

Influence of Type and Amount of Carbon in Cigarette Filters on Smokers' Mouth Level Exposure to “Tar”, Nicotine, 1,3-Butadiene, Benzene, Toluene, Isoprene, and Acrylonitrile *

by

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SUMMARY

Activated carbons are effective adsorbents for many volatile organic compounds and are used in cigarette filters to remove selected smoke toxicants. Polymer-derived carbon is more effective in removing many vapour phase toxicants found in cigarette smoke than coconut-shell-derived carbon. We compared mouth-level exposure to “tar”, nicotine and five vapour phase constituents (1,3-butadiene, benzene, toluene, isoprene, acrylonitrile) in two groups of Romanian smokers of 4-mg or 8-mg International Organization for Standardization (ISO) “tar” bands. Test cigarettes with 4 and 8 mg ISO “tar” were manufactured for the study with two target levels of polymer-derived carbon (30 mg and 56 mg), along with control cigarettes containing a target level of 56 mg of coconut-shell-derived carbon in both “tar” bands. No significant differences were found between mouth-level exposure to “tar” or nicotine yields obtained from control and test products ($p > 0.05$) in either ISO “tar” band. Mouth-level exposure to each of the five vapour phase constituents was significantly lower from the test products with polymer-derived carbon ($p < 0.0001$) than from control cigarettes with coconut-shell-derived carbon, by an average of 25% with 30 mg polymer-derived carbon and around 50% with 56 mg. [Beitr. Tabakforsch. Int. 27 (2016) 40–53]

ZUSAMMENFASSUNG

Aktivkohlen sind wirksame Adsorbentien für viele flüchtige organische Verbindungen und werden in Zigarettenfiltern verwendet, um bestimmte Giftstoffe aus dem Rauch zu entfernen. Die Kohle aus Polymer ist effektiver bei der Entfernung vieler Giftstoffe aus Zigarettenrauch in der Dampfphase als die Kohle aus Kokosnussschale. Es wurde die Exposition im Mundraum gegenüber “Teer”, Nikotin und fünf Bestandteilen der Dampfphase (1,3-Butadien, Benzol, Toluol, Isopren, Acrylnitril) in zwei Gruppen rumänischer Raucher mit 4-mg oder 8-mg “Teerwert”, bestimmt nach ISO (International Organization for Standardization), verglichen. Für die Studie wurden Testzigaretten mit 4 und 8 mg “Teer” mit zwei Zielkonzentrationen der Kohle aus Polymer (30 mg und 56 mg) sowie Kontrollzigaretten mit einem Zielwert von 56 mg Kohle aus Kokosnussschale in beiden “Teerniveaus” hergestellt. In keinem ISO-“Teerniveau” wurden signifikante Unterschiede zwischen der Exposition im Mundraum gegenüber “Teer” oder Nikotinausbeuten aus Kontroll- oder Testzigaretten ($p > 0,05$) festgestellt. Die Exposition im Mundraum gegenüber jedem der fünf Bestandteile der Dampfphase lag bei den Testprodukten mit Kohle aus Polymer signifikant niedriger ($p < 0,0001$) als die bei den Kontrollzigaretten mit Kohle aus Kokosnussschale, und zwar durchschnittlich um 25% bei 30 mg Kohle aus Polymer und etwa 50% bei 56 mg. [Beitr. Tabakforsch. Int. 27 (2016) 40–53]

RESUME

Les charbons actifs sont de puissants agents d'adsorption, capables de capturer de nombreux composés organiques volatiles; ils sont utilisés dans les filtres de cigarette afin de retenir une sélection de substances toxiques présentes dans la fumée. Comparativement au charbon dérivé de la coque de noix de coco, le charbon dérivé de polymères s'avère plus efficace à éliminer de nombreuses substances toxiques en phase gazeuse présentes dans la fumée de cigarette. Nous avons mis en regard les données d'exposition au niveau buccal pour la nicotine, le goudron et cinq constituants en phase gazeuse (1,3-butadiène, benzène, toluène, isoprène, acrylonitrile) parmi deux groupes de fumeurs roumains répertoriés dans les plages de goudron normalisées ISO à 4 mg ou 8 mg. Aux fins de la présente étude, des cigarettes d'essai contenant du goudron normalisé ISO à 4 ou 8 mg furent fabriquées avec deux niveaux ciblés de charbon dérivé de polymères (30 mg et 56 mg), ainsi que des cigarettes de référence contenant un niveau ciblé de 56 mg de charbon dérivé de coque de noix de coco dans les deux plages de goudron. Aucune différence significative ne fut observée entre les données d'exposition relevées, au niveau buccal, pour la teneur en nicotine ou en goudron, sur les produits d'essai et de référence ($p > 0,05$) quel que soit la plage de goudron normalisée ISO. Les données d'exposition au niveau buccal pour chacun des cinq constituants en phase gazeuse furent significativement inférieures pour les produits d'essai contenant du charbon dérivé de polymères ($p < 0,0001$) que pour les cigarettes de référence contenant du charbon dérivé de coque de noix de coco, cet écart avoisina une moyenne de 25% dans le cas du dispositif à 30 mg de charbon dérivé de polymères et 50% dans le cas du dispositif à 56 mg. [Beitr. Tabakforsch. Int. 27 (2016) 40–53]

KEY WORDS:

Carbon, smokers, mouth-level exposure, nicotine, filter tip

ABBREVIATIONS

CA	cellulose acetate
CSD	coconut-shell-derived
HCI	Health Canada Intense
ISO	International Organization for Standardization
MLE	mouth-level exposure
PD	polymer-derived
PFM	part-filter method

1. INTRODUCTION

Cigarette smoking may cause serious disease through frequent and sustained exposure to toxicants (1), and health risks may be dose related (2). Tobacco smoke comprises more than 6,000 constituents (3) and exposure to many of these has been linked with disease in humans (4). Typically, "tar", nicotine and carbon monoxide are measured using smoking machine methods based on the International

Organization for Standardization (ISO) (5–8) testing regime to provide a ranking of brands, but this ranking may not transfer to other smoke constituents (9–10). Also, machine smoking provides a ranking measurement that is not necessarily representative of the yields achieved by smokers. The World Health Organization Study Group on Tobacco Product Regulation (9) has recommended a strategy of characterising products on the basis of smoke toxicity, and a priority list of toxicants for reduction in tobacco smoke has been proposed (9). The US Food and Drug Administration Tobacco Products Scientific Advisory Committee has identified 93 harmful or potentially harmful tobacco smoke constituents (11), of which 20 must be measured and reported by US tobacco manufacturers. Future product standards may include limits on smoke toxicants (11).

Activated carbons are effective adsorbents for many volatile organic compounds (12), and are used in cigarette filters to remove selected toxicants from cigarette smoke. It has been shown previously that different levels of carbon can influence the magnitude of the reduction in smokers' exposure to toxicants (13).

Typically, coconut-shell derived (CSD) carbon is used in cigarette filters, but a synthetic polymer-derived (PD) carbon (Blücher GmbH, Erkrath, Germany) is approximately twice as effective in removing many vapour phase toxicants (14). Cigarette designs that included PD carbon in the filter have reduced smokers' exposure to some toxicants, but the level of influence of the PD carbon could not be separated from that of other design differences (15–17). To investigate the impact on smokers' mouth-level exposure (MLE) of using PD carbon in the filter, we designed a study to compare differences in MLE for "tar", nicotine and five vapour phase compounds (1,3-butadiene, benzene, toluene, isoprene, acrylonitrile) between smokers of test and control cigarettes in which the only difference was the type of carbon or its mass loading. These five vapour phase compounds were selected because a common method is used to determine their concentrations in mainstream smoke. Two of the compounds (1,3-butadiene and benzene) are on the Study Group on Tobacco Product Regulation list of toxicants for which lowering is mandated (9), and all five are in the US Food and Drug Administration list of 20 recommended for measuring and reporting (11).

Various assays and methods are available to assess smokers' MLE to tobacco smoke constituents by analysis of cigarette filters (18). To assess smokers' MLE to "tar", nicotine and the five compounds of interest, we used a method based on the part-filter method (PFM) described by ST.CHARLES *et al.* (19), which relies on the relationships between the mainstream smoke yields of "tar" and nicotine and the amounts of "tar", nicotine or solanesol (20–27) retained within the filter. The PFM has also been used to estimate smokers' MLE to other smoke constituents: tobacco-specific nitrosamines (10, 27, 28, 30, 31), polycyclic aromatic hydrocarbons (20, 28, 29, 31), hydroxybenzene (21), volatile organic compounds (22, 28), solanesol (23–25), carbonyl compounds (26, 28, 31) and carbon monoxide (28). In our study, filter nicotine was used as a marker for the mainstream yields of nicotine and the five compounds of interest. The methodology is unobtrusive and, therefore, more likely to reflect normal smoking

behaviour than other laboratory-based methods of smoking behaviour assessments. Studies have also shown that MLE estimates from filter studies correlate well with biomarkers of exposure and, hence, provide a useful way to collect exposure information in large populations without the need for clinical studies (32–33).

The study objective was to use PFM to compare MLE to “tar”, nicotine and five vapour phase constituents in smokers of 4-mg and 8-mg ISO “tar” cigarettes with two levels of PD carbon and controls containing CSD carbon. Additionally, we assessed the sensory effects of PD carbon in the filter compared with that of CSD carbon.

2. MATERIALS AND METHODS

The filters were manufactured at Essentra Filter Products (Tyne and Wear, UK) and the cigarettes by British American Tobacco (Bayreuth, Germany). The analytical work was carried out by Labstat International ULC (Kitchener, ON, Canada). Benzene, 1,3-butadiene, toluene, isoprene, and acrylonitrile were measured in the vapour phase of smoke. Nicotine extracted from smoked part filters was used to estimate the yields of these five vapour phase compounds.

2.1 Products

Ten thousand of each of the study test cigarettes were manufactured: two control cigarettes at 4-mg and 8-mg ISO “tar” levels, each with a target of 56 mg CSD carbon in the filter, two test products at 4-mg ISO “tar” with targets of 30 mg or 56 mg of PD carbon in the filter and the same in test products with 8 mg ISO “tar”. These values encompass the loadings typically used in commercial products. The cigarettes were specified to be 83 mm long with 25 mm circumference. The cigarette filters comprised two 7.5-mm cellulose acetate (CA) sections at the mouth end and tobacco end and a 12.0-mm section comprising CA and carbon in the middle.

Mainstream yields of the five vapour phase compounds were measured under three machine smoking regimes: ISO (5–8), Massachusetts (34) and Health Canada intense (HCI) (35). The vapour phase compounds were measured using Health Canada Method determination of 1,3-butadiene, isoprene, acrylonitrile, benzene and toluene in mainstream tobacco smoke (36). “Tar” and nicotine yields were measured under the ISO and HCI regimes (5–8, 35). These smoking regimes increase in intensity by the use of ventilation blocking, increased puff volume and reduced puffing interval, but they all provide a ranking of the study products.

All product and pack labelling, including health warnings, pack prints and tax stamps, were compliant with Romanian cigarette packaging regulation.

2.2 Participants

The target recruitment was 60 smokers of single commercial brands in the 4-mg and 8-mg “tar” bands to ensure that a minimum of 50 participants in each group would complete the study.

The inclusion criteria for smokers were age 19–49 years and regular smoking of at least eight Kent brand cigarettes per day with ISO “tar” 4-mg or 8-mg for at least 6 months. Exclusion criteria were pregnancy and breastfeeding. We aimed to recruit 50% men and 50% women. All participants gave written informed consent and were free to leave the study at any time. Each individual who completed the study was compensated at a level of € 22.

2.3 Protocol

Fieldwork was carried out by ISRA Centre Marketing Research (Bucharest, Romania) in December, 2014.

The study comprised four visits to a central location. On the first visit, each participant was advised of the activities they were being asked to take part in prior to providing signed informed consent. All individuals were issued a filter cutter designed to cut a 5-mm portion from the mouth-end CA section of the filter and instructions for use. Each filter cutter was uniquely labelled with the participant’s identification code and the test product code. Each participant was issued with study products to take away with them and smoke in their normal environment. The number of cigarettes each person received was based on their self-reported average daily cigarette consumption (ADC) rounded up to the nearest pack. The order of issue of the three study products was randomised to eliminate bias due to order effect. A diary was also given to participants along with instructions about how to record their ADC.

Participants were required to smoke study products over 5 days, with day 1 being the day of the visit. On days 3 and 4 they were required to use the filter cutter to cut and collect at least 15 filters from the study cigarettes they had smoked before returning to the central location for visit 2 on day 5. ISRA staff collected the filter cutters and diaries. To determine the cigarettes’ acceptability, participants were asked to complete a sensory questionnaire about those they had been smoking during the previous 5 days. The required response used a sensory magnitude scale (where 1 was low and 5 was high), and a “just right” (JR) scale (where 1 was “too low”, 3 was “just right” and 5 was “too high”) to evaluate how close sensory characteristics of the smoked products were to the participants’ ideal. The attributes assessed were overall acceptability, taste quality, natural tobacco taste, mouth drying and aftertaste (sensory magnitude only), plus flavour amount, draw effort, mouthful of smoke, immediate smoke delivery, irritation and impact in mouth and throat (sensory magnitude and “just right”). This process was repeated until the study was completed.

Participants were asked not to return to smoking their usual product in between smoking the test products. Those who completed all aspects of the study were compensated for their time. Following completion of the study, the filter cutters containing 5 mm cut mouth-end sections of the participants’ spent cigarettes were shipped to Labstat International ULC by air and stored at ambient temperature until analysis.

2.4 Filter analysis

PFM was used to estimate smokers’ MLE to “tar” and nicotine, as previously described (19). The principle of the methodology was the same, but in this study the PFM was used, to estimate smokers’ MLE to “tar”, nicotine and the

five vapour phase compounds. All analyses were performed under contract by Labstat, Canada using methodology described by ST.CHARLES *et al.* (19) and Health Canada Methods (35, 36). Copies of Health Canada Methods are available on request (e-mail to: tr_rrrt@hc-sc.gc.ca).

Mainstream nicotine yields generated during smoking the study products were determined by Labstat, Canada using methodology based on Health Canada Method T-115 (35). The five mainstream vapour phase compounds generated during the smoking of the calibration brands were determined under contract by Labstat, Canada, using Health Canada Method T-116 (36).

Five cigarettes were smoked for each smoke run to prepare each sample for analysis using a single impinger containing 20 mL methanol below -70°C . Standards were prepared at lower concentrations based on the British American Tobacco method determination of selected volatiles in mainstream smoke (see BAT Science website at http://www.bat-science.com/groupms/sites/BAT_9GVJXS.nsf/vwPagesWebLive/DO7AXGCL?opendocument.) All samples were analysed using gas chromatography-mass spectroscopy operating in selective ion monitoring (SIM) mode.

Smoking replicates were produced across two days to allow for day to day variation of the smoking procedure. Following the completion of each smoke run, a 5-mm filter CA mouth-end section cut was taken from each cigarette and each set of five cut filters were stored at ambient temperature to be analysed alongside cut filters from the field study. The calibration smoking was performed in the same time period as the study field work to enable similar aging of the part filters (19).

The human smoked tips were split into three sets of five tips, in cases where there were less than 15 tips, where possible, the number of tips in each replicate was reduced rather than reducing the number of replicates (19). In this study approximately 90% of the replicates comprised five part filters and more than 95% of the samples comprised three replicates. Each replicate was analysed on a separate day to remove the concern of analytical failure issues losing all the data points for one participant. The human tips and calibration tips were intermixed within each tip batch to ensure concurrent testing of each participant sample.

The length of each tip was measured (± 0.1 mm) and recorded before five tips were extracted in 20 mL methanol and analysed for tip nicotine by GC (19).

MLE to nicotine was obtained for each extract by using the nicotine values from human smoked cigarettes and the linear regression equation obtained by plotting mainstream smoke nicotine yield versus tip nicotine data obtained during calibration. Similarly, MLE to each of the five vapour phase compounds was obtained using measured human tip nicotine values and the linear regression equation derived by plotting the appropriate vapour phase yield versus tip nicotine data obtained during calibration.

2.5 Data analysis

Minitab 17 (Minitab Inc., State College, PA, USA) and SAS 9.3 (SAS Institute Inc., Cary, NC, USA) statistical software packages were used to conduct data analysis.

Machine smoking regime results are portrayed as mean and standard deviation (SD). Data below the level of quantification (LOQ) are reported as half the LOQ. Vapour phase compound yields from the study products were compared

within each of the ISO “tar” bands using the mixed model procedure by machine smoking test regime. Where differences were found ($p < 0.05$), Tukey’s post hoc test was used to investigate the source of the difference.

Smokers’ MLE to “tar”, nicotine and vapour phase compound results are shown as mean and SD. The MLE data were compared within each of the ISO “tar” bands using Analysis of Variance General Linear Model (ANOVA GLM) with participant as a random factor. Where differences were found ($p < 0.05$), Tukey’s post hoc test was used to investigate the source of the difference.

The average daily cigarette consumption of study and non-study cigarettes reported in the diaries are shown as mean and SD. Cigarette consumption for each smoker group were compared using ANOVA GLM with participant as a random factor. Where differences were found ($p < 0.05$), Tukey’s post hoc test was used to investigate the source of the difference.

The ‘sensory’ scores were compared using the mixed model procedure with participant as a repeated factor. The ‘just right’ scores were compared using a 1-sample t-test with the hypothesised mean of 3 (just right).

3. RESULTS

3.1 Products

The objective of the study relied on the assumption that the manufactured test products had “tar” deliveries of 4 mg and 8 mg under ISO smoking conditions and that the products within an ISO “tar” band were closely matched. Comparison of “tar”, and nicotine yields generated from the ISO testing showed some significant differences within ISO “tar” bands (Table 1).

Table 1. ISO and HCl smoking regime results for “tar” and nicotine. Tukey’s test is represented by superscript letters. The same letter (within a comparison) denotes no significant difference $p > 0.05$.

Smoking regime	Product	Nicotine (mg/cig)	“Tar” (mg/cig)
		mean \pm SD	
ISO (n = 2)	56 mg CSD 4 mg	0.41 \pm 0.00 ^a	4.5 \pm 0.04
	56 mg PD 4 mg	0.36 \pm 0.01 ^b	4.3 \pm 0.13
	30 mg PD 4 mg	0.39 \pm 0.00 ^{ab}	4.4 \pm 0.01
	p value	0.013	0.144
	56 mg CSD 8 mg	0.66 \pm 0.00 ^a	8.3 \pm 0.12 ^a
	56 mg PD 8 mg	0.64 \pm 0.00 ^b	8.3 \pm 0.07 ^a
	30 mg PD 8 mg	0.59 \pm 0.00 ^c	7.7 \pm 0.04 ^b
	p value	0.001	0.009
HCl (n = 5)	56 mg CSD 4 mg	1.64 \pm 0.05 ^b	19.9 \pm 0.50
	56 mg PD 4 mg	1.78 \pm 0.13 ^a	20.6 \pm 1.44
	30 mg PD 4 mg	1.61 \pm 0.02 ^b	19.7 \pm 0.69
	p value	0.014	0.363
	56 mg CSD 8 mg	1.67 \pm 0.05 ^{ab}	21.6 \pm 0.57 ^b
	56 mg PD 8 mg	1.74 \pm 0.04 ^a	21.2 \pm 0.54 ^b
	30 mg PD 8 mg	1.64 \pm 0.04 ^b	22.8 \pm 0.42 ^a
	p value	0.006	0.001

Abbreviations: CSD: coconut-shell-derived; HCl: Health Canada Intense; PD: polymer-derived.

As these differences were within 10% and 15%, respectively, they were deemed to be within the acceptable levels of repeatability of matched cigarette samples (7).

All six study products were also smoked under HCI conditions. Some significant differences were found between the “tar” and nicotine yields within ISO “tar” bands (Table 1), but all were within 10%.

Physical measurement data of the study cigarettes are shown in Table 2. The weight of the PD carbon in the 56-mg test products were within 4% and 1% respectively of the target quantity. The weight of PD carbon in the 30-mg test products was within 12% and 18%, whereas the weight of the CSD carbon in the control products was over the target weight, but within 22% and 12% respectively.

Table 2. Physical measurement results.

Product	Tobacco weight (mg) (n = 10)	Total cigarette weight (mg) (n = 10)	Filter carbon weight (mg) (n = 10)	Open pressure drop (mmWG) (n = 10)	Tip ventilation (%) (n = 10)
Mean ± SD					
56 mg CSD 4 mg	591	930	68.1 ± 4.7	85 ± 2.2	56.7 ± 0.5
56 mg PD 4 mg	623	947	54.1 ± 3.1	86 ± 2.1	58.5 ± 1.83
30 mg PD 4 mg	591	896	33.4 ± 2.0	84 ± 2.0	56.8 ± 1.2
56 mg CSD 8 mg	663	993	65.5 ± 3.5	115 ± 4.6	30.1 ± 2.27
56 mg PD 8 mg	640	962	55.6 ± 4.5	105 ± 5.1	29 ± 1.98
30 mg PD 8 mg	670	972	35.3 ± 1.0	112 ± 3.1	30 ± 1.33

Abbreviations: CSD: coconut-shell-derived; PD: polymer-derived; mmWG: mm water gauge.

3.2 Participants

Of the 60 people recruited, 54 completed the study for the 4-mg ISO “tar” level, and 56 for the 8-mg “tar” level. Table 3 lists the number of participants by gender and age.

Table 3. Participants’ demographics.

Smoker group	Total number	Male	Female	Age 19 – 29 years	Age 30 – 49 years	Cigarette consumption (per day) ^a
4 mg	54	28	26	25	29	20.1 ± 2.3
8 mg	56	26	30	27	29	19.8 ± 2.2

^a Included is the average self-reported daily cigarette consumption for each person’s usual product, reported at recruitment. (Mean ± SD)

3.3 Regulatory smoking regimes data

The six products were smoked according to ISO, Massachusetts and HCI test regimes and the results for five vapour phase compounds are displayed in Table 4. It should be noted that under the ISO regime some of the values for acrylonitrile, benzene and toluene were below

the LOQ of the methodology.

In two cases there was a small increase in 1,3-butadiene yield for the test products containing 30 mg PD carbon compared with the control products containing 56 mg CSD carbon in the filter. In all other cases within an ISO “tar” band, the test products containing PD carbon yielded less than the control product. Comparison within each ISO “tar” band across all smoking regimes and mainstream yields found significant reductions for PD carbon in all cases except two (4-mg ISO “tar” band, HCI for 1,3-butadiene and ISO for toluene). Comparisons were conducted where $p < 0.05$ and showed that 56-mg PD product had significantly lower yields in every case than the control. Regarding the 30-mg PD products, under the ISO regime both the 4-mg and 8-mg ISO “tar” bands showed significantly lower yields of all five vapour phase compound yields than the control in all cases apart from 4-mg ISO “tar” band, toluene. For the Massachusetts and HCI test regimes, 30-mg PD products yielded significantly lower yields of isoprene, toluene and benzene than with the control, the 8-mg ISO “tar” product yielded significantly lower acrylonitrile yields and no significant differences were found for 1,3-butadiene yields.

3.4 Calibration smoking

The calibration curves from the calibration smoking and filter analysis performed by Labstat International ULC were linear and the relationships between the vapour phase constituents and tip nicotine ranged R^2 values of 0.84–0.97.

3.5 Smokers’ MLE

Smokers’ MLE to “tar”, nicotine and the five vapour phase compounds are displayed in Table 5. Within an ISO “tar” band, no significant differences were found in MLE to “tar” or nicotine yields between control and test products ($p > 0.05$). MLE to the five vapour phase compounds obtained from the test product with PD carbon differed significantly from that with controls. In all cases, the 56-mg PD products yielded significantly lower yields of the five vapour phase compounds than did the applicable CSD control, leading to MLE reductions, on average, of around 50%. In all cases except the PD 8-mg “tar” band products for acrylonitrile, the 30-mg PD products yielded significantly lower yields of the vapour phase compounds than the CSD control, with an average MLE reduction of around 25%.

The distribution of MLE to “tar” and nicotine data for the 4-mg and 8-mg ISO “tar” band smokers, by product, are shown as cumulative frequency plots in Appendix Figures 1 and 2. The study cigarette yields under the ISO and HCI regimes are also shown. MLE did not differ for “tar” and nicotine between the control and test products within ISO “tar” bands. Yields under the ISO regime are representative of around the fifth percentile of smokers in the two “tar” bands. Those under the HCI regime are representative of the 95th percentile for the 4-mg ISO “tar” band smokers and the 80th–85th percentiles for the 8-mg ISO “tar” band smokers.

Table 4. Machine smoking at three regulatory regimes for five vapour phase compounds (n = 3).

Regime	Product	1,3-Butadiene (µg/cig) mean ± SD	p value	Isoprene (µg/cig) mean ± SD	p value	Acrylonitrile (µg/cig) mean ± SD	p value	Benzene (µg/cig) mean ± SD	p value	Toluene (µg/cig) mean ± SD	p value
ISO	56 mg CSD 4 mg	13.0 ± 0.7 ^a		85.3 ± 1.2 ^a		1.35 ± 0.1 ^a		6.51 ± 0.36 ^a		5.56 ± 2.42	
	56 mg PD 4 mg	5.2 ± 0.5 ^c	< 0.0001	22.4 ± 3.6 ^c	< 0.0001	0.47 ± 0.00 ^b	0.0077	2.32 ± 0.00 ^b	< 0.0001	4.16 ± 0.00	0.4219
	30 mg PD 4 mg	8.5 ± 1.2 ^b		37.1 ± 6.0 ^b		0.69 ± 0.39 ^b		2.32 ± 0.00 ^b		4.16 ± 0.00	
	56 mg CSD 8 mg	26.3 ± 0.9 ^a		170 ± 5.7 ^a		3.2 ± 0.4 ^a		13.8 ± 2.4 ^a		16.1 ± 2.3 ^a	
	56 mg PD 8 mg	10.1 ± 1.6 ^c	< 0.0001	46.4 ± 9.5 ^c	< 0.0001	1.4 ± 0.3 ^b	0.0011	3.11 ± 1.36 ^b	0.0005	4.16 ± 0.00 ^c	0.0001
	30 mg PD 8 mg	16.9 ± 1.6 ^b		77 ± 4.2 ^b		2.2 ± 0.2 ^b		6.97 ± 0.26 ^b		8.85 ± 0.40 ^b	
Mass	56 mg CSD 4 mg	51.1 ± 2.7 ^a		387 ± 36 ^a		7.7 ± 0.2 ^a		26.6 ± 0.2 ^a		38.6 ± 2.0 ^a	
	56 mg PD 4 mg	38.3 ± 2.2 ^b	0.0005	194 ± 10 ^c	0.0001	5.3 ± 0.2 ^b	0.0002	13.6 ± 1.1 ^c	< 0.0001	21.5 ± 3.0 ^b	0.0013
	30 mg PD 4 mg	54.0 ± 2.5 ^a		301 ± 8 ^b		7.5 ± 0.5 ^a		20.4 ± 1.8 ^b		28.4 ± 3.8 ^b	
	56 mg CSD 8 mg	68.8 ± 9.6 ^a		463 ± 59 ^a		11.6 ± 1.1 ^a		42.4 ± 4.2 ^a		58.2 ± 6.7 ^a	
	56 mg PD 8 mg	44.3 ± 3.5 ^b	0.0128	209 ± 17 ^c	0.0006	6.7 ± 0.2 ^c	0.0003	18.0 ± 0.6 ^c	< 0.0001	26.8 ± 1.3 ^c	0.0004
	30 mg PD 8 mg	61.3 ± 6.4 ^{ab}		339 ± 23 ^b		9.5 ± 0.2 ^b		27.8 ± 1.1 ^b		38.4 ± 3.5 ^b	
HCI	56 mg CSD 4 mg	85.8 ± 2.8		645 ± 14 ^a		16.0 ± 0.4 ^a		45.4 ± 2.4 ^a		72.9 ± 3.0 ^a	
	56 mg PD 4 mg	71.2 ± 7.8	0.0889	416 ± 50 ^b	0.0030	11.8 ± 1.0 ^b	0.0016	23.7 ± 2.8 ^c	0.0004	32.1 ± 4.7 ^c	<0.0001
	30 mg PD 4 mg	80.0 ± 7.9		523 ± 63 ^b		15.5 ± 1.0 ^a		34.0 ± 3.8 ^b		50.0 ± 2.1 ^b	
	56 mg CSD 8 mg	87.0 ± 4.3 ^a		642 ± 56 ^a		16.9 ± 0.1 ^a		54.5 ± 2.6 ^a		87.7 ± 1.3 ^a	
	56 mg PD 8 mg	59.3 ± 4.2 ^b	0.0002	299 ± 7 ^c	0.0002	9.8 ± 1.1 ^b	< 0.0001	22.2 ± 0.5 ^c	< 0.0001	32.4 ± 0.8 ^c	< 0.0001
	30 mg PD 8 mg	88.1 ± 3.4 ^a		517 ± 47 ^b		15.6 ± 0.6 ^a		44.0 ± 0.3 ^b		64.0 ± 2.0 ^b	

Tukey's test represented by superscript letters. The same letter (within a comparison) denotes no significant difference $p > 0.05$. Abbreviations: CSD: coconut shell derived; PD: polymer derived.

Table 5. Smokers' MLE to "tar", nicotine and five vapour phase compounds.

Product	Number of participants	"Tar" mean \pm SD	Nicotine mean \pm SD	1,3-butadiene mean \pm SD	Isoprene mean \pm SD	Acrylonitrile mean \pm SD	Benzene mean \pm SD	Toluene mean \pm SD
56 mg CSD 4 mg	52	13.5 \pm 3.8	1.09 \pm 0.29	38.6 \pm 10.8 ^a	311 \pm 94 ^a	7.3 \pm 2.4 ^a	28.5 \pm 9.1 ^a	41.6 \pm 14.0 ^a
56 mg PD 4 mg	53	13.0 \pm 3.7	1.07 \pm 0.30	24.3 \pm 8.4 ^c	141 \pm 53 ^c	4.7 \pm 1.8 ^c	11.4 \pm 4.3 ^c	15.1 \pm 5.6 ^c
30 mg PD 4 mg	54	13.3 \pm 4.0	1.11 \pm 0.32	33.9 \pm 11.3 ^b	215 \pm 78 ^b	6.0 \pm 2.1 ^b	17.4 \pm 6.4 ^b	22.8 \pm 8.4 ^b
p value ANOVA GLM		0.078	0.173	< 0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001
56 mg CSD 8 mg	56	17.6 \pm 7.6	1.30 \pm 0.56	53.1 \pm 21.8 ^a	398 \pm 175 ^a	12.1 \pm 6.1 ^a	40.3 \pm 18.8 ^a	56.1 \pm 27.8 ^a
56 mg PD 8 mg	55	18.0 \pm 6.3	1.32 \pm 0.44	33.5 \pm 14.3 ^c	185 \pm 82 ^c	6.8 \pm 3.1 ^b	16.9 \pm 7.6 ^c	21.3 \pm 9.3 ^c
30 mg PD 8 mg	56	17.4 \pm 6.8	1.28 \pm 0.49	47.3 \pm 19.7 ^b	301 \pm 137 ^b	10.9 \pm 5.5 ^a	28.9 \pm 13.8 ^b	37.5 \pm 18.3 ^b
p value ANOVA GLM		0.789	0.890	< 0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001

Abbreviations: ANOVA GLM: analysis of variance general linear model; CSD: coconut-shell-derived; PD: polymer-derived.

The distribution of MLE to the five vapour phase compounds in smokers of the 4-mg and 8-mg ISO "tar" band products are shown in Appendix Figures 3–7, along with the yields under the ISO and HCI regimes. Similar to MLE to "tar" and nicotine, yields under the ISO regime are representative of around the fifth percentile of smokers for all five vapour phase compounds in the two "tar" bands. Those under the HCI regime are representative of the 95th percentile of smokers for all five vapour phase compounds for the 4-mg ISO "tar" band. In the 8-mg ISO "tar" band, HCI yields are representative of the 95th percentile of smokers for 1,3-butadiene and isoprene and the 75th–85th percentiles for the other compounds. Significant MLE reductions were observed for all five compounds over the whole range of human exposures.

3.6 Average daily cigarette consumption

The average number of study cigarettes recorded in the diaries is listed in Table 6. Most participants reported that they only smoked study cigarettes throughout the study. Comparison of the numbers of study cigarettes reported in the diaries found no significant differences between users of the control and test products within either "tar" band (4-mg ISO, $p = 0.065$, 8-mg ISO $p = 0.712$). Cigarette consumption reported in the diaries was significantly higher than average daily cigarette consumption (ADC) reported at recruitment (4-mg smokers ADC = 20.1, $p = 0.021$, 8-mg smokers ADC = 19.8, $p < 0.0001$).

Table 6. Average daily study cigarette consumption reported in diaries. (Mean across 3 days \pm SD)

56 mg CSD 4 mg	56 mg PD 4 mg	30 mg PD 4 mg
4-mg smoker group		
20.5 \pm 4.3	21.1 \pm 3.8	21.5 \pm 3.8
56 mg CSD 8 mg	56 mg PD 8 mg	30 mg PD 8 mg
8-mg smoker group		
22.7 \pm 5.4	22.4 \pm 6.0	22.9 \pm 5.5

3.7 Sensory scores

Comparison of overall acceptability of the 4-mg and 8-mg ISO "tar" band products found no significant differences between users of the control and test products (4-mg ISO

$p = 0.5925$, 8-mg ISO $p = 0.9931$). Comparison of sensory magnitude scores of the 4-mg and 8-mg ISO "tar" band smoking groups found no significant differences $p < 0.05$. Comparison of "just right" scores showed that the control product was scored marginally lower for the attributes amount of flavour (mean = 2.8, $p = 0.0489$), mouthful of smoke (2.8, $p = 0.0204$), immediate smoke delivery (mean = 2.8, $p = 0.0240$) and impact in mouth and throat (mean = 2.8, $p = 0.0489$).

4. DISCUSSION

The aim of the experimental work reported here was to investigate the influence of type and amount of carbon on smokers' MLE to "tar", nicotine and five selected vapour phase compounds found in tobacco smoke. Comparison of the "tar" and nicotine yields generated under machine smoking regimes ISO and HCI within an ISO "tar" band found some significant differences. However, these differences were not systematically related to carbon type or carbon amount and were likely to be due to the small variation between machine smoking replicate data measured in this study. Previously, BRANTON *et al.* (14) reported that carbon type (CSD versus PD) did not influence the "tar" or nicotine yields generated under the machine smoking regimes ISO and HCI within an ISO "tar" band.

The carbon type and level did not significantly influence smokers' "tar" or nicotine MLE yields between products within an ISO "tar" band. Previously, SARKAR *et al.* (13) found no significant impact on levels of nicotine biomarkers when smokers switched to test cigarettes with high levels of filter carbon.

Mean MLE to "tar" was 13.0–13.5 mg/cig for the 4-mg ISO product and 17.4–18.0 mg/cig for the 8-mg ISO product, and MLE to nicotine was 1.07–1.11 mg/cig for the 4-mg ISO product and 1.28–1.32 mg/cig for the 8-mg ISO product. MLEs were in agreement with those reported previously in a study conducted in Romania with 4-mg and 7-mg ISO "tar" products containing carbon in the filter (4-mg ISO product, "tar" MLE 11.9 \pm 2.9 mg/cig, nicotine MLE 1.04 \pm 0.28 mg/cig; 7-mg ISO product, "tar" MLE 16.3 \pm 4.2 mg/cig, nicotine MLE 1.40 \pm 0.33 mg/cig) (37). The absolute values of the smoke toxicants from the ISO test regime measured in our study were of a similar order of magnitude to those previously reported by BRANTON *et al.* (14) for cigarettes with 60 \pm 1 mg CSD carbon in the filter

(nicotine 0.85 mg/cig, 1,3-butadiene 16.0 µg/cig, benzene 18.2 µg/cig, toluene 29.5 µg/cig, isoprene 117 µg/cig and acrylonitrile 4.8 µg/cig) and by COUNTS *et al.* (38) for a European carbon product with similar nicotine yields under the ISO regime (nicotine 0.65 mg/cig, 1,3-butadiene 25.0 µg/cig, benzene 17.6 µg/cig, toluene 24.2 µg/cig, isoprene 182 µg/cig, acrylonitrile 3.6 µg/cig).

We found that PD carbon was more effective at removing the selected vapour phase compounds investigated in this study than the CSD carbon and it was more effective at the level of 56 mg than 30 mg. This difference was seen in machine smoking yields generated under ISO, Massachusetts and HCI regimes and in smokers' MLE.

BRANTON *et al.* (14) found that PD carbon was approximately twice as effective as CSD carbon at removing selected smoke constituents. SARKAR *et al.* (13) found reductions in biomarkers of exposure to selected smoke constituents in smokers who switched from a conventional product to a product with 180 mg of activated CSD carbon in the filter. In our study, comparison of yields from cigarettes containing 56 mg CSD with cigarettes containing 56 mg PD showed reductions in smokers' MLE of on average 50% for the five vapour phase compounds. Reductions versus 56 mg CSD were also achieved with 30 mg PD, but were on average around 25%.

We found some evidence that daily cigarette consumption increased by a small, but significant amount during the study. It may have been due to participants underestimating their ADC at recruitment, but equally, it may have been the result of being provided with free cigarettes. This has been observed previously (16) and we took measures to ensure participants were provided cigarettes based on their self-reported ADC; however, rounding up to the nearest pack may have influenced the numbers smoked.

Synthetic PD carbon could be used to reduce selected mainstream smoke constituents and may assist with mandated reductions for some toxicants. However, reduced exposure does not necessarily mean reduced risk (16), and the impact of any reduction in smoke constituents on smokers' health is unknown. Reduction in smokers' total exposure is dependent on them not increasing their cigarette consumption or smoking more intensely.

5. CONCLUSIONS

In conclusion, the use of synthetic PD carbon in the filter resulted in significant reductions of five vapour phase compounds (1,3-butadiene, benzene, toluene, isoprene, acrylonitrile) in machine smoking yields and smokers' MLE compared with equivalent loadings of CSD carbon. At lower levels of PD carbon, the reductions versus CSD carbon were less but were, on the whole, still significant. Moreover, these reductions were achieved with little or no effect on overall acceptability or consumption.

CONFLICT OF INTEREST STATEMENT

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Marian Liviu Ionita: Study logistics
John McAughey: Scientific review

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APPENDIX 1

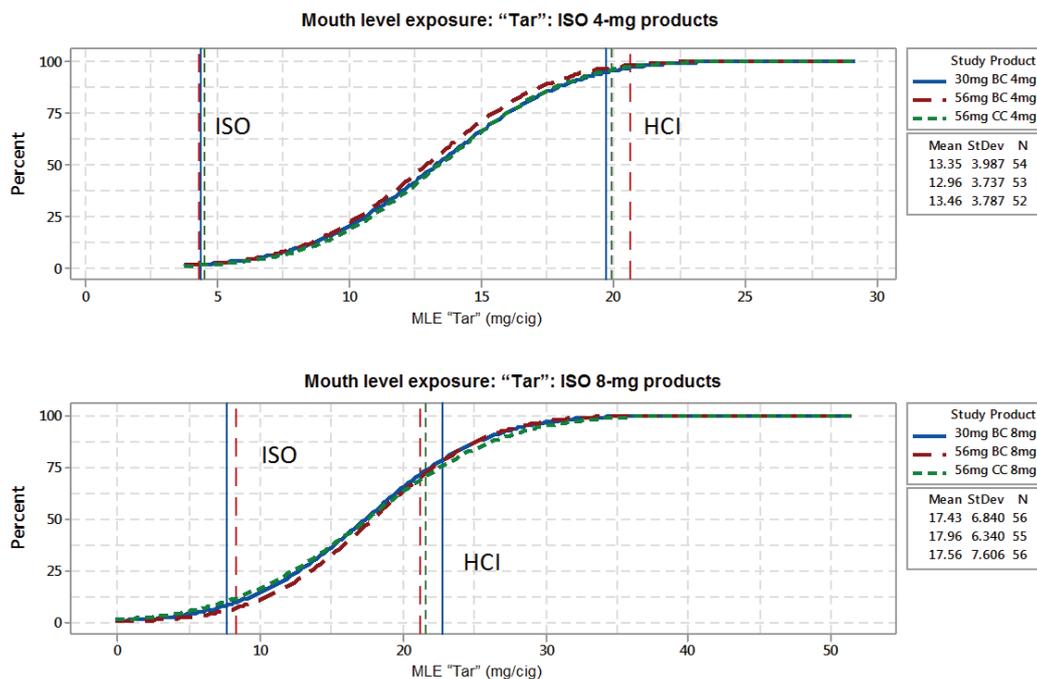


Figure 1. Cumulative frequency distribution of MLE to "tar" for 4-mg and 8-mg ISO "tar" band smokers versus ISO and HCI yields. The vertical lines from the x axis correspond to the ISO and HCI yields.

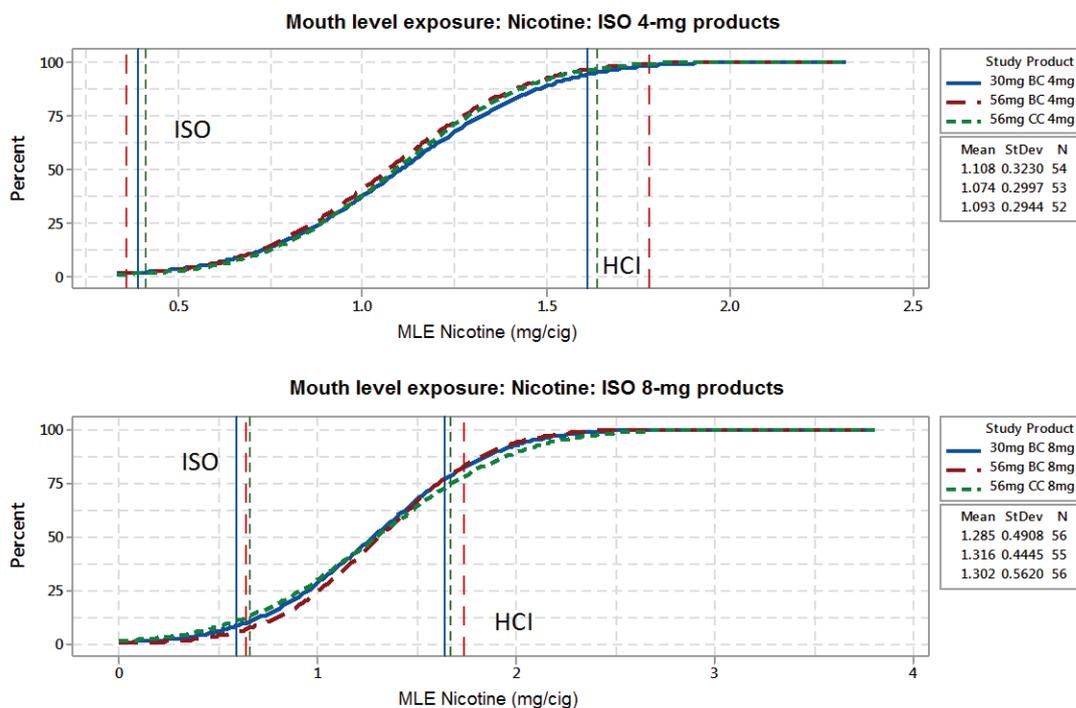


Figure 2. Cumulative frequency distribution of MLE to nicotine for 4-mg and 8-mg ISO "tar" band smokers versus ISO and HCI yields. The vertical lines from the x axis correspond to the ISO and HCI yields.

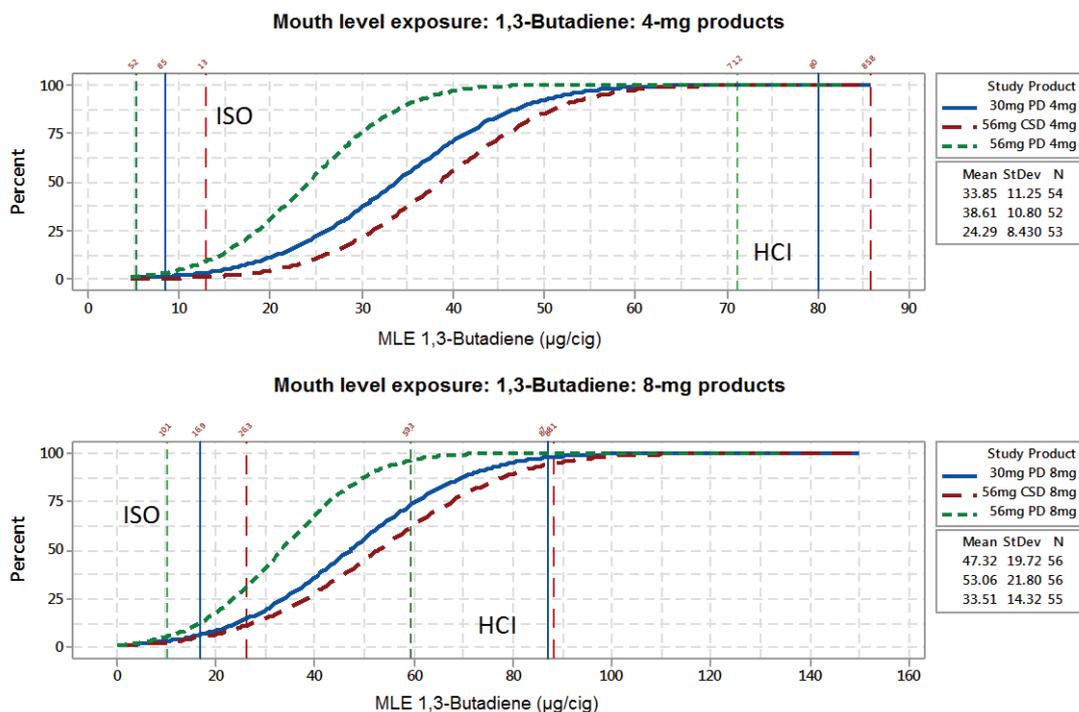


Figure 3. Cumulative frequency distribution of MLE to 1,3-Butadiene for 4-mg and 8-mg ISO “tar” band smokers versus ISO and HCl yields. The vertical lines from the x axis correspond to the ISO and HCl yields.

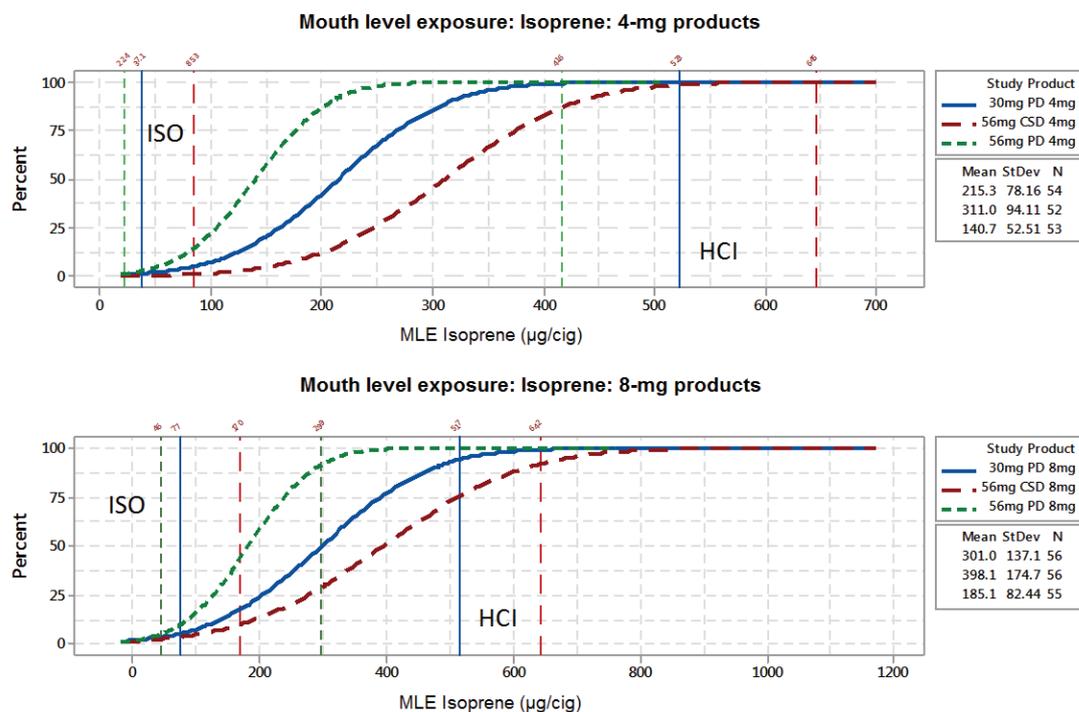


Figure 4. Cumulative frequency distribution of MLE to isoprene for 4-mg and 8-mg ISO “tar” band smokers versus ISO and HCl yields. The vertical lines from the x axis correspond to the ISO and HCl yields.

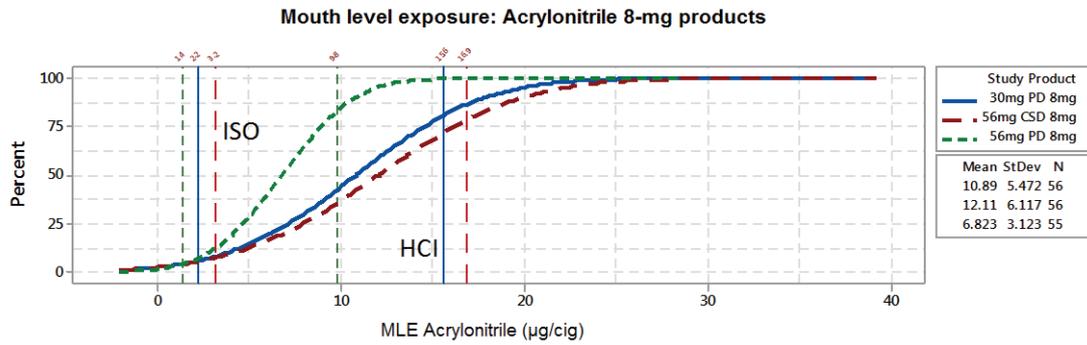
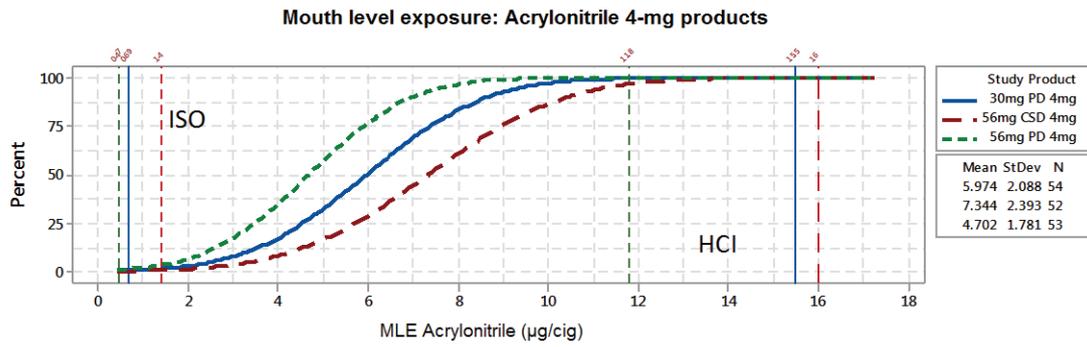


Figure 5. Cumulative frequency distribution of MLE to acrylonitrile for 4-mg and 8-mg ISO “tar” band smokers versus ISO and HCl yields. The vertical lines from the x axis correspond to the ISO and HCl yields.

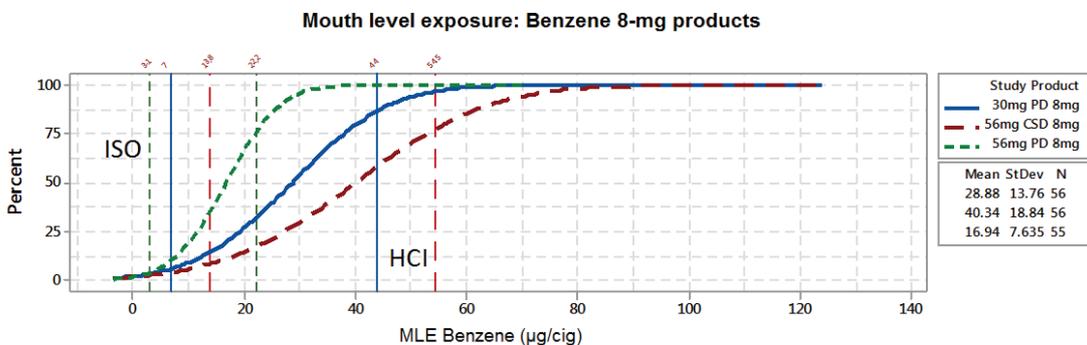
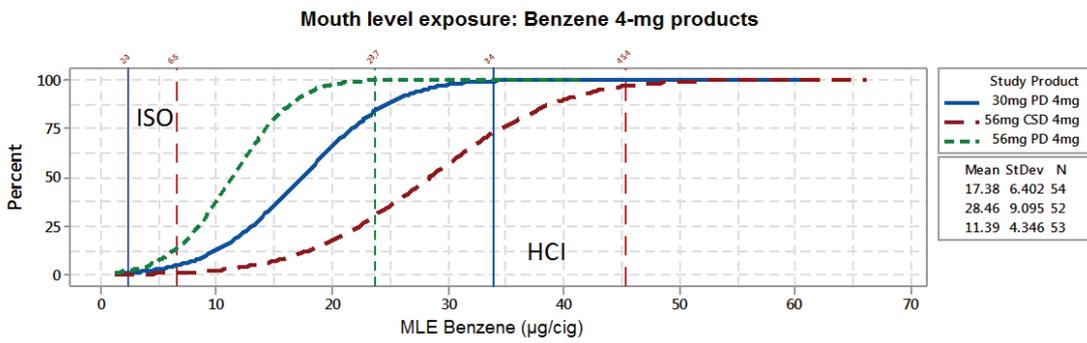


Figure 6. Distribution of MLE to benzene for 4-mg and 8-mg ISO “tar” band smokers versus ISO and HCl yields. The vertical lines from the x axis correspond to the ISO and HCl yields.

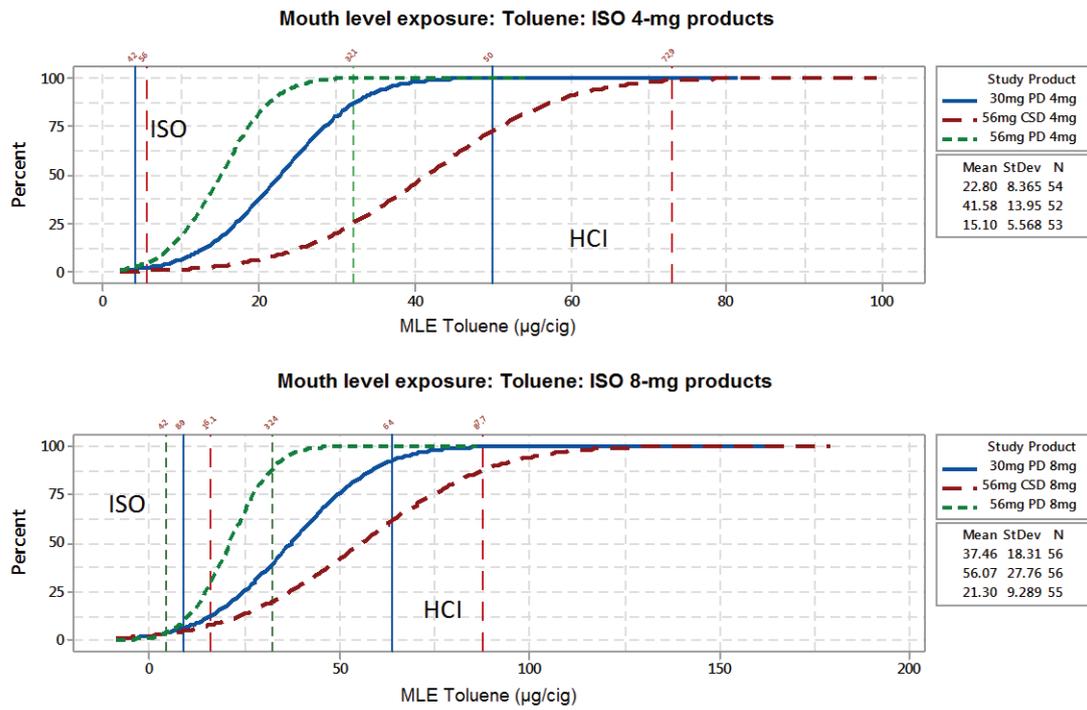


Figure 7. Distribution of MLE to toluene for 4-mg and 8-mg ISO “tar” band smokers versus ISO and HCl yields. The vertical lines from the x axis correspond to the ISO and HCl yields.