yes	no	no
Licorice Extract (A.)	n/s	yes	yes	no	no
Flavours below 0.1% w/w (A.)	n/s	yes	yes	no	no

Table 2: Mean concentrations (C_{mean} PM_{10}, $PM_{2.5}$ and PM_1) with standard deviation of all tested tobacco products.

	3R4F Reference cigarette	Lucky Strike Original Red	Lucky Strike Original Blue	Lucky Strike Straight Red	Lucky Strike Straight Blue
Cmean PM10 [µg/m³]	1803 ± 320	2557 ± 726	3076 ± 321	2704 ± 261	2300 ± 340
Cmean PM2.5 [µg/m³]	1801 ± 320	2550 ± 724	3068 ± 319	2700 ± 261	2294 ± 339
Cmean PM1 [µg/m³]	1762 ± 302	2402 ± 624	2865 ± 282	2530 ± 231	2145 ± 306

Table 3: P-values of statistical two sample t-test of C_{mean} (PM_{10}, $PM_{2.5}$ and PM_1) for the Lucky Strike brands.

	Original Red vs. Original Blue	**Original Red vs. Straight Red**	**Original Red vs. Straight Blue**	**Original Blue vs. Straight Red**	**Original Blue vs. Straight Blue**	**Straight Red vs. Straight Blue**
p-value Cmean PM10	0.0072	0.4097	0.1713	0.0004	<0.0001	0.0002
p-value Cmean PM2.5	0.0072	0.4016	0.5237	0.0004	<0.0001	0.0002
p-value Cmean PM1	0.0067	0.4183	0.1243	0.0003	<0.0001	<0.0001
p<0.05	yes	no	no	yes	yes	yes

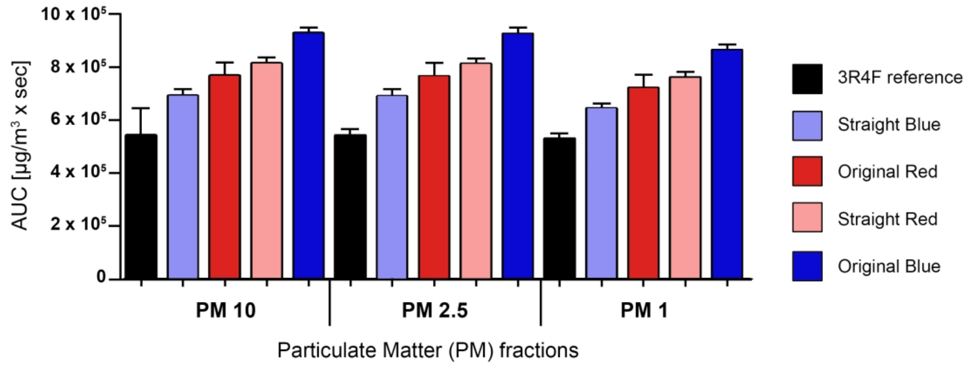

Figure 1: Comparison of AUC (PM10, PM2.5 and PM1) for all tested tobacco products.

234x93mm (300 x 300 DPI)

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Particulate matter emissions of four types of one cigarette brand with and without additives: a laser spectrometric particulate matter analysis of second-hand smoke

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Particulate matter emissions of four types of one cigarette brand with and without additives: a laser spectrometric particulate matter analysis of second-hand smoke

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Abstract:

Objective: Inhaled Particulate Matter (PM) in second-hand smoke (SHS) is deleterious for smokers and non-smokers. Different additives in cigarettes might have influence on the amount of PM. The aim of this study was to assess the influence of additives on the PM emissions from different cigarette types in SHS.

Design: An experimental study of measuring SHS of cigarettes without exposing any person.

Method: The concentrations of PM (PM_{10}, $PM_{2.5}$ and PM_1) of four different types of cigarettes of the brand Lucky Strike, two types with additives (Original Red, Original Blue) and two types without additives (Straight Red, Straight Blue), were analysed in comparison to the 3R4F reference cigarette. An automatic environmental tobacco smoke emitter (AETSE) generated SHS in an enclosed room with a volume of 2.88 m^3, followed by measuring of PM with a laser aerosol spectrometer (Grimm model 1.109). Afterwards, the measuring values of the four test cigarette brands and the reference cigarette were statistically analysed.

Results: The Lucky Strike Straight Blue, a cigarette type without additives and lower tar amount, shows 10 % to 25 % lower PM mean values compared to the other tested Lucky Strike products, but 21 % (PM_1) respectively 27 % ($PM_{2.5,}PM_{10}$) higher mean values than the reference cigarette. The PM mean of all measured smoke-free baseline values, that means clean air, was 1.6 µg/m³. This increased up to about 1800 µg/m³ for the reference cigarette and about 3070 µg/m³ for the Lucky Strike Original Blue.

Conclusions: These results are possibly an indication that additives in tobacco products increase the PM amount in SHS. For validation, further comparative studies are necessary that are focusing on the comparison of the PM concentration of cigarettes with and without additives. This study showed the massive increase of PM amount by smoking cigarettes in enclosed rooms.

Implications:

Due to the exposure to second-hand smoke (SHS) 890,000 people die each year worldwide. Particulate matter (PM) in SHS endangers the health of both non-smokers and smokers. This study considers the effect of additives like aromatics and humectant agents in cigarettes on PM in SHS. Do additives increase the amount of PM?

Keywords:

Second-hand smoke, Environmental tobacco smoke; particulate matter; additives; aromatics; humectant agents

Strength and limitations of this study:

- The used automatic environmental tobacco smoke emitter (AETSE) with a standardised smoking protocol enables the generating of reproducible and reliable particulate matter (PM) levels.
- The application of the applied methodology allows the comparison of different types of tobacco products.
- The used aerosol spectrometer has a measuring range from 0.25 to 30 µm.
- The mechanism simulates reliable and comparable conditions by avoiding subjective deviations and without exposing test persons or the investigator.
- The developed technique applied in this study is not able to imitate the human smoking behavior accurate in every detail.

Introduction:

Airborne particulate matter (PM) as a part of air pollution causes tremendous adverse health effects especially cardiovascular and respiratory diseases (1) and aggravates airway inflammation and hyperresponsiveness in asthmatic patients (2). The exposure to PM is also associated with increased risk of ischemic stroke (3) and breast cancer mortality (4). Several studies have shown that morbidity and mortality increase in relation to PM exposure (5). PM is a differently sized mixture of solid and liquid particles that varies in composition and origin (6). One option to classify PM is by the size of the particles, that determines how deep they penetrate the respiratory system. The smaller the particles the deeper they penetrate (7, 8). Indeed, the size-related definition of PM is the most relevant one. The U.S. Environmental Protection Agency (EPA) differentiates between $PM_{10,}$ inhalable coarse particles equal or smaller than 10 µm, and $PM_{2.5,}$ fine inhalable particles equal or smaller than 2.5 µm (9). In addition, PM_1 is the fraction of particles equal or smaller than 1 µm. The majority of PM derives from tobacco smoke (10). Worldwide approximately one billion adults smoke (11). Each year more than 7 million people are killed due to tobacco use, and 890,000 of those are non-smokers being exposed to second-hand smoke (SHS) , also called environmental tobacco smoke (12). SHS is a mixture of mainly side-stream smoke emitted from the smoldering tobacco product and the exhaled mainstream smoke from the smoker (13, 14) and is considered to be the major risk factor for air pollution in indoor spaces (15).

Previous studies showed variations of PM levels within different brands and types of cigarettes (16-18). Different additives, e.g. aromatics, humectant agents and the content of tar and nicotine might affect the amount of PM (19). Based on these conclusions it seems to be reasonable and necessary to compare different cigarette types of a special brand with and without additives. Therefore, the particle size fractions of PM_{10}, $PM_{2.5}$ and PM_1 of four specific types of the popular cigarette brand Lucky Strike (20) as well as the ones of the reference cigarette 3R4F developed by the Kentucky Tobacco Research and Development

Center (University of Kentucky, USA) (21) were analysed. At the time of the study, Lucky Strike offered each two cigarette types with nearly congruent amounts of tar, nicotine and carbon monoxide with and without additives like aromatics and humectant agents. The four cigarette types of Lucky Strike to be examined were Original Red and Original Blue (with additives) (22, 23), and Straight Red and Straight Blue (without additives) (24, 25). They differ among others in their content of tar, nicotine, carbon monoxide and additives as shown in table 1. For more extensive information, it is referred to the Federal Ministry of Food and Agriculture of Germany (*Bundesministerium für Ernährung und Landwirtschaft*) (22-25).

The aim of this study was to investigate only the influence of additives on PM emissions. To minimise other influences on the amount of PM, like e.g. dissimilar manufacture processes of different tobacco companies, test cigarettes of one cigarette brand were selected.

Methods:

Nineteen cigarettes of four tobacco products from Lucky Strike, each two with and without additives, and 19 reference cigarettes were smoked using an automatic environmental tobacco smoke emitter (AETSE). The various PM-levels (PM_{10}, $PM_{2.5}$, PM_1) were recorded and evaluated. A modified smoking protocol according to the Tobacco Smoke Particles and Indoor Air Quality (ToPIQ) studies (16, 26) was used.

In a glass chamber with a volume of 2.88 m^3 the AETSE was located. This smoke pump for medical research, designed and engineered by Schimpf-Ing, Trondheim, Norway (27), allows the smoking of tobacco products in a reproducible way without exposing test persons and the investigator. A stepper motor moved via a linear actuator a 200 ml glass syringe for imitating the smoking process. The puff volume (40 ml), puff flow rate (13 ml/sec), puff frequency (2/min), inter puff interval (22 sec) and the amount of 9 puffs were adjusted by a microcontroller. The smoking protocol was departed in four different phases of five minutes each and started with the pre-ignition phase and measurement of the baseline values. After the cigarette was lighted and smoked in the combustion phase, the post-combustion phase followed, which started with the extinguishing of the cigarette. In the final suction phase, the indoor air was cleaned by using an industrial suction device, before the next cycle started. Each cycle lasted 20 min. The PM concentrations in the chamber were measured in a dilution of 1:10 with compressed air by a Grimm Portable Laser Aerosol Spectrometer and Dust Monitor model 1.109 (28, 29). The dilution with air was necessary to protect the spectrometer against blockage of the laser measuring chamber by high particle concentrations. Subsequently the dilution ratio was considered. The spectrometer detected via light scattering airborne particles with a size range from 0.25 µm to 32 µm. It displayed the output of measurement data as particle count [l^{-1}], detailed dust mass fractions in 31 channels [$\mu g/m^3$], output of the measurement results important for occupational health according to European standard EN 481, inhalable, thoracic and alveolic [$\mu g/m^3$] (30) and the PM_{10}, $PM_{2.5}$ and PM_1

values according to EPA (9) in real-time. The received data was recorded every six seconds. The PM values were analysed using the area under the curve (AUC) and the mean concentration (C_{mean}) referring to the mean value of 19 cigarettes of each type. In this study, the AUC describes the area under a concentration-time curve in the five minutes lasting interval from ignition to extinction of a cigarette. A test for Gaussian normality was performed. All cigarette type samples passed the test. Afterwards the Kruskal-Wallis test followed by the Dunn's multiple comparison test (post-hoc test) were done to compare the investigated cigarette types among themselves.

Patient involvement: Patients were not involved.

Results:

All results of the AUC-PM values and the C_{mean} values allow a direct comparison of the PM emission differences among the cigarette brands, because all tobacco products were tested exactly the same way with identical suction volume, speed and puff amount. The results of the AUC-PM values are shown in figure 1. The results of the C_{mean} values are shown in table 2. Figure 2 shows that the main part of SHS was composed by PM_1 fraction with 97.7 % (reference cigarette 3R4F), 93.9 % (Original Red), 93.1 % (Original Blue), 93.6 % (Straight Red) and 93.3 % (Straight Blue), respectively.

The data of the AUC-PM of all types of the Lucky Strike cigarettes were 21-71 % higher than the data of the reference cigarette 3R4F. The values of C_{mean}-PM of all types of Lucky Strike cigarettes were 22-71 % higher than the values of C_{mean}-PM of the reference cigarette 3R4F as well. Furthermore, the AUC and the C_{mean} of PM_1, $PM_{2.5}$ and PM_{10} of all Lucky Strike brands except Straight Blue showed a high statistical significance compared to the AUC-PM and C_{mean} of the reference cigarette 3R4F.

More specifically, the Straight Blue, a tobacco product without additives and lower tar amount (table 1), shows 10 to 25 % lower PM mean values compared to the other tested Lucky Strike products. The differences between Straight Blue and Original Blue are highly significant ($p<0.05$). The differences between Original Red and Original Blue are significant, too ($p<0.05$). The Straight Blue showed also less PM values than the Straight Red but no statistical significance ($p>0.05$). Details are shown in table 3. In comparison of the Straight Blue to the reference cigarette, the AUC-PM_1 mean values were only 21 %, AUC-$PM_{2.5}$ and AUC-PM_{10} 27 % higher. Accordingly, the C_{mean} values of PM_1, $PM_{2.5}$ and PM_{10} of the Straight Blue were 22 %, 27 % and 28 % higher than the values of the reference cigarette.

In contrast, the PM mean values as well as the C_{mean} values of the Lucky Strike Original Blue, a cigarette with additives, but the same tar amount as the Straight Blue, were substantial higher with 63 %, 70 % and 71 % higher values for C_{mean} and AUC of PM_1, $PM_{2.5}$ and PM_{10} compared to the reference cigarette. The measuring data of the Lucky Strike Straight Red and Original Red were in between with 44 % higher C_{mean} values of PM_1 and 50 % higher C_{mean} values of $PM_{2.5}$ and PM_{10} of the Straight Red, and 37 % higher C_{mean} values of PM_1 and 42 % higher C_{mean} values of $PM_{2.5}$ and PM_{10} of the Original Red compared to the reference cigarette. According to this, the AUC mean values of the Straight Red were 43% (PM_1) and 50% ($PM_{2.5}$ and PM_{10}), and of the Original Red 36% (PM_1), 41% ($PM_{2.5}$) and 42% (PM_{10}) higher.

The fact, that the additive-free Straight Blue with a lower tar amount showed the lowest measurement values of all four Lucky Strike cigarette products, suggests that additives in cigarettes in combination with a higher tar amount increase the PM emissions.

The PM mean of all measured baseline values, that means clean air, was 1.6 µg/m³. The measured C_{mean} of PM_{10} increased up to 1803 µg/m³ (± 320 µg/m³) for the reference cigarette and 3076 µg/m³ (± 321 µg/m³) for the Lucky Strike Original Blue and in case of $PM_{2.5}$ up to 1801 µg/m³ (± 320 µg/m³) respectively 3068 µg/m³ (± 319 µg/m³). For PM_1 the values raised up to 1762 µg/m³ (± 302 µg/m³) respectively 2865 µg/m³ (± 282 µg/m³).

Discussion:

Different studies show a hazardous increase of PM levels in smoking rooms and households (31) (32) (33) (34). The results of our study show that tobacco smoke in an enclosed space of 2.88 m^3 (capacity of the measuring cabin) increased the particulate matter emissions compared to smoke free air (baseline values) more than a 1000-fold. The measured $PM_{2.5}$ values exceeded the daily average concentration of at most 25 µg/m^3 according to the *WHO Air quality guidelines* (35) approximately 70- to 120-fold, depending on the cigarette brand. This is to illustrate how massive the PM burdens under the study conditions were. For example, a compact car, classified by the EPA with a total passenger and cargo volume of 2.832 m^3 to 3.087 m^3 (36), has a comparable indoor volume. The used modified smoking regime is similar to conditions in a compact car with closed windows and no ventilation or air conditioning. This is a fundamentally important aspect of our study design, because many people smoke in cars. The passive smoke with the contained particulate matter is not only hazardous to the health of smokers but also of passengers, which are often children.

The aim of this study was to investigate only the influence of additives on PM emissions. To minimise influences on PM emissions by e.g. production processes of different cigarette manufacturers it seemed to be suggestive to choose cigarette types of one manufacturer. Of all tested Lucky strike brands the cigarette type without additives in combination with lower tar amount (Straight Blue) emitted less particulate matter than the type with no additives but higher tar amount (Straight Red), respectively the cigarette with additives but lower tar amount (Original Blue). The results lead to the assumption that cigarettes without additives emit less particulate matter. In contrast, the Straight Red and the Original Red had similar PM values. Nevertheless, it must be emphasised, that all tested cigarettes of the brand Lucky Strike emitted significant higher PM levels than the reference cigarette. Hence, it could not be ascertained beyond doubt that additive free cigarettes produced by one manufacturer emit less

PM than cigarettes with additives. Therefore, further studies are necessary to prove the assumption that cigarettes without additives and lower tar amount emit less PM.

Only a few studies are published with respect to an effect of additives on PM with contradictory conclusions. In 2002, Rustemeier et al. (19) performed a study, in which 333 commonly used ingredients were added in three different groups to the 1R4F reference cigarette. The results showed an increase of 13-28 % of PM relative to the cigarettes without added additives. In 2011, Wertz et al. (37) analysed previously secret tobacco industry documents and found among others four peer reviewed publications which concluded no evidence of substantial toxicity as well as total particulate matter (TPM). They ascertained that internal documents of the tobacco industry showed post hoc changes in protocols after primary statistical findings of an additive-associated increase in toxicity and TPM concentrations in cigarette smoke with additives. No significant difference of PM of cigarettes with and without the additive menthol showed the studies of Gaworski et al. (38) and Gerharz et al. (39). Similar results were described by Wasel et al. (17). They found no significant differences in the PM amount of L&M cigarettes with and without additives. But higher PM values of the L&M filtered cigarillo could be shown.

In this study, by far the largest part of PM is represented by particles ≤ 1 µm and > 0.25 µm. Figure 2 shows the particle distribution pattern of all investigated cigarettes. Depending on the cigarette brand over 93 % to nearly 98 % of the measured PM was $PM_{1.}$ It seems that the smaller the particles, the more extensive are the health effects (7) (40). Already in 1960, Keith and Derrick (41) published similar results that most of the particles in tobacco smoke has a size between 0.1 µm and 1 µm peaked between 0.2 µm and 0.25 µm. Nazaroff and Klepeis (42) described SHS with a particle size between 0.02 µm and 2 µm in diameter. There is no common agreement on the peak size. On the one hand side-stream smoke particles were described with geometric mean diameters of 0.1µm (43, 44). As opposed to that, Haustein and

Groneberg (45) reported mean diameters of 0.5 µm. The technical limitation of the used aerosol spectrometer Grimm model 1.109 to detect particles with a minimum size of 0.25 µm resulted in a nonconformity with the EPA definition, where particles down to 0.1 µm are included. Here it must be mentioned that the used laser aerosol sprectrometer, built for continuous measurement of PM and also common used in monitoring networks (46), has a measuring range from 0.25 µm to 30 µm. This limitation effected that a proportion of PM_1 could not be measured and for particles smaller than 0.25 µm a new measurement system is essential. The used aerosol spectrometer measures PM, including PM_1 and semi-volatile fractions (e.g. water, ammonium nitrate, some organic compounds) via light scattering in real time (47). This allows to investigate the PM amount of each single tobacco product. By contrast, the EPA Federal Reference Methods (FRMs) for PM measuring often rest upon 24 h sample collection of PM_{10} and $PM_{2.5}$, but not PM_1, followed by gravimetric measurement of the collected PM, or the likewise real time measurement device Tapered Element Oscillating Microbalance (TEOM) Monitor (47, 48). The European standard EN 12341 for determination of the PM_{10} and $PM_{2.5}$ is also a gravimetric measurement method (49). It must be said, that one listed FRM is an automated equivalent method with the Grimm model EDM 180, which measures PM via light scattering (48). Several studies showed that the measurement results of a Grimm model 1.107, 1.108 or 1.109 are very similar to the results of TEOM Monitors, Grimm model EDM 180 or gravimetric methods (47, 50). Fromme et al. concluded in 2007, that gravimetric methods generates higher results than laser aerosol spectrometer but with a high correlation of the rank order of measured values (51). Thus, the measured values of the used Grimm model 1.109 are valid, but it is very important not to change the method of measurement during a study.

In addition, the AETSE is not able to imitate SHS and the smoking behaviour of humans exactly, because in the respiratory tract the inhaled mainstream smoke is humidified and due to hygroscopic growth the exhaled smoke particles are approximately 1.5-fold larger (52, 53).

By using the AETSE, it is not possible to differentiate between inhaled and exhaled mainstream smoke, but it should be enhanced that SHS consists only of approx. 15 % mainstream smoke and approx. 85 % side-stream smoke (54, 55). However, the AETSE is able to imitate side-stream smoke as realistically as possible. Hence, the measured PM emissions are very similar to SHS. It is worth pointing out that reproducible results have been ensured by avoiding inter-individual deviations without the exposure of a test person to any health risk. The used modified smoking regime differed from the already existing protocols like, e.g. ISO/TR 17219 (56) or the *Standard operating procedure for intense smoking of cigarettes* by the WHO (57). On that point, it must be said that no "gold standard" exists for smoking regimes (58-61). Moreover, this study as well as all previous ToPIQ-studies focuses on data comparison to the 3R4F reference cigarette and not on absolute numbers. Therefore, the use of the modified protocol and the application of AETSE can be considered as valid.

In conclusion and in general, this study showed repeatedly the massive increase of particulate matter in enclosed rooms in consequence of smoking of tobacco products. The possibility of the decrease of PM emissions due to the absence of additives in tobacco products should be verified in further studies.

Author Contributions: This article is part of the thesis of Elsa-Luise Fromm, whereas Markus Braun, Alexander Gerber, Doris Klingelhöfer, Ruth Müller and David A. Groneberg contributed significantly to the conception and design of the study. Moreover, they prepared the experiments, which were performed by Elsa-Luise Fromm. Elsa-Luise Fromm and Alexander Gerber analysed the data. Ruth Müller reanalysed the data. The manuscript was written by Markus Braun and critically reviewed by all authors. All authors have participated sufficiently in the work to take public responsibility for appropriate portions of the content and agreed to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved. All authors have read and approved the final manuscript.

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Ethics approval: Not required

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Figure legends:

Figure 1: Comparison of AUC (PM_{10}, $PM_{2.5}$ and PM_1) for all tested tobacco products.

Figure 2: Distribution pattern of $PM_{10\text{-}2.5}$, $PM_{2.5–1}$ and PM_1 of all investigated cigarettes.

Table 1: List of the cigarette ingredients with reference to tar, nicotine, carbon monoxide and additives (21-25). H.A. = Humectant Agent, A. = Aromatic

	3R4F Reference cigarette	**Lucky Strike Original Red**	**Lucky Strike Original Blue**	**Lucky Strike Straight Red**	**Lucky Strike Straight Blue**
Tar [mg]	9.4	10	7	10	7
Nicotine [mg]	0.73	0.8	0.6	0.9	0.6
Carbon monoxide [mg]	12	10	8	10	8
Glycerin (H.A.)	yes	yes	yes	no	no
Sugar (A.)	yes	yes	yes	no	no
Cocoa Powder (A.)	n/s	yes	yes	no	no
Licorice Extract (A.)	n/s	yes	yes	no	no
Flavours below 0.1% w/w (A.)	n/s	yes	yes	no	no

Table 2: Mean concentrations (C_{mean} PM_{10}, $PM_{2.5}$ and PM_1) with standard deviation of all tested tobacco products.

	3R4F Reference cigarette	**Lucky Strike Original Red**	**Lucky Strike Original Blue**	**Lucky Strike Straight Red**	**Lucky Strike Straight Blue**
Cmean PM10 [µg/m³]	1803 ± 320	2557 ± 726	3076 ± 321	2704 ± 261	2300 ± 340
Cmean PM2.5 [µg/m³]	1801 ± 320	2550 ± 724	3068 ± 319	2700 ± 261	2294 ± 339
Cmean PM1 [µg/m³]	1762 ± 302	2402 ± 624	2865 ± 282	2530 ± 231	2145 ± 306

Table 3: P-values of statistical Dunn's multiple comparisons test of AUC (PM_{10}, $PM_{2.5}$ and PM_1) for the Lucky Strike brands (significant results are highlighted by bold font type).

	Original Red vs. Original Blue	**Original Red vs. Straight Red**	**Original Red vs. Straight Blue**	**Original Blue vs. Straight Red**	**Original Blue vs. Straight Blue**	**Straight Red vs. Straight Blue**
C_{mean} PM10	**0.0424**	>0.9999	>0.9999	0.6131	**0.0005**	0.2817
C_{mean} PM2.5	**0.0424**	>0.9999	>0.9999	0.6131	**0.0005**	0.2775
C_{mean} PM1	**0.0465**	>0.9999	>0.9999	0.6131	**0.0002**	0.1829

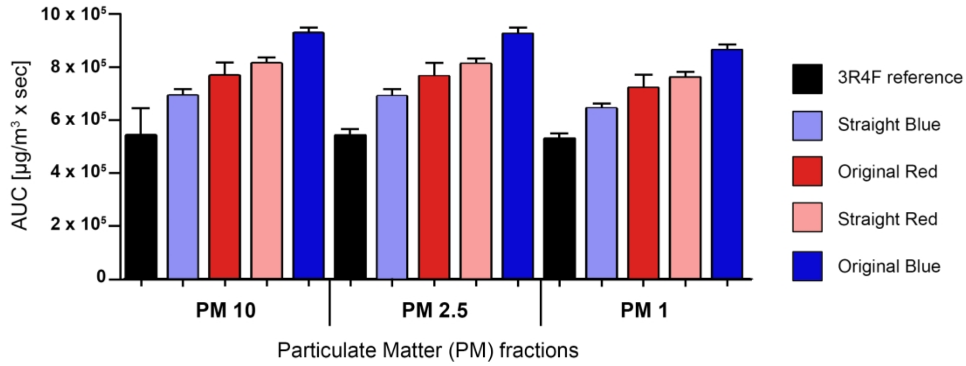

Figure 1: Comparison of AUC (PM10, PM2.5 and PM1) for all tested tobacco products.

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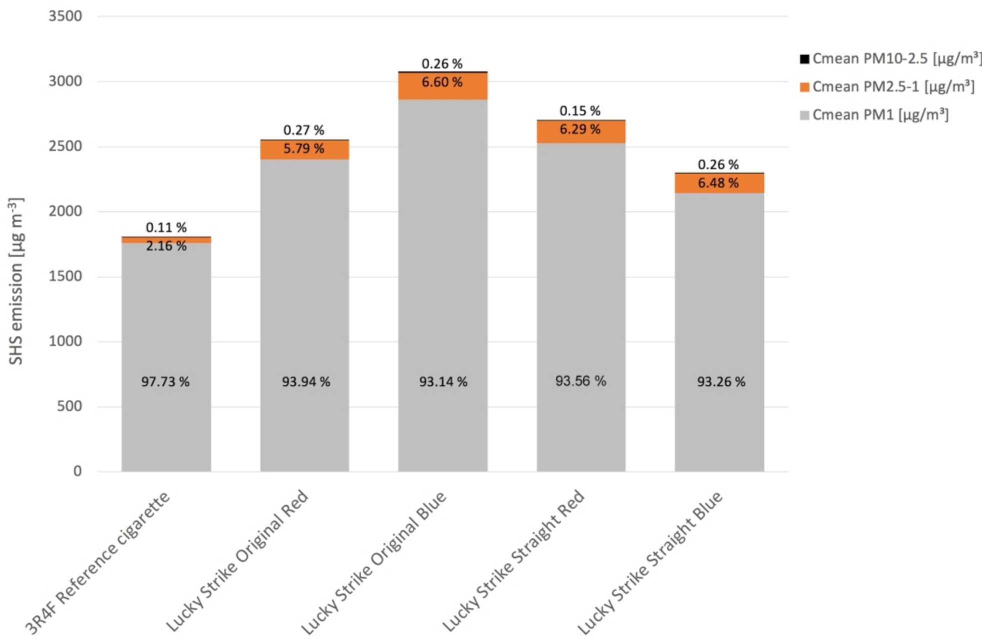

Figure 2: Distribution pattern of PM10-2.5, PM2.5–1 and PM1 of all investigated cigarettes.

199x129mm (300 x 300 DPI)

BMJ Open

Particulate matter emissions of four types of one cigarette brand with and without additives: a laser spectrometric particulate matter analysis of second-hand smoke

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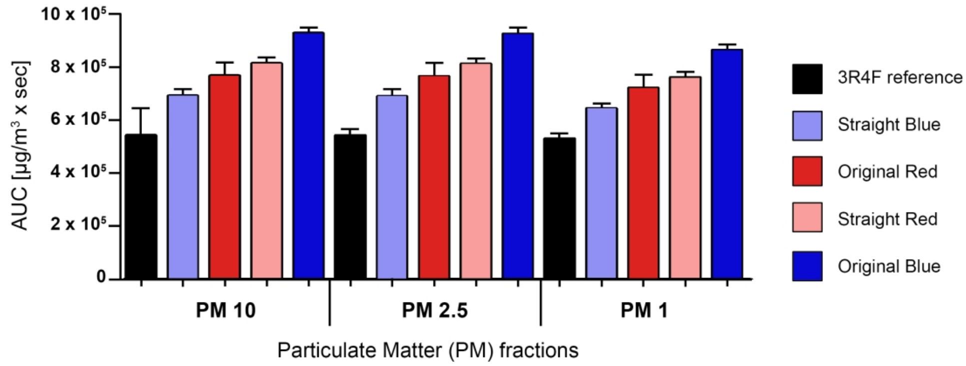
Note: The following files were submitted by the author for peer review, but cannot be converted to PDF. You must view these files (e.g. movies) online.	
BMJ_Open_PM_Lucky_Strike_R2_v2.docx	



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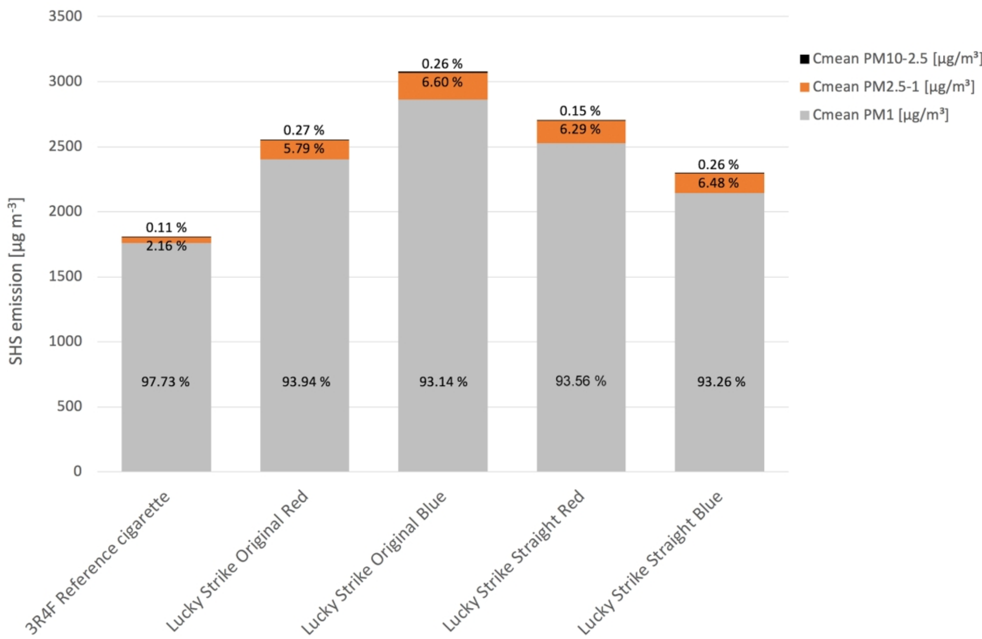

Figure 2: Distribution pattern of PM10-2.5, PM2.5–1 and PM1 of all investigated cigarettes.

199x129mm (300 x 300 DPI)