Validation of Methods for Determining Consumer Smoked Cigarette Yields from Cigarette Filter Analysis*

by

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SUMMARY

Methods based on the analyses of cigarette filters have been used to estimate 'tar' and nicotine yields to smokers. These methods rely on the measurement of filtration efficiencies (FEs). However FEs may be influenced by both cigarette design features e.g., type of filter and levels of filter ventilation, and human smoking behaviour factors such as puff flow-rates and cigarette butt lengths. Two filter analysis methods are considered in our study. One is based on the analysis of whole filters using average values of FEs obtained from a range of machine smoking regimes. The other, a 'part filter' method, analyses a 10 mm section from the mouth end of the filter where the FE remains relatively constant irrespective of puff flow rates and butt lengths. Human puffing behaviour records were obtained from 10 smokers, each smoking six commercial cigarettes ranging from 1 mg to 12 mg 'tar' yields [International Standard (ISO) values]. These records were used to drive a human smoke duplicator and the resulting 'tar' and nicotine yields obtained from duplication were compared with the estimates obtained from 'whole' and 'part filter' analysis. The results indicated that whilst both filter methods gave good correlations with nicotine and 'tar' yields obtained from smoke duplication, the 'part filter' method was less susceptible to the effect of nicotine condensation and changes in FEs and hence gave a more accurate assessment of yields than the 'whole filter' method. [Beitr. Tabakforsch. Int. 22 (2006) 176–184]

ZUSAMMENFASSUNG

Zur Abschätzung der Kondensat- und Nikotinaufnahme durch Raucher können Methoden verwendet werden, die auf Analysen von Zigarettenfiltern beruhen. Diesen Methoden liegt die Messung der Filtrationseffizienz (FE) zu Grunde. Die Filtrationseffizienz kann jedoch sowohl durch das Zigarettendesign, wie zum Beispiel Filtertyp und Grad der Filterventilation, als auch durch Faktoren des menschlichen Rauchverhaltens, wie die Flussrate während eines Zuges und die Stummellänge, beeinflusst werden. In unserer Untersuchung wurden zwei Filteranalysemethoden berücksichtigt. Die eine Methode basiert auf der Analyse der ganzen Filter. Hierbei werden Durchschnittswerte der FE verwendet, die unter verschiedenen maschinellen Abrauchbedingungen ermittelt wurden. Bei der anderen, einer „Teil-Filter" Methode, wird ein 10 mm großes Stück vom mundseitigen Ende des Filters untersucht. In diesem Bereich bleibt die FE, unabhängig von den Flussraten während des Zuges und der Stummellänge, relativ konstant. Die Werte des menschlichen Rauchverhaltens stammen von 10 Rauchern, die jeweils sechs handelsübliche Zigaretten mit Kondensatgehalten von 1 bis 12 mg (Methode der Internationalen Organisation für Normung, ISO) rauchten. Mit diesen Parametern wurde ein Rauch-Duplikator betrieben und die ermittelten Kondensat- und Nikotinwerte wurden mit den Schätzungen verglichen, die mit der Gesamt- und der Teil-Filter Methode ermittelt wurden. Diese Ergebnisse zeigen, dass beide Filteranalysemethoden eine gute Korrelation mit den aus dem Rauch-Duplikator erhaltenen Nikotin- und Kondensatwerten aufwiesen, die Teil-Filter Methode jedoch weniger empfind-

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lich auf Nikotinkondensation und Veränderungen der FE reagierte und somit eine genauere Bestimmung der Rauchwerte ermöglicht als die Gesamtfilter Methode. [Beitr. Tabakforsch. Int. 22 (2006) 176–184]

RESUME

Des études ont été menées par des méthodes d'analyse des filtres pour estimer les rendements en goudron et nicotine chez les fumeurs. Ces méthodes sont basées sur la mesure de l'efficacités de filtration (FE). Cependant les FEs peuvent être influencées par les caractéristiques de conception des cigarettes, par exemple le type de filtre, les niveaux de ventilation, et par des facteurs du fumage humain, comme le profil du débit de la bouffée et la longueur du mégot. Dans notre étude deux méthodes d'analyse de filtre ont été prises en considération. Une méthode est basée sur l'analyse des filtres entiers en utilisant des valeurs moyennes des FEs, obtenues sous plusieurs régimes de machines à fumer. L'autre méthode examine une section de 10 mm à partir de l'extrémité buccale des filtres, où les FEs restent relativement constantes indépendamment des profils du débit de la bouffée et de la longueur du mégot. Le recensement du comportement de fumage chez les fumeurs est basé sur 10 fumeurs, chacun fumant six cigarettes vendues dans le commerce d'un rendement de 1 à 12 mg de goudron (méthode normalisée de l'Organisation internationale de normalisation, ISO). Ces valeurs ont été utilisées pour piloter une machine à duplication du fumage humain et les rendements en goudron et nicotine obtenus par la duplication ont été comparés avec les estimations obtenues des analyses de filtres entiers ou des sections de filtres. Les résultats indiquent que les deux méthodes fournissent une bonne corrélation avec les rendements en nicotine et goudron obtenus par duplication de la fumée, cependant la méthode examinant des sections de filtres est moins sensible aux effets de la condensation de la nicotine et des changements des FEs et fournit ainsi une évaluation plus précise des rendements que l'analyse des filtres entiers. [Beitr. Tabakforsch. Int. 22 (2006) 176–184]

INTRODUCTION

Standardised methods for the measurement of 'tar' and nicotine yields from cigarettes such as those of the Federal Trade Commission (FTC) and the International Organization for Standardization (ISO) have been in use for the past four decades. These methods measure the amounts of nicotine, 'tar' and carbon monoxide produced by cigarettes when smoked by a machine taking 35 mL, 2 s puffs once a minute until the cigarette reaches a pre-determined length. Standardised smoking regimes were adopted to provide consumers with information on the ranking of cigarettes in terms of 'tar' and nicotine yields rather than purporting to represent the absolute amounts of 'tar' and nicotine smokers would receive from cigarettes (1, 2)

Cigarette smokers vary considerably in their puffing behaviour styles and encompass a wide range of puff volumes, puff durations and inter-puff intervals. The 1988 Report of the US SURGEON GENERAL (3) provides a good example of the range of puffing indices reported in studies of smokers. As smokers can markedly deviate from the standardised puffing parameters used in the FTC/ISO methods, the validity of these methods have been questioned over the past three decades (2–8). Consequently, it would be desirable to have methods of determining the 'tar' and nicotine yields from cigarettes when smoked under normal conditions by consumers. This would help to a) establish whether the FTC/ISO ranking of cigarettes is maintained across groups of consumers and b) whether alternatives to the FTC/ISO methods provide a better correlation with the yields obtained by groups of smokers.

There have been a number of approaches to the development of methods for determining smoke yields obtained by smokers as opposed to smoking machines. One of the least invasive approaches is the collection of 'spent' cigarette filters (cigarette butts) from smokers and the analysis of these filters for nicotine and/or 'tar' content (9–14). With this method, the FEs for nicotine and/or 'tar' for each cigarette type are measured. Then nicotine or 'tar' yields are calculated from the combination of the nicotine or 'tar' retained in the filter and the respective FEs. Unfortunately, the FEs for nicotine and 'tar' can vary according to the flow-rate or velocity of the smoke passing through the filter and, to some extent, the length of the tobacco rod smoked. The flow-rate dependency can be very marked when flow-rates fall below 17.5 mL/s (15). This creates a problem when one considers the FEs of the filter section upstream of the ventilation zone of highly ventilated cigarettes where the flow-rates may fall within this range (16). Consequently it is essential that flow-rate effects are accounted for when attempting to determine nicotine and 'tar' yields from filter analyses, particularly of highly ventilated cigarettes.

BAKER *et al.* (13) attempted to minimise the flow-dependency effect by measuring nicotine FEs over a range of machine smoking regimes and using the average FE to determine nicotine yields from the filters collected from smokers ('whole filter' method). ST.CHARLES *et al.* (17) described a method involving the analysis of a 10 mm portion of the filter taken from the mouth end ('part filter' method). This portion will be downstream of the ventilation zones of most filters and thus filtration in this section will not be subject to a high degree of flow dependency.

The aim of this study was to evaluate and validate the 'whole filter' and 'part filter' methods by comparing the estimates of 'tar' and nicotine yields derived from filter analysis with those obtained directly from a human smoke duplicator, i.e., a modified smoking machine which reproduced the puffing behaviour patterns recorded from smokers.

METHODS

Cigarette products

Six commercial cigarettes were selected to cover the range of yields available within the European Union. Details of these products are shown in Table 1.

The two 1 mg 'tar' yield products, A and B, were chosen due to their differing filter designs [Paper/Cellulose Acetate (CA) dual vs. a mono CA filter], and blend differences

Table 1. Details of the study products

Product code	A	B	C	D	E	F
Source market	Germany	UK	UK	Germany	Germany	Germany
ISO nicotine (mg/cig)	0.1	0.1	0.3	0.4	0.5	0.8
ISO 'tar' yield (mg/cig)	1	1	3	4	6	11
Blend style	USB [a]	VA [b]	VA [b]	USB [a]	USB [a]	USB [a]
Filter type	Paper/CA [c]	CA [c]	CA [c]	CA [c]	CA [c]	CA [c]
Filter length (mm)	25	25	25	27	27	21
Filter ventilation (%)	78	84	70	63	41	18

[a] USB refers to a blend containing Virginia, burley and Oriental tobaccos.
[b] VA refers to a blend containing predominantly Virginia tobaccos.
[c] CA refers to a cellulose acetate filter.

(US blended vs. Virginia respectively). Products C and D were selected on the basis of their similar ISO yields but different blend characters (Virginia vs. US blended). Products E and F were selected as being representative of a typical Lights (E) and a typical Full flavour product (F) in Europe.

In order to minimise variability between cigarettes of the same product type, the cigarettes used in the study were weight (± 3% of mean weight) and pressure drop (± 3% of mean open pressure drop) selected.

Study participants

The smoking behaviour profiles from ten smokers (9 male, 1 female) were used in this study. The smokers were over 18 years of age and regular smokers. The profiles were obtained from participants in a smoking behaviour research programme conducted at the University of Reading. Permission to recruit participants for the smoking behaviour studies was given by the University of Reading ethics committee. The participants were not regular smokers of the products used in this study. This was deemed appropriate since the objective of the study was simply to obtain a range of human smoking behaviour profiles in order to compare estimated yields from filter analysis with actual yields obtained by smoke duplication. All subjects were familiar with the smoking analyser and had previously taken part in similar studies using this equipment.

Smoking behaviour recording, smoke duplication and filter collection

Smoking behaviour recording was achieved by attaching an orifice-plate cigarette holder to the cigarette (18). The differential pressure across the orifice was digitised and stored and was subsequently used to derive puffing parameters such as puff volume, puff duration etc. The puff flow-rate signal was also used to drive a smoke duplicator.

Each participant smoked each product four times. Two of the behavioural records per subject were used for comparing duplicated yields with estimates from 'whole filters', the other two were used for 'part filter' estimates.

The smoking behaviour records were duplicated using a modified single-port Borgwaldt RM1 Plus smoking machine (Borgwaldt Technik GmbH). This device accurately reproduces the puff flow-rate profile for each puff recorded from each smoker. Consequently the duplicator provides measures of 'tar' and nicotine obtained from smoking patterns closely resembling those produced by the smoker. The machine incorporated a valve downstream of the filter that was open to the atmosphere between puffs and closed during the puff, mimicking the removal of the cigarette from the mouth between puffs. Smoke produced during the duplication process was collected on a pre-weighed 44 mm Cambridge Filter (CF) pad. Smoke condensate from five cigarettes was collected onto a single pad for each record from the 1–4 mg products. Three cigarettes were smoked onto a single pad for the higher yield products.

Filters were collected from the cigarettes after each smoke duplication run. The cigarettes were extinguished by removing the coal, and butt lengths were determined by measuring from the filter end to maximum and minimum burn lines. All remaining ash and tobacco was removed from the filter prior to storage in an appropriately labelled 4 mL amber glass screw-top vial. Part filters were obtained by accurately slicing a 10.0 mm section from the mouth end of the filter using a Sodim Model C.04 Filter Cutter (Sodim Instrumentation) immediately after the cigarette had been extinguished.

The 'whole filter' samples were stored in the dark at room temperature and were analysed within eight weeks of collection. The 'part filter' samples were stored in the dark at −20 °C pending analysis for 4 to 5 months after collection.

'Whole filter' analysis and calibration procedures

The calibration for the 'whole filter' method involved the machine smoking of each product using four different regimes in order to cover a range of puff flow-rates from the ISO standard of 17.5 mL/s to a high extreme for human smoking behaviour of 60 mL/s. The machine smoked regimes were (puff volume/puff duration): 35 mL/2 s, 60 mL/2 s, 90 mL/2 s; and 90 mL/1.5 s. A puff frequency of 1 puff per minute was used for all of the smoking regimes. Ten cigarettes were smoked for each brand and calibration regime. The mainstream smoke yields of Total Particulate Matter (TPM), nicotine, water and Nicotine Free Dry Particulate Matter (NFDPM or 'tar') were determined according to ISO methods 3308 (19), 4387 (20), 10362-1 (21) and 10315 (22).

The 'whole filters' were partially split longitudinally and then cut in half transversally before being transferred to a

150 mL conical flask. The 5 'whole filters' from the lower yield cigarettes, or 3 filters from the higher yield cigarettes, were pooled for each duplication run. The filters were extracted into 20 mL of a methanol solution containing 1 mg/mL sodium hydroxide and 0.24 mg/mL *n*-heptadecane internal standard. The flask containing the extraction solution was stoppered and shaken on a flat-bed orbital shaker at medium speed for a minimum of 40 min. The nicotine content of the extract was determined by using an Agilent 5890 gas chromatograph with a Varian CP Wax 52 CB, 0.53 mm × 2 µm × 25 m, fused silica column and flame ionization detector (FID) using CORESTA (Cooperation Center for Scientific Research Relative to Tobacco) Recommended Method No 9 (23).

Filters obtained from the calibration procedures were extracted and analysed in a similar manner to the filters obtained from smoke duplication. However as 10 'whole filters' were obtained from each calibration run the extraction volume was increased from 20 mL to 40 mL. Filters from unsmoked cigarettes were also analysed for residual nicotine content. The measured levels of residual nicotine content were subtracted from the values of nicotine in the filters obtained from the duplication and calibration processes to provide values termed corrected filter nicotine (corrFiltN). Filter Nicotine Filtration Efficiency (NFE) values were calculated for each product at each regime using Eqn. [1].

$$\text{NFE} = \frac{\text{corrFiltN}}{(\text{corrFiltN} + \text{mainstream nicotine yield})} \cdot 100\% \quad [1]$$

A mean value of NFE (mNFE) was calculated across all four smoking regimes for each product.

'Tar'/nicotine ratios (T/N) were calculated for each product at each regime and a mean T/N value (mT/N) was determined by averaging the T/N ratios obtained from the four regimes.

Cigarette yield estimation from 'whole filters'

Two approaches were used to estimate the nicotine and NFDPM yields using the 'whole filters' obtained from the duplication process. The first approach used the amounts of nicotine retained in the filters (corrFiltN), the mean nicotine filtration efficiencies (mNFE) and the mean 'tar' to nicotine ratios (mT/N) obtained from the Cambridge pads produced during the calibration smokings. This is shown in the following two equations:

$$\text{Estimated Nicotine Yield (mg/cig)} = \left(\frac{\text{corrFiltN}}{\text{mNFE}} \cdot 100\right) - \text{corrFiltN} \quad [2]$$

$$\text{Estimated NFDPM Yield (mg/cig)} = \text{Estimated Nicotine yield} \cdot \text{mT/N} \quad [3]$$

The second approach used nicotine filtration efficiencies and T/N ratios corresponding to the average puff flow-rate produced by each smoker. Average puff flow rates were obtained from each individual human smoking record and 'personal' NFE values were interpolated from the relationships between NFE and puff flow-rates for each product type. Similarly 'personal' T/N ratios were calculated from the T/N vs. flow-rate relationships for each product type. The 'personal' NFE and T/N ratios were used instead of the mNFE and mT/N values in Eqns. [2] and [3].

'Part filter' analysis, calibration and estimation of yields

The part filters from the duplication of each smoking profile were split up to give three extractions using 5 mL methanol with decanol internal standard per filter tip. For the calibration tips, four tips were pooled for each extraction and extracted with 20 mL solution. Glass stoppered Erlenmeyer flasks were used for extraction. Flasks were shaken on a flat-bed orbital shaker at 200 rpm for 40 min. This method is similar to CORESTA Recommended Method No. 9 (23) but the *n*-heptadecane was replaced as internal standard by 0.038 mg/mL decanol and NaOH was removed from the extraction solution. The removal of NaOH from the extracting solution was to eliminate possible extract turbidity found to occur with cork tipped cigarettes. Turbidity was a potential problem with the use of the ultraviolet (UV) absorbance method for determining the 'tar' contents of 'part filters' but was not an issue for the 'whole filter' method as the UV absorbance method was not used for 'whole filters'.

The extract was analysed for nicotine using an Agilent 5890 Series II gas chromatograph with a J&W Scientific DB-Wax 0.53 mm × 1.0 µm × 30 m, Megabore® fused silica column and FID. The 'tar' content of the extracts were estimated with a UV absorbance method (24) using a Beckman MDQ capillary electrophoresis instrument with no applied voltage and a 312 nm interference filter on the UV detector. Quinoline in methanol solutions (20, 50, 100, 200, 400 and 1000 ppm) were used as instrument check standards. Selected concentrations were used with each sample batch throughout the study. The coefficients of variation for the standards (N = 10 to 32) were approximately 5% for all but the highest (2.2%) and lowest (8.4%) concentrations. Absorbance vs. concentration was linear for this range of standards (N = 96, R^2 = 0.997).

The calibration procedure for the 'part filter' method differed from that used for the 'whole filter' method. Since the filtration efficiencies of the 'part filters' are relatively insensitive to changes in puff flow-rate over the range typically produced by smokers (15, 16) the 'part filter' calibration was simply a linear regression for each product of nicotine yield vs. nicotine per tip and NFDPM yield vs. UV absorbance per tip over a wide range of nicotine and 'tar' yields. This was achieved by using the range of machine smoking regimes described in Table 2.

The UV absorbance and nicotine values obtained from the 'part filters' were converted into estimated NFDPM and nicotine yields by use of the regression equations.

Secondary part filter experiment

Subsequent to this initial experiment, questions arose as to whether ventilation blocking was necessary for the part filter calibration. The reason for using the fully ventilation blocked calibration routine was to extend the range of the upper end of the calibration curve. This can also be

Table 2. Details of the machine smoking regimes for the 'part filter' calibration

Brand	Puff vol/Interval (mL/s)	Ventilation holes	Length smoked
All	35/60	Open	Over tipping + 3 mm
All	35/60	Closed [a]	Over tipping + 3 mm
All	70/40	Closed [a]	Over tipping + 3 mm
All	70/60	Open	Over tipping + 3 mm
A–E	70/60	Closed [a]	4 puffs
A–E	70/60	Closed [a]	Over tipping + 3 mm
F	35/60	Open	4 puffs
F	70/60	Open	5 puffs

[a] Filter ventilation zones were covered with adhesive tape. The puff duration was 2 s for all regimes.

achieved by using a high puff volume and flow-rate, short inter-puff interval regime without ventilation blocking. To test this method of calibration, we duplicated the smoking behaviour records from another study on Products B and C along with a 10 mg ISO 'tar' yield product from the UK. The subjects were current smokers of the products tested. The records from nine subjects per product were duplicated in triplicate. Two calibration smoking methods were used with the 'part filter' method. One was identical with the calibration with ventilation blocking described above. The other calibration method did not use ventilation blocking. Puff volumes used were 40, 50 and 70 mL. For each puff volume, cigarettes were smoked to overtip plus 3 mm with puff intervals of 20, 30 (except 70 mL), and 60 sec and to 4 puffs with puff intervals of 60 sec. Filter tip analyses for nicotine were conducted within 2 weeks with no storage at −20 °C. This also addressed two further questions in the initial study regarding differences in laboratories and storage regimes. This secondary experiment was conducted in the same laboratory and used the same storage regime as the whole filter method described above.

Statistical analyses

Paired *t*-tests with the Bonferroni correction for multiple significance tests (25) were used to compare the mean values of nicotine and NFDPM produced by the duplicator with those estimated from the 'whole filter' and the 'part filter' methods. Additionally the correlations between yields produced by the duplication process and those estimated from the two filter methods were calculated for each product type using the Pearson's correlation coefficient. The statistical calculations were performed using Minitab Statistical Software Version 13.31.

RESULTS AND DISCUSSION

The ten subjects produced a wide range of puffing behaviour parameters from the six products. Puff numbers ranged from 10 to 24 puffs per cigarette, puff volumes covered a range of 29 mL to 85 mL and mean puff flow-rates ranged from 19 to 58 mL/s. This provided an extensive range of human puffing behaviour conditions for testing the accuracy of both filter methods.

The nicotine and 'tar' yields estimated using the 'whole filter' and 'part filter' methods are compared with the mainstream yield data obtained from the smoke duplicator in Figure 1 (individual points) and Table 3 (mean values by brand).

The nicotine and NFDPM yield estimates from the 'whole filter' analysis method produced good correlations with the corresponding values obtained from smoke duplication. For the regressions in Figure 1, the 95% confidence intervals (CIs) (for the intercepts were −0.20 to +0.08 mg nicotine/cig and −2.5 to +0.075 mg NFDPM/cig (neither significantly different from zero). However the slopes were significantly different from one with 95% CIs of 1.04 to 1.27 for nicotine and 1.08 to 1.25 for NFPDM. Using the (estimated – measured) difference for the individual points, the bias was +1.15 mg NFDPM/cig and 0.12 mg nicotine/cig and the root mean square (RMS) difference was 3.2 mg NFDPM/cigarette and 0.25 mg nicotine/cig. Overall the method produced mean nicotine and NFDPM values for the group of 6 products which were 10% and 8% higher than the means obtained from the duplicator. These differences were statistically significant ($p < 0.001$). The data in Table 3 demonstrate that the largest deviations from the smoke duplicator values were for products C, D and especially E.

Mean nicotine retention efficiencies and T/N ratios were used to derive the 'whole filter' data in Table 3 and Figure 1. The use of 'personal' NFE and T/N ratio values based on each subject's average puff flow-rates did not improve the relationships between estimated and duplicated smoke yields. For example, the mean estimated NFDPM and nicotine yields for all products combined were the same for both NFE approaches.

The 'part filter' method produced a close approximation to the nicotine and NFDPM yields obtained from the smoke duplicator. For the regressions in Figure 1, the 95% CIs for the intercepts were −0.05 to +1.2 mg nicotine/cig and +0.2 to +1.8 mg NFDPM/cig. The 95% CIs for the slopes were 0.93 to 1.07 mg for nicotine and 0.87 to 0.98 for NFDFM. From the (estimated – measured) difference for the individual points the bias was −0.06 mg NFDPM/cig and +0.035 mg nicotine/cig and the RMS difference was 1.8 mg NFDPM/cig and 0.16 mg nicotine/cig. The 'part filter' method over-estimated the average duplicator derived nicotine yield for the group of six products by only 3% and the corresponding value for NFDPM was under-estimated by only 0.4%. Analysing the data on a per product basis (Table 3) showed close agreements between 'part filter' and duplicator yields for most of the products. The considerable over-estimation of the yields of products C, D, and E seen with the 'whole filter' method was not present with the 'part filter' method.

An influence of changing machine smoking regimes on the NFE values of the 'whole filters' from the cigarettes used in the study was apparent from an examination of the calibration curves. The results from a highly ventilated product (B) and the least ventilated product (F) are shown in Figure 2. This Figure clearly indicates a reduction in NFE for the 'whole filter' when average puff flow-rates are increased from 17.5 mL/s to 60 mL/s especially for the highly ventilated product B.

Some researchers (9, 11, 28) have used NFEs measured under conditions which were essentially the same as the

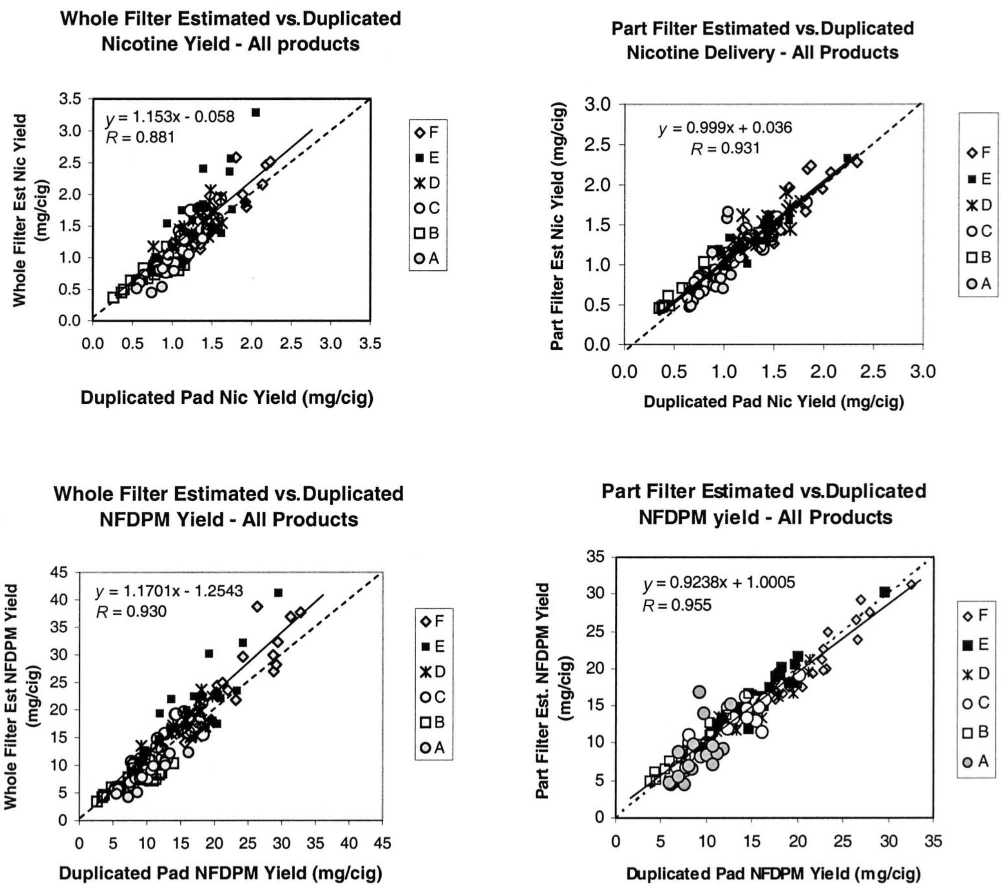

Figure 1. A comparison between estimated and smoke duplicator nicotine and NFDPM deliveries for all products (dashed line is the line of unity; solid line is the linear regression fit)

Table 3. Estimated 'tar' and nicotine yields vs. duplicated yields by product for the 'whole filter' and 'part filter' methods; values expressed as mean ± standard deviation (a,b,c = significant differences, paired *t* -test with Bonferroni correction)

	'Tar' yield (mg)			Nicotine yield (mg)		
Product	Duplicated	Estimated	*R*-value	Duplicated	Estimated	*R*-value
			'Whole filter' method			
All	14.6 ± 6.4	15.3 ± 8.1 [a]	0.93	1.16 ± 0.41	1.28 ± 0.53 [b]	0.88
A	9.4 ± 2.9	7.9 ± 2.3	0.91	0.88 ± 0.23	0.83 ± 0.24	0.85
B	8.1 ± 3.2	7.1 ± 1.9	0.87	0.75 ± 0.27	0.76 ± 0.20	0.85
C	12.8 ± 3.6	14.3 ± 3.8	0.83	1.16 ± 0.27	1.30 ± 0.34	0.85
D	15.0 ± 3.5	16.7 ± 3.7 [c]	0.84	1.27 ± 0.27	1.46 ± 0.33 [c]	0.80
E	17.4 ± 5.6	21.4 ± 7.4 [a]	0.86	1.34 ± 0.38	1.71 ± 0.59 [c]	0.78
F	22.4 ± 5.9	24.4 ± 7.5 [c]	0.91	1.57 ± 0.36	1.63 ± 0.50	0.92
			'Part filter' method			
All	13.9 ± 6.0	13.8 ± 5.8	0.93	1.15 ± 0.40	1.19 ± 0.40	0.96
A	8.8 ± 2.0	8.3 ± 3,5	0.64	0.85 ± 0.16	0.84 ± 0.32	0.69
B	8.2 ± 3.0	9.2 ± 3.1 [b]	0.98	0.74 ± 0.26	0.80 ± 0.25	0.93
C	12.6 ± 3.9	12.6 ± 3.4	0.89	1.18 ± 0.33	1.16 ± 0.32	0.95
D	15.3 ± 3.7	14.8 ± 3.2	0.94	1.27 ± 0.28	1.35 ± 0.29	0.88
E	17.1 ± 4.2	17.6 ± 4.3	0.95	1.35 ± 0.32	1.39 ± 0.32	0.93
F	22.5 ± 4.3	21.5 ± 4.7	0.94	1.59 ± 0.33	1.64 ± 0.38	0.88

[a] $p < 0.01$; [b] $p < 0.001$; [c] $p < 0.05$.

current ISO or FTC smoking regimes to convert levels of nicotine retained in filters into estimated nicotine deliveries to the smokers. The application of NFE data obtained under ISO conditions to the 'whole filter' levels of nicotine recorded in our study shows that estimated nicotine yields for the highly ventilated cigarettes (A and B) would have been around 30% lower than the actual values obtained from the duplication process. The difference was less marked for the less ventilated products, for example the estimated nicotine value for product F was only 9% lower

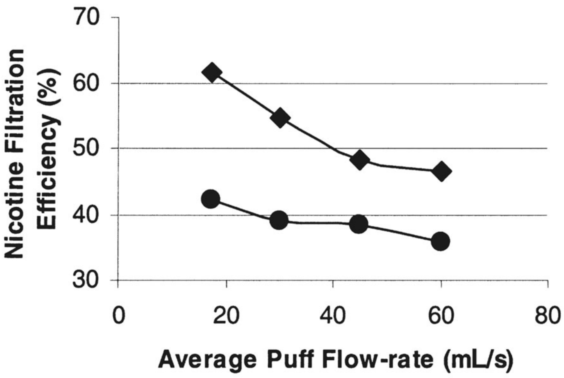

Figure 2. Effect of puff flow-rate on nicotine filtration efficiency (NFR) of product B (♦) and product F (●)

than the duplicator derived value. This can be explained by the fact that puff-flow rates obtained from the smokers were higher than the 17.5 mL/s ISO flow-rate and that the FEs of the highly ventilated cigarettes exhibited a higher degree of flow dependency than those of the less ventilated cigarettes (Figure 2).

The 'whole filter' method attempted to accommodate the influence of puff flow rate on NFE by using an average of four FEs measured over a wide range of flow-rate regimes. This produced estimated nicotine yields for the two highly ventilated cigarettes, A and B, and the least ventilated cigarette, F, which were very close to the actual yields produced by smoke duplication. Unfortunately, the estimated yields for products C, D and E were significantly higher ($p < 0.001$) than the measured yields from smoke duplication. Since the estimate of delivery is dependent on the value of NFE used, this was investigated further. The NFE values obtained during smoke duplication for products A, B and F were within approximately 2% of the calibration NFE values used to estimate nicotine delivery. The estimates for these products were close to the nicotine yields obtained from the smoke duplicator. The NFE values obtained during calibration for the remaining three products were 5% to 12% lower than the duplication values. Therefore, this would give rise to an overestimate of delivery in keeping with the difference in the observed difference in NFE i.e., the greatest overestimate was for product E.

The reason for these NFE discrepancies is unlikely to be an influence of puff-flow rates, as mean flow-rates of approximately 20 mL/s would have been required to achieve the NFE values obtained during the smoke duplication of products C, D and E. However, the smokers' mean flow-rates used in the smoke duplication for these products were all in excess of 20 mL/s.

The most likely explanation for the 'whole filter' method over-estimation of the nicotine yields for products C, D and E is nicotine condensation. As nicotine vapour self-condenses to form the smoke aerosol, it also condenses on the cooler substrate (tobacco or filter) over a 20 mm range behind the coal (26, 27). During the NFE calibration procedures the cigarettes are smoked down to a fixed butt length. However, smokers produce different butt lengths and thus the amount of nicotine condensing onto the filter may vary according to the butt length produced by the smokers and the smoking machine. Consequently, one could have a situation where two smokers may obtain similar amounts of nicotine from their cigarettes. If one smoker left a long butt and the other left a very short butt, the amount of nicotine retained in the filter of the short butt would be higher than the levels in the longer butt due to the effect of nicotine condensation. This would result in the filter analysis technique producing a higher estimated nicotine yield for the 'shorter butt' smoker than for the 'longer butt' smoker.

Comparisons between the average 'butt lengths' of the cigarettes at the end of the smoke duplication process and the cigarette filter lengths give some insight into why the condensation effect may have produced a greater influence on products C, D and E than on the other products. The differences between 'butt length' and filter length (i.e. the amount of unburnt tobacco rod) were greatest for products B and F, two products for which the duplicated NFEs and consequently the delivery estimates were as predicted. This longer tobacco section would have ensured that any nicotine condensation that could have occurred during the puff would have remained predominantly in the tobacco rod. Products C, D and E had much shorter lengths of unburnt tobacco and nicotine vapour may well have condensed into the filters of these products giving rise to artificially high filter nicotine levels for these cigarettes. This would have led to significantly higher delivery estimates than the actual deliveries from these products. The one apparent anomalous result was for product A which had a small unburnt tobacco length (similar to D and E) and might have been expected to also give rise to an overestimate due to nicotine condensation. However, this product incorporated a dual filter with a very high efficiency front section. Consequently, the amounts of nicotine retained in the filters of this product were almost twice that of all other filters in the study and hence any nicotine condensation would have been swamped by the inherent smoke nicotine present.

Confirmation of the potential influence of butt length and nicotine condensation on the 'whole filter' estimates of nicotine was found by examining the data from products E and F. The data for these two products were split into two groups according to butt length i.e. a short and a long butt length group each comprising of 10 data records. The 'whole filter' analysis for product E showed a large effect of butt length. The mean estimated nicotine yield for the shorter butt length group (mean butt length 32.2 mm) was 39% higher than the duplicator derived mean value. The longer butt length group for product E (mean length 40.0 mm) produced estimated yields that were only 13% higher than the duplicator values. Additionally the NFE value obtained from the duplicator was 55.3% for the shorter and 50.4% for the longer butt length groups. The corresponding values for product F were an average of 9% over-estimate of yields for the shorter butt length group (mean length 32.2 mm) and a less than 1% under-estimate for the longer butt length group (mean length 38.8 mm).

A similar analysis of the 'part filter' derived estimates of nicotine yield showed no effect of butt length on the differences in 'part filter' derived and duplicator measured nicotine yields for products E and F. This is not too surprising as nicotine condensation during a puff taken when the coal is in close proximity to the filter is likely to occur in the section of the filter which adjoins the tobacco rod. For most cigarettes, nicotine condensation is unlikely to occur to any great extent in the 10 mm mouth section used in the 'part filter' method. However, with some products, a nicotine condensation effect may be detectable with the 'part filter' method. Cigarettes with very low filtration efficiency filters, or dual filters with low efficiency mouth sections can fall into this category. In these cases, the amount of nicotine filtered and subsequently measured in the mouth section is inevitably very low. Any trace of nicotine condensation that reaches the 10 mm mouth section can therefore significantly influence the filter nicotine measurement and thus lead to potential overestimation of delivery. In this study, Product A was just such a cigarette being equipped with a dual filter with a very low efficiency mouth end. The most intense calibration regimes (highest flows/fully blocked ventilation/smoking to overtip + 3 mm) resulted in an apparent increase in filtration efficiency. It was concluded that this was due to low levels of nicotine condensation into the mouth section. Since the higher flow-rate, full ventilation blocked regimes are unlikely to be adopted by smokers because maximum, deliberate blocking of the ventilation holes with lips or fingers results in a blockage of only around half of the ventilation holes (29, 30) and blocking ventilation holes is associated with a reduction in puff flow-rate (31, 32), these regimes were removed from the calibration data for this product.

For the secondary part filter experiment, slopes and intercepts for the two methods of calibration were not significantly different from each other at the 95% confidence level. Using Analysis of Variance, the two calibration methods predicted yields that were not significantly different from each other or from the actual cigarette yields. The RMS difference between estimated and measured nicotine was 0.20 mg/cig for the calibration with ventilation holes blocked and 0.27 mg/cig for the calibration without ventilation holes blocked. This means that either calibration method could be used. However, in certain circumstances, calibration without ventilation blocking may be more appropriate. For example this avoids the condensation issues with low efficiency mouth sections noted above. In addition, this second experiment showed that tips stored for 2 weeks at room temperature or 4 to 5 months at −20 °C gave equivalent predictions of actual cigarette yields.

Although our study identified some problems with the use of the 'whole filter' method, the results indicated that the 'whole filter' method has the ability to provide a reasonable indication of the yields of 'tar' and nicotine obtained by smokers. However, the data clearly demonstrated that the 'part filter' method produced a more accurate estimation of these yields; thus, we conclude that the 'part filter' method rather than the 'whole filter' method should be used for yield estimation studies in smokers.

Recently, WATSON *et al.* (14) published a method to assess cigarette smoke intake. This was based on the analysis of solanesol retained in filters after smoking. Whilst WATSON *et al.* (14) demonstrated that solanesol retained in filters could be used to estimate 'tar' and nicotine yields, they identified an 'intensive' machine smoking condition i.e., fully blocked filter ventilation holes, where the relationships between the amounts of solanesol retained in the filter and the amounts of 'tar' or nicotine delivered exhibited marked deviations from a linear response. This is consistent with an influence of puff flow-rate on filtration efficiencies. The cigarettes used for the ventilation block condition in the WATSON *et al.* (14) study were highly ventilated 1 mg 'tar' yield cigarettes. Although puff-flow rates were not changed in the study, blocking the filter ventilation zone would have resulted in an increase in the flow-rate through the filter section upstream of the ventilation zone. This flow-rate increase would have caused a reduction in the filtration efficiency of the 'upstream' section and thus the amounts of solanesol retained in the filter. Consequently, we believe that the solanesol method described by WATSON *et al.* (14) could be improved by the analysis and calibration of only the 'mouth section' of the filter rather than using the whole filter.

Our results demonstrate that the filter analysis methods can provide a useful and simple means of measuring 'tar' and nicotine yields obtained from cigarettes by smokers in their normal smoking environments. The 'part filter' method gave more accurate estimates of smoke yields than the 'whole filter' because the former was less susceptible to problems caused by smoker behaviour factors such as differences in puff flow rates and cigarette butt lengths. Data obtained from filter analysis could provide a useful means of assessing the relationships between ISO/FTC yields and those obtained by smokers and thus assist in determining the utility of the current ISO/FTC methods and alternative machine based techniques for measuring smoker-relevant smoke yields.

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