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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 secondhand smoke

Markus Braun, Elsa-Luise Fromm, Alexander Gerber, Doris Klingelhöfer, Ruth Müller, David A Groneberg

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Institute of Occupational, Social and Environmental Medicine, Goethe University Frankfurt, Frankfurt am Main, Germany

Correspondence to
Mr Markus Braun;
m.braun@med.uni-frankfurt.de

ABSTRACT

Objective Inhaled particulate matter (PM) in secondhand smoke (SHS) is deleterious for smokers and non-smokers. Different additives in cigarettes might effect the amount of PM. This study aimed to assess the influence of additives on the PM emissions from different cigarette types in SHS.

Design An experimental study of PM measuring in SHS of cigarettes without exposition of any person.

Method The concentrations of PM (PM_{10}, $PM_{2.5}$ and PM_1) in SHS 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), in comparison to the reference cigarette 3R4F were analysed. An automatic environmental tobacco smoke emitter generated SHS in an enclosed space with a volume of 2.88 m^3. PM was measured with a laser aerosol spectrometer (Grimm model 1.109). Afterwards, the measuring values of the four Lucky Strike brands and the reference cigarette were statistically evaluated and visualised.

Results Lucky Strike Straight Blue, a cigarette type without additives and lower tar amount, showed 10% to 25% lower PM mean values compared with 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 (clean air) was 1.6 µg/m³. It increased up to about 1800 µg/m³ for the reference cigarette and up to about 3070 µg/m³ for the Lucky Strike Original Blue.

Conclusions The findings of this study show the massive increase of PM amount by smoking cigarettes in enclosed spaces and suggest that additives in tobacco products increase the PM amount in SHS. For validation, further comparative studies are necessary focusing on the comparison of the PM concentration of cigarettes with and without additives.

Implications Due to the exposure to SHS, 890 000 people die each year worldwide. 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 in tobacco products increase the amount of PM?

Strength and limitations of this study

- Different types of tobacco products with and without additives were checked against each other regarding airborne particulate matter (PM) directly.
- The automatic environmental tobacco smoke emitter generated reproducible and reliable PM levels in accordance with a standardised smoking protocol.
- The used laser aerosol spectrometer measures the emitted particles in a size range of 0.25 µm to 32 µm in real time.
- The mechanism simulated reliable and comparable conditions without exposing test persons or the investigator.
- The applied technique was not able to imitate accurately the human smoking behaviour 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 hyper-responsiveness in asthmatic patients[2] should be mentioned in this context. The exposure to PM is also associated with increased risk of ischaemic stroke[3] and breast cancer mortality.[4] Several studies showed increase in morbidity and mortality in relation to higher PM exposure.[5]

PM is a mixture of solid and liquid particles varying in size, composition and origin.[6] The most relevant and common option to classify PM is by size of the particles that determines how deep they penetrate the respiratory system. The US 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.[7] In addition, PM_1 is the fraction of particles equal or smaller than 1 µm. The smaller the particles the deeper they penetrate in the respiratory system and the more extensive are the health effects.[8–10]

The majority of PM derives from tobacco smoke.[11] Worldwide approximately one billion adults smoke.[12] 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 (SHS), also called environmental tobacco smoke.[13] SHS mainly consists of sidestream smoke emitted directly from the smouldering tobacco product and the exhaled mainstream smoke from the smoker.[14 15] It is considered to be the major risk factor for air pollution in indoor spaces.[16]

Previous analyses revealed variations of PM levels within different brands and types of cigarettes.[17–19] The content of tar, nicotine and various additives (eg, aromatics and humectant agents) might affect the amount of PM.[20]

Based on these findings, it is reasonable and necessary to compare different cigarette types of a special brand with and without additives. Therefore, the aim of this study was to investigate the influence of additives on PM emissions of cigarettes. Hence, the particle size fractions of PM_{10}, $PM_{2.5}$ and PM_1 of four different types of the popular cigarette brand Lucky Strike[21] and of the reference cigarette 3R4F, developed by the Kentucky Tobacco Research and Development Center (University of Kentucky, USA),[22] were analysed. At the time of the study, Lucky Strike offered each two cigarette types with and without additives with nearly congruent amounts of tar, nicotine and carbon monoxide. The Lucky Strike cigarette types were Original Red and Original Blue (with additives)[23 24] and Straight Red and Straight Blue (without additives)[25 26] were included in the analysis. They differed among others in content of tar, nicotine, carbon monoxide and additives as shown in table 1. For more detailed information, refer to the Federal Ministry of Food and Agriculture of Germany (*Bundesministerium für Ernährung und Landwirtschaft*).[23–26]

To minimise other influences on the amount of PM, for example dissimilar manufacture processes of different tobacco companies, test cigarettes of one cigarette brand were selected.

Table 1 List of the cigarette ingredients with reference to tar, nicotine, carbon monoxide and additives[22–26]

	Reference Cigarette 3R4F	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
Glycerine (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

A, aromatic; HA, humectant agent.
n/s, not significant.

METHODS

Nineteen cigarettes of four tobacco products from Lucky Strike, two each with and without additives, and 19 reference cigarettes were smoked using an automatic environmental tobacco smoke emitter (AETSE). The individual 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[17 27] was applied.

The AETSE is located in a closable chamber with a volume of 2.88 m^3. This smoke pump for medical research, designed and engineered by Schimpf-Ing, Trondheim, Norway,[28] allows the smoking of tobacco products in a reproducible way without exposing test persons and the investigator. A stepper motor moves via a linear actuator a 200 mL glass syringe that imitates the process of smoking. Puff volume (40 ml), puff flow rate (13 ml/s), puff frequency (2/min), inter-puff interval (22 s) and amount of nine puffs is adjusted by a microcontroller. The smoking protocol is divided into four different phases of each 5 min. It starts with the pre-ignition phase and measurement of the baseline values. Then, the cigarette is lighted and smoked in the combustion phase. The post-combustion phase follows, which starts with the extinguishing of the cigarette. In the last cycle phase, the indoor air is cleaned by using an industrial suction device before the next cycle starts. Each cycle lasts 20 min. The PM concentrations in the chamber is measured in a dilution of 1:10 with compressed air by a Grimm Portable Laser Aerosol Spectrometer and Dust Monitor model 1.109.[29 30] The dilution with compressed air is necessary to protect the spectrometer against blockage of the laser measuring chamber by high particle concentrations. When evaluating the measurement results, the dilution ratio of 1:10 has to be taken into account. The spectrometer detects in real time via light scattering airborne particles with a size range from 0.25 to 32 µm. It displays the output of measurement data as particle count [l^{-1}] and detailed dust mass fractions in 31 channels (µg/m^3). Furthermore, it is possible to present the data output as inhalable, thoracic and alveolic (µg/m^3) according to European standard EN 481[31] or as PM_{10}, $PM_{2.5}$ and PM_1 values according to the US EPA.[7] The received data is recorded every 6 s. Subsequently, the PM values are statistically evaluated by calculating the area under the curve

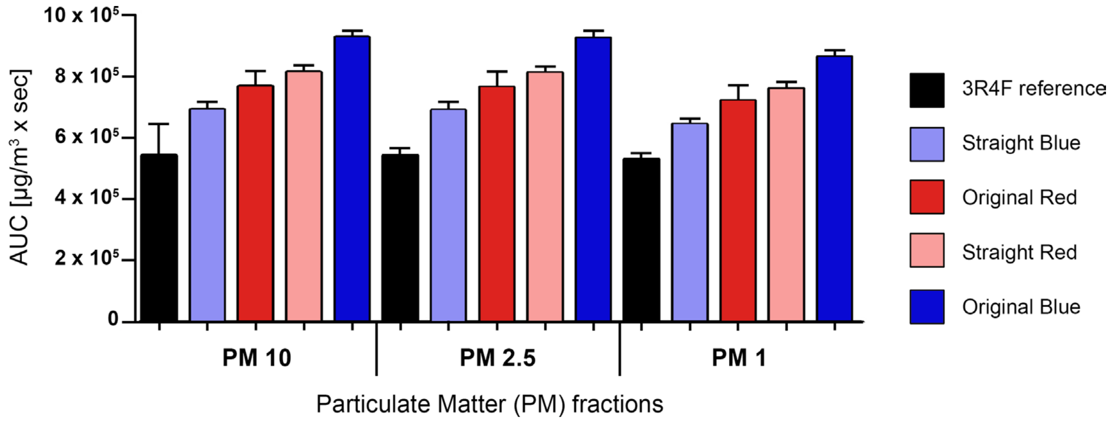

Figure 1 Comparison of area under the curve (AUC) (PM_{10}, $PM_{2.5}$ and PM_1) for all tested tobacco products.

(AUC) and the mean concentration (C_{mean}). In this study, the AUC describes the area under a concentration-time curve in the 5 min lasting interval from ignition to extinction of a cigarette. Since all measured data of the cigarette samples passed the test for Gaussian normality, the Kruskal-Wallis test followed by the Dunn's multiple comparison test (post hoc test) were done to compare the individual values of the investigated cigarette types with each other.

Patient involvement

Patients were not involved.

RESULTS

The data of AUC-PM of all types of Lucky Strike cigarettes were 21%–71% higher than the data of the reference cigarette (figure 1). 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 as well (table 2). Furthermore, the differences of AUC and 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 with the AUC-PM and C_{mean} of the reference cigarette.

The main part of SHS consists of the PM_1 fraction with 97.7% (reference cigarette), 93.9% (Lucky Strike Original Red), 93.1% (Lucky Strike Original Blue), 93.6% (Lucky Strike Straight Red) and 93.3% (Lucky Strike Straight Blue), respectively (figure 2).

More specifically, Lucky Strike Straight Blue, a tobacco product without additives and lower tar amount (table 1), showed 10%–25% lower PM mean values compared with the other tested Lucky Strike products. The differences between Lucky Strike Straight Blue and Lucky Strike Original Blue were highly significant ($p \leq 0.0005$), and between Lucky Strike Original Red and Lucky Strike Original Blue significant too ($p<0.05$). Lucky Strike Straight Blue showed also less PM values than Lucky Strike Straight Red but no statistical significance ($p>0.05$). Details are shown in table 3. In a comparison of Lucky Strike Straight Blue with the reference cigarette, the AUC-PM_1 mean values were only 21%, the AUC-$PM_{2.5}$ and AUC-PM_{10} mean values 27% higher, respectively. Accordingly, the C_{mean} values of PM_1, $PM_{2.5}$ and PM_{10} of Lucky Strike Straight Blue were 22%, 27% and 28% higher than the values of the reference cigarette.

In contrast, the AUC-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 Lucky Strike Straight Blue, were substantially higher (PM_1 63%, $PM_{2.5}$ 70%, PM_{10} 71%) compared with the reference cigarette. The measuring data of Lucky Strike Straight Red were in between 43% (PM_1) and 50% ($PM_{2.5}$ PM_{10}) higher compared with the reference cigarette. Furthermore, the measured values of Lucky Strike Original Red were in between 36% (PM_1) and 41% ($PM_{2.5}$) respectively 42% (PM_{10}) higher than the values of the reference cigarette.

The fact that the additive-free Lucky Strike 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 (smoke-free air) was 1.6 µg/m³. The measured C_{mean} of PM_{10}

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

	Reference Cigarette 3R4F	Lucky Strike Original Red	Lucky Strike Original Blue	Lucky Strike Straight Red	Lucky Strike Straight Blue
$C_{mean}PM_{10}$(µg/m³)	1803±320	2557±726	3076±321	2704±261	2300±340
$C_{mean}PM_{2.5}$(µg/m³)	1801±320	2550±724	3068±319	2700±261	2294±339
$C_{mean}PM_1$(µg/m³)	1762±302	2402±624	2865±282	2530±231	2145±306

PM, particulate matter.

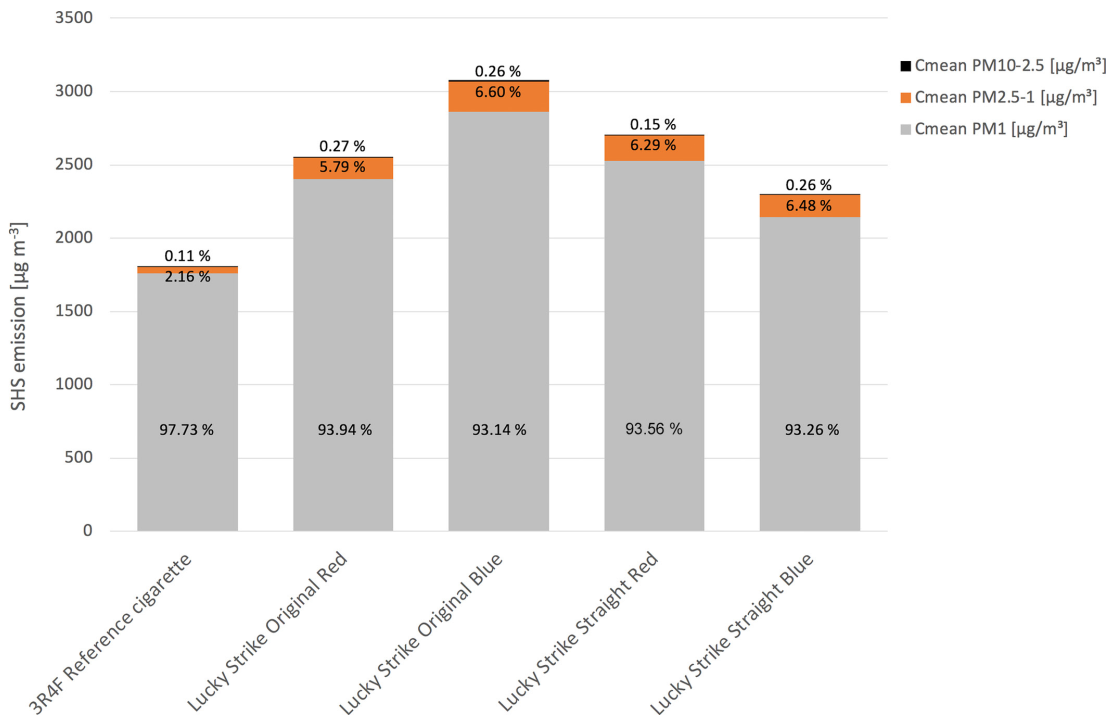

Figure 2 Distribution pattern of $PM_{10-2.5}$, $PM_{2.5-1}$ and PM_1 of all investigated cigarettes. PM, particulate matter; SHS, secondhand smoke.

increased up to 1803 µg/m³ (±320 µg/m³) for the reference cigarette and 3076 µg/m³ (±321 µg/m³) for Lucky Strike Original Blue. In case of $PM_{2.5}$ it went up to 1801 µg/m³ (±320 µg/m³) respectively 3068 µg/m³ (±319 µg/m³). The values for PM_1 increased up to 1762 µg/m³ (±302 µg/m³) respectively 2865 µg/m³ (±282 µg/m³).

DISCUSSION

The findings of the presented study show that tobacco smoke in an enclosed space of 2.88 m³ (capacity of the measuring cabin) increased the particulate matter emissions compared with smoke-free air (baseline values) more than a 1000-fold. The measured $PM_{2.5}$ values exceeded the daily average concentration of the maximum of 25 µg/m³ according to the WHO air quality guidelines[32] approximately 70-fold to 120-fold, depending on the cigarette brand. This illustrates the massiveness of PM burdens under the study conditions.

A compact car, classified by the EPA with a total passenger and cargo volume of 2.832 m³ to 3.087 m³,[33] has a indoor volume that is comparable to the measuring cabin. The modified smoking regime that was used is similar to conditions in a compact car with closed windows and no ventilation or air conditioning. This is a fundamentally important aspect of the 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 to passengers who are often children.

Different studies show a hazardous increase of PM levels in smoking rooms and households,[34–37] but only a few studies were published with respect to an effect of additives on PM with contradictory conclusions. In 2002, Rustemeier *et al*[20] performed a study, in which 333 commonly used ingredients were added to the reference cigarette 1R4F. The results showed an increase of 13%–28% of PM relative to the cigarettes without added additives. In 2011, Wertz *et al*[38] analysed formerly secret documents of the tobacco industry. They found among others four peer-reviewed publications that concluded no correlation between additives and toxicity as well as total particulate matter (TPM). Regarding this, internal documents of the

Table 3 P values of the statistical Dunn's multiple comparisons test of AUC (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
AUC PM_{10}	**0.0424**	>0.9999	>0.9999	0.6131	**0.0005**	0.2817
AUC $PM_{2.5}$	**0.0424**	>0.9999	>0.9999	0.6131	**0.0005**	0.2775
AUC PM_1	**0.0465**	>0.9999	>0.9999	0.6131	**0.0002**	0.1829

Significant results are highlighted by bold font type.
AUC, area under the curve; PM, particulate matter.

tobacco industry showed post hoc changes in protocols after previous statistical findings of an additive-associated increase in toxicity and TPM concentrations. Wasel *et al*[18] found no significant differences in the PM amount of L&M cigarettes with and without additives. Similar findings showed the studies of Gaworski *et al*[39] and Gerharz *et al*.[40] They could not prove the influence of the additive menthol.

Therefore, the main focus of this study was to investigate the impact of cigarette additives on PM emissions. It seemed to be advisable to choose cigarette types of one manufacturer to minimise influences on PM emissions by, for example, production processes. Of the four tested Lucky Strike types, the cigarettes without additives in combination with lower tar amount (Lucky Strike Straight Blue) emitted less PM than those without additives but with higher tar amount (Lucky Strike Straight Red), respectively those with additives but lower tar amount (Lucky Strike Original Blue). That would justify the claim that additives have an impact on the concentration of PM in SHS, though the smoke of Lucky Strike Straight Red (without additives) and Lucky Strike Original Red (with additives) contained similar PM amounts. However, all tested cigarettes of the brand Lucky Strike emitted significant higher PM levels than the reference cigarette. In conclusion, the findings could not ascertain beyond doubt that additive-free cigarettes produced by one manufacturer emit less PM than cigarettes with additives. Therefore, further studies are necessary to prove this assumption.

In this study, by far the largest part of PM is represented by particles ≤1 µm and ≥0.25 µm. Depending on the cigarette brand, over 93% to nearly 98% of the measured PM was PM_1 (figure 2). Already in 1960, Keith and Derrick[41] showed that most of the particles in tobacco smoke has a size between 0.1 µm and 1 µm and 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 0.1 µm.[43 44] As opposed to that, Haustein and Groneberg[45] reported mean diameters of 0.5 µm. In this respect, it has to be mentioned that the used aerosol spectrometer Grimm model 1.109 only detects particles with a minimum size of 0.25 µm. Thus, the part of PM_1 smaller than 0.25 µm could not be measured. This led to a deviation of the PM_1 content in accordance with the EPA definition that includes particles down to 0.1 µm.

It must be mentioned that the used laser aerosol spectrometer, built for continuous measurement of PM, is also commonly used in monitoring networks.[46] An advantage of the spectrometer is the possibility to measure PM, including PM_1 and semi-volatile fractions (eg, water, ammonium nitrate, some organic compounds) via light scattering in real time.[47] This allowed to investigate the amount of PM of each single tobacco product. By contrast, the EPA Federal Reference Methods (FRMs) for measuring PM often rest on 24 hours sample collection of PM_{10} and $PM_{2.5}$, but not of 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 the determination of PM_{10} and $PM_{2.5}$ is also a gravimetric measurement method.[49] 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 were 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 generated 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 can be recognised as valid, but it is very important not to change the method of measurement during a study.

A limitation of the applied AETSE is the inability to imitate SHS and the smoking behaviour of humans exactly, because in the respiratory tract the inhaled mainstream smoke is humidified and the exhaled smoke particles are approximately 1.5-fold larger due to hygroscopic growth.[52 53] By using the AETSE, it is not possible to differentiate between inhaled and exhaled mainstream smoke, but it should be emphasised that SHS consists only of approximately 15% mainstream smoke and approximately 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 were very similar to SHS.

It is worth pointing out that reproducible results had 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, for example, ISO/TR 17219[56] or the standard operating procedure for intense smoking of cigarettes by the WHO.[57] At this 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 focused 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, this study showed repeatedly the massive increase of PM in enclosed rooms in consequence of smoking of tobacco products. Although the reduction of PM emissions due to the absence of additives in tobacco products should be verified in further studies.

Contributors Author Contributions: This article is part of the thesis of E-LF. MB, AG, DKö, RMü and DAG contributed significantly to the conception and design of the study. Moreover, MB, AG, DA.G and E-LF prepared the experiments, which were performed by E-LF. E-LF and AG analysed the data. RMü reanalysed the data. The manuscript was written by MB and critically reviewed by all authors. All authors 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 read and approved the final manuscript.

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REFERENCES

1. Brunekreef B, Holgate ST. Air pollution and health. *Lancet* 2002;360:1233–42.
2. Castaneda AR, Bein KJ, Smiley-Jewell S, *et al*. Fine particulate matter (PM2.5) enhances allergic sensitization in BALB/c mice. *J Toxicol Environ Health A* 2017;80:197–207.
3. Zhang C, Meng Q, Zhang X, *et al*. Role of astrocyte activation in fine particulate matter-enhancement of existing ischemic stroke in Sprague-Dawley male rats. *J Toxicol Environ Health A* 2016;79(9-10):393–401.
4. Tagliabue G, Borgini A, Tittarelli A, *et al*. Atmospheric fine particulate matter and breast cancer mortality: a population-based cohort study. *BMJ Open* 2016;6:e012580.
5. Anderson JO, Thundiyil JG, Stolbach A. Clearing the air: a review of the effects of particulate matter air pollution on human health. *J Med Toxicol* 2012;8:166–75.
6. Pope CA. Epidemiology of fine particulate air pollution and human health: biologic mechanisms and who's at risk? *Environ Health Perspect* 2000;108 Suppl 4(Suppl 4):713–23.
7. Epa. US, Agency EP, Matter P. PM) Pollution. 2017 https://www.epa.gov/pm-pollution/particulate-matter-pm-basics#PM.
8. Brown JS, Gordon T, Price O, *et al*. Thoracic and respirable particle definitions for human health risk assessment. *Part Fibre Toxicol* 2013;10:12.
9. Zwozdziak A, Sówka I, Willak-Janc E, *et al*. Influence of PM_1 and $PM_{2.5}$ on lung function parameters in healthy schoolchildren-a panel study. *Environ Sci Pollut Res Int* 2016;23:23892–901.
10. Kim KH, Kabir E, Kabir S. A review on the human health impact of airborne particulate matter. *Environ Int* 2015;74:136–43.
11. Van Deusen A, Hyland A, Travers MJ, *et al*. Secondhand smoke and particulate matter exposure in the home. *Nicotine Tob Res* 2009;11:635–41.
12. Society AC. The Tobacco Atlas. 5th edition, 2015. http://3pk43x313ggr4cy0lh3tctjh.wpengine.netdna-cdn.com/wp-content/uploads/2015/03/TA5_2015_WEB.pdf.
13. WHO. World Health Organization Tobacco Fact sheet. 2017 http://www.who.int/mediacentre/factsheets/fs339/en/.
14. Juranić B, Rakošec Ž, Jakab J, *et al*. Prevalence, habits and personal attitudes towards smoking among health care professionals. *J Occup Med Toxicol* 2017;12:20.
15. U.S U, Service PH. *The Health Consequences of Smoking—50*. Years of Progress: A Report of the Surgeon General - Executive Summary, 2014. https://www.surgeongeneral.gov/library/reports/50-years-of-progress/full-report.pdf.
16. DKFZ. *Passivrauchen - ein unterschätztes Gesundheitsrisiko*. Heidelberg, Germany: Deutsches Krebsforschungszentrum, 2006:1–72.
17. Gerber A, Hofen-Hohloch AV, Schulze J, *et al*. Tobacco smoke particles and indoor air quality (ToPIQ-II) - a modified study protocol and first results. *J Occup Med Toxicol* 2015;10:5.
18. Wasel J, Boll M, Schulze M, *et al*. Brand Cigarillos: Low Price but High Particulate Matter Levels-Is Their Favorable Taxation in the European Union Justified? *Int J Environ Res Public Health* 2015;12:9141–53.
19. Kant N, Müller R, Braun M, *et al*. Particulate Matter in Second-Hand Smoke Emitted from Different Cigarette Sizes and Types of the Brand Vogue Mainly Smoked by Women. *Int J Environ Res Public Health* 2016;13:799.
20. Rustemeier K, Stabbert R, Haussmann HJ, *et al*. Evaluation of the potential effects of ingredients added to cigarettes. Part 2: chemical composition of mainstream smoke. *Food Chem Toxicol* 2002;40:93–104.
21. Tobacco BA. Germany,Unsere Marken. 2015 http://www.bat.de/group/sites/BAT_7TYF37.nsf/vwPagesWebLive/DO7VFL7S?opendocument.
22. UK. University of Kentucky. Kentucky Tobacco Research and Development Center. 3R4F Preliminary Analysis. 2017 https://ctrp.uky.edu/resources/pdf/webdocs/3R4F%20Preliminary%20Analysis.pdf.
23. BMEL. Bundesministerium für Ernährung und Landwirtschaft. Tabakzusatzstoffe: Lucky Strike Original Red. 2011 https://service.bmel.de/tabakerzeugnisse/index2.php?detail_id=106700&site_key=153&stichw_suche=Lucky+Strike+Original+Red&zeilenzahl_zaehler=5.
24. BMEL. Bundesministerium für Ernährung und Landwirtschaft. Tabakzusatzstoffe: Lucky Strike Original Blue. 2011 https://service.ble.de/tabakerzeugnisse/index2.php?detail_id=105676&site_key=153&stichw_suche=Lucky+Strike+Original+Blue&zeilenzahl_zaehler=3.
25. BMEL. Bundesministerium für Ernährung und Landwirtschaft. Tabakzusatzstoffe: Lucky Strike Straight Blue. 2011 https://service.bmel.de/tabakerzeugnisse/index2.php?detail_id=105475&site_key=153&stichw_suche=Lucky+Strike+Straight+Blue&zeilenzahl_zaehler=1.
26. BMEL. Bundesministerium für Ernährung und Landwirtschaft. Tabakzusatzstoffe: Lucky Strike Straight Red. 2011 https://service.bmel.de/tabakerzeugnisse/index2.php?detail_id=105476&site_key=153&stichw_suche=Lucky+Strike+Straight+Red&zeilenzahl_zaehler=1.
27. Mueller D, Uibel S, Braun M, *et al*. Tobacco smoke particles and indoor air quality (ToPIQ) - the protocol of a new study. *J Occup Med Toxicol* 2011;6:35.
28. Schimpf-Ing. Electronics Development. 2015 http://www.schimpf-ing.no/index_e.html.
29. Grimm. Grimm Aerosol Technik GmBH & Co.KG Ainring. Portable laser aerosolspectrometer and dust monitor Model 1.108/1.109 (Manual). 2010 http://wmo-gaw-wcc-aerosol-physics.org/files/OPC-Grimm-model-1.108-and-1.109.pdf.
30. Grimm. Grimm Aerosol Technik GmBH & Co.KG Ainring. GRIMM Software für Optical Particle Counter Tragbares Aerosolspektrometer 1.108/1.109. 2012 http://wiki.grimm-aerosol.de/images/c/c6/M_d_labview_software_rev_2p1.pdf.
31. CEN. *European Committee for Standardization: Workplace atmospheres-size fraction definitions for measurement of airborne particles (Report No. BS EN 481:1993)*. London, England: British Standards Institute, 1993.
32. WHO. World Health organization. Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide. 2005. Global update 2005. Summary of risk assessment. http://apps.who.int/iris/bitstream/handle/10665/69477/WHO_SDE_PHE_OEH_06.02_eng.pdf.
33. EPA.U.S. Environmental Protection Agency. U.S. Department of Energy(DOE). Fuel Economy Guide, 2018. https://www.fueleconomy.gov/feg/pdfs/guides/FEG2018.pdf.
34. Weitzman M, Yusufali AH, Bali F, *et al*. Effects of hookah smoking on indoor air quality in homes. *Tob Control* 2016;26:586–91.
35. Zhou S, Behrooz L, Weitzman M, *et al*. Secondhand hookah smoke: an occupational hazard for hookah bar employees. *Tob Control* 2017;26:40–5.
36. Soule EK, Maloney SF, Spindle TR, *et al*. Electronic cigarette use and indoor air quality in a natural setting. *Tob Control* 2017;26:109–12.
37. Semple S, Apsley A, Azmina Ibrahim T, *et al*. Fine particulate matter concentrations in smoking households: just how much secondhand smoke do you breathe in if you live with a smoker who smokes indoors? *Tob Control* 2015;24–e205–e211.
38. Wertz MS, Kyriss T, Paranjape S, *et al*. The toxic effects of cigarette additives. Philip Morris' project mix reconsidered: an analysis of documents released through litigation. *PLoS Med* 2011;8:e1001145.
39. Gaworski CL, Dozier MM, Gerhart JM, *et al*. 13-week inhalation toxicity study of menthol cigarette smoke. *Food Chem Toxicol* 1997;35:683–92.
40. Gerharz J, Bendels MHK, Braun M, *et al*. Particulate matter emissions of different brands of mentholated cigarettes. *J Air Waste Manag Assoc* 2018;68:608–15.
41. Keith CH, Derrick JC. Measurement of the particle size distribution and concentration of cigarette smoke by the “conifuge”. *J Colloid Sci* 1960;15:340–56.

42. Nazaroff WW, Klepeis NE. *Environmental Tobacco Smoke Particles. Indoor Environment: Airborne Particles and Settled Dust*. Weinheim: Wiley-VCH, 2003:245–74.
43. Guerin MR, Higgins CE, Jenkins RA. Measuring environmental emissions from tobacco combustion: Sidestream cigarette smoke literature review. *Atmospheric Environment* 1987;21:291–7.
44. Ueno Y, Peters LK. Size and Generation Rate of Sidestream Cigarette Smoke Particles. *Aerosol Science and Technology* 1986;5:469–76.
45. Haustein KO, Groneberg DA. *Groneberg. Tabakabhängigkeit – Gesundheitliche Schäden durch das Rauchen*: Springer-Verlag Berlin Heidelberg:, 2008.
46. Burkart J, Steiner G, Reischl G, *et al*. Characterizing the performance of two optical particle counters (Grimm OPC1.108 and OPC1.109) under urban aerosol conditions. *J Aerosol Sci* 2010;41:953–62.
47. Grimm H, Eatough DJ. Aerosol measurement: the use of optical light scattering for the determination of particulate size distribution, and particulate mass, including the semi-volatile fraction. *J Air Waste Manag Assoc* 2009;59:101–7.
48. EPA.United States Environmental Protection Agency. List of Designated Reference and Equivalent Methods, 2017. https://www3.epa.gov/ttn/amtic/files/ambient/criteria/AMTIC_List_June_2017_update_6-19-2017.pdf. (accessed 07 Aug 2018).
49. CEN. European Committee for Standardization. *CEN/TC 264 - Air quality EN 12341. Ambient air - Standard gravimetric measurement method for the determination of the PM10 or PM2,5 mass concentration of suspended particulate matter*. London: British Standard Institute, 2014. https://standards.cen.eu/dyn/www/f?p=204:110:0::::FSP_PROJECT,FSP_ORG_ID:29133,6245&cs=1DC6EB16DD302E384B46A7097AAC67CB5. (accessed 07 Aug 2018).
50. Bolte G, Heitmann D, Kiranoglu M, *et al*. Exposure to environmental tobacco smoke in German restaurants, pubs and discotheques. *J Expo Sci Environ Epidemiol* 2008;18:262–71.
51. Fromme H, Twardella D, Dietrich S, *et al*. Particulate matter in the indoor air of classrooms—exploratory results from Munich and surrounding area. *Atmos Environ* 2007;41:854–66.
52. McGrath C, Warren N, Biggs P, *et al*. Real-time measurement of inhaled and exhaled cigarette smoke: Implications for dose. *Journal of Physics: Conference Series* 2009;151:012018.
53. Sahu SK, Tiwari M, Bhangare RC, *et al*. Particle Size Distribution of Mainstream and Exhaled Cigarette Smoke and Predictive Deposition in Human Respiratory Tract. *Aerosol Air Qual Res* 2013;13:324–32.
54. Keil U, Prugger C, Heidrich J. Passivrauchen. *Public Health Forum* 2016;24:84–7.
55. Nowak D, Raupach T, Radon K, *et al*. Passivrauchen als Gesundheitsrisiko. *Pneumologe* 2008;5:386–92.
56. ISO. International Organization for Standardization. ISO/TR 17219:2013(en). Review of human smoking behaviour and recommendations for a new ISO standard for the machine smoking of cigarettes. 2013 https://www.iso.org/obp/ui/#iso:std:iso:tr:17219:en.
57. WHO. World Health Organization. WHO TobLabNet Official Method SOP 01. Standard operating procedure for intense smoking of cigarettes. 2012 http://apps.who.int/iris/bitstream/10665/75261/1/9789241503891_eng.pdf.
58. Hammond D, Wiebel F, Kozlowski LT, *et al*. Revising the machine smoking regime for cigarette emissions: implications for tobacco control policy. *Tob Control* 2007;16:8–14.
59. Liu C, G. McAdam K, A. Perfetti T. Some Recent Topics in Cigarette Smoke Science. *Mini Rev Org Chem* 2011;8:349–59.
60. Marian C, O'Connor RJ, Djordjevic MV, *et al*. Reconciling human smoking behavior and machine smoking patterns: implications for understanding smoking behavior and the impact on laboratory studies. *Cancer Epidemiol Biomarkers Prev* 2009;18:3305–20.
61. Wright C. Standardized methods for the regulation of cigarette-smoke constituents. *TrAC Trends in Analytical Chemistry* 2015;66:118–27.