# Migration and Delivery of Filter Flavors\*

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# INTRODUCTION

The current trend toward low tar delivery cigarettes has resulted in a concurrent decrease in the taste experienced by the smoker. Cigarette manufacturers are faced, therefore, with the problem of delivering adequate taste while maintaining low levels of tar and nicotine. There are several ways to increase cigarette flavor. Blending different tobacco types for increased flavor and adding flavorants to the tobacco are two techniques which are commonly used (1). However, these methods possess the shortcoming that flavoring associated with the tobacco is altered and reduced as it traverses the filter segment (2). Another technique for enhancing cigarette taste involves incorporating flavor additives into the filter, where they can be eluted into the smoke aerosol as it passes through the filter. Relatively high flavor delivery efficiencies can frequently be achieved in this manner despite low tar deliveries. Although this flavoring technique is currently recognized by cigarette manufacturers (3), very little information describing the behavior of filter flavors exists. This study addresses several fundamental questions relating to interactions between flavorant and filter and how such interactions ultimately affect the efficiency with which a filter flavor is delivered to the smoker.

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### EXPERIMENTAL

Experimental filter cigarettes were prepared by using 65 mm blended tobacco rods and 20 mm flavored filters. Filter tips were fabricated from 2.9 denier/fil., 41,000 total denier, Y cross section filter tow. Ten model flavor compounds were applied to the tow as a 10 mg/ml solution of triacetin with a brush applicator. Finished 20 mm tips weighed 148.5  $\pm$  1.3 mg each and contained 10% triacetin by weight and 0.134 mg of each flavor. This flavor concentration was chosen to facilitate the analytical measurement and because it was about a factor of ten less than the menthol concentration in mentholated brands. However, because flavoring formulations are a closely guarded secret, it was not known if this level accurately simulated existing commercial cigarettes. The following compounds obtained from Eastman Organic Chemicals and Pfaltz and Bauer, Inc., were used as model flavors: cinnamyl isovalerate, cinnamyl propionate, 4-ethoxyacetophenone, eugenol, isoamyl benzoate, isoamyl cinnamate, isoamyl isovalerate, isoamyl phenylacetate, methyl benzoate, and methyl cinnamate.

Test cigarettes with an average wet TPM\* delivery of 21.4 mg were all prepared within 24 hours of filter manufacture by manually attaching the flavored filter tips to conditioned 65 mm blended tobacco columns with tipping paper and polyvinyl alcohol adhesive. Cigarettes

<sup>\*</sup> Presented at the 33rd Tobacco Chemists' Research Conference held in Lexington, Kentucky, in 1979.

<sup>\*</sup> total particulate matter

were stored in sealed glass jars and sampled initially and 1, 2, 4, 6, and 8 weeks after preparation for the determination of migration and delivery levels. Migration was determined by separating the filter tips from the tobacco rods and extracting and analyzing each for its flavor content. Flavor delivery was determined by analyzing the flavor content of an extract of total particulate matter collected on Cambridge filter pads during smoking. A series of ventilated cigarettes were also prepared at 25% and 50% ventilation, and their flavor delivery was measured as a function of time.

The quantitative measurement of model flavor compounds was achieved by extracting filters, tobacco columns, or Cambridge pads with a solution of absolute ethanol containing 0.020 mg/ml decylaldehyde obtained from Aldrich as an internal standard. Complete extraction was demonstrated by spiking samples with a known amount of flavored triacetin and then analyzing the samples after 24 hours. Analysis of flavors present in the extracts was performed by gas chromatography on a temperature programmed 30 meter soft glass WCOT (wall coated open tubular) OV101 capillary column obtained from J and W Chromatographic Supplies\*. A Carlo Erba Fractovap series 2900 gas chromatograph equipped with flame ionization detector (FID) was used in the splitless injection mode for the analysis. Peak height ratios with the internal standard were used for the measurement based on a seven-point calibration curve determined for each model compound. Each extract was chromatographed twice and represented a group of five experimental cigarettes. Four such groups of five cigarettes were measured to obtain each migration and delivery data point. Interferences to the analysis were not a problem except in the case of the tobacco column extract where sample matrix effects made it impossible to determine the migration of four of the model flavors. Part of the problem encountered resulted from the fact that the migration of these compounds was so low that only a small amount of each compound was present in the tobacco column extract and was, therefore, highly subject to interference.

\* J and W Scientific Inc.

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Flavor	Migration (percentage of total)							
	week 1	week 2	week 4	week 6	week 8			
Isoamyl isovalerate	67	67	66	65	64			
Methyl benzoate	20	24	28	28	31			
Methyl cinnamate	10	15	18	23	22			
4-Ethoxyacetophenone	6.1	7.9	9.9	12	14			
Cinnamyl propionate	4.7	5.8	6.0	8.7	8.1			
Cinnamyl isovalerate	3.8	7.8	9.2	19	16			

Table :	2.	Flavor	migration	to	tobacco	V\$.	time
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#### Table 3. Kinetics of filter flavor migration.

Flavor	Y <sup>0.01</sup>	P <sup>0</sup> <sub>25</sub> (torr)	γ <sup>0.01</sup> <sub>25</sub> p <sup>0</sup> <sub>25</sub> (torr)	k (week <sup>-1</sup> )	t <sub>½</sub> (week)	М <sub>ЕQ</sub> (%)
Isoamyl isovalerate	6.72	0. <del>9</del>	6.05	≥ 8.98	≤ 0.077	66 ± 2
Methyl benzoate	1.05	0.4	0.42	1.18 ± 0.15	0.59	28 ± 2
Methyl cinnamate	0.79	0.26	0.021	0.60 ± 0.27	1.2	21 ± 2
4-Ethoxyacetophenone	1.39	0.004	0.0056	0.51 ± 0.38	1.4	13 ± 2
Cinnamyl propionate	0.97	0.002	0.0019	0.68 ± 0.40	1.0	8 ± 1
Cinnamyl isovalerate	1.36	0.001	0.0014	0.33 ± 0.33	2.1	16 ± 5
Menthol C	10.4	0.1	1.04	1.20 ± 0.10	0.58	60 ± 1

### **RESULTS AND DISCUSSION**

### Efficiency of Flavor Application

Table 1 lists the percentages of each flavor in freshly fabricated filter tips relative to the amount of flavor in the plasticizer. A flavor in plasticizer solution which is quantitatively applied by the brush applicator has an application efficiency of 100 %. However, Table 1 shows that significant losses have occurred for isoamyl isovalerate and methyl benzoate. These two compounds are more volatile than the other additives and are more readily vaporized from the high surface area mist of plasticizer droplets generated by the brush applicator. The other less volatile compounds are not lost in this manner and are applied in virtually quantitative amounts. In general, it appears that compounds which boil below 250 °C are not quantitatively applied by a brush applicator because of evaporation from the plasticizer mist.

# Migration of Filter Flavors

Table 2 shows the percentage of the total amount of each flavor compound applied to the filter which migrated to the tobacco column as a function of cigarette age. Migration is not reported for four of the flavors because low migration and interferences prohibited their measurement. For those compounds measured, it was observed that the flavor originally placed on the filter was quantitatively recovered from the tobacco and filter. Table 2 shows that migration takes place most rapidly during the first week after the test cigarettes are fabricated and that the rate of migration decreases with increasing cigarette age. After some time, equilibrium migration is achieved and no further migration takes place. The rate at which equilibrium is achieved and its final value vary from flavor to flavor and, therefore, must be determined largely by the properties of the individual flavor compounds.

The observed migration appeared to follow an exponential time dependence in which the physical limitations of

Table 1. Efficiency of brush applicator for filter flavors.

Flavor	Boiling- point (°C)	Application efficiency (%)
Isoamyl isovalerate	193	51 ± 3
Methyl benzoate	200	81 ± 2
Eugenol	253	100 ± 6
Methyl cinnamate	254	88 ± 3
Isoamyi benzoate	262	92 ± 2
Isoamyl phenylacetate	268	92 ± 3
4-Ethoxyacetophenone	269	94 ± 3
Cinnamyl propionate	289	94 ± 3
Isoamyl cinnamate	298	105 ±12
Cinnamyl isovalerate	313	95 ± 7
Average of last 8		95 ± 5

no migration at week zero and attainment of equilibrium migration after some period of time are obeyed. The following model was developed to describe flavor migration:

$$M(t) = M_{EQ} \times (1 - e^{-kt})$$

where M(t) is the migration at time t,  $M_{EO}$  is the migration at equilibrium, e is the natural base, and k is a first-order rate constant with units of reciprocal time. The model was fitted to the experimental data by using a non-linear least squares regression analysis, and the predicted time dependence is shown in Figure 1 as solid lines through the experimental data points. Figure 1 also shows data for menthol migration from an earlier study (4). The excellent agreement between the experimental data and the theoretical model supports the model's validity. Table 3 lists the best fit values of k and MEO determined by using the model for the six flavors for which migration could be measured and for menthol (4). Values of t<sub>1/2</sub>, which is the time required for migration to attain half its equilibrium value, are also given, since  $t_{1/2}$  is physically more meaningful than k.

Values for the flavor migration rate constant were proportional to the vapor pressures of the pure flavor compounds, and an excellent correlation of 0.9994 was calculated between the rate constant and flavor volatility in triacetin solution. Volatility is defined as the vapor pressure of the pure compound times an activity coefficient which corrects for non-ideal solution behavior such as thermodynamic solubility and concentration effects. Values of activity coefficients were determined at 25 °C for a solute mole fraction of 0.01 by using the Universal Quasi-Chemical Functional Group Activity Coefficients (UNIFAC) method (5). The relationship

Figure 1. Migration of flavors.



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discovered between k and flavor volatility is described by the following equation:

$$k = 0.53 + 1.4 \gamma_{25}^{0.01} p_{25}^{0}$$

where  $\gamma_{25}^{0.01}$  is the activity coefficient and  $p_{25}^0$  is the vapor pressure of the pure compound. The data for menthol migration were not included in the analysis because filter menthol levels used to obtain these data were many times higher than the filter flavor levels used in this study, and a plasticizer other than pure triacetin was used.

Although a strong correlation was observed between the flavor migration rate constant and flavor volatility, it should be noted that because of the distribution of the data, this correlation is influenced by the extreme behavior of isoamyl isovalerate. More data for flavors of intermediate volatility are required to characterize the exact dependence of migration kinetics on flavor volatility, but the observed correlation does indicate that these quantities vary proportionally. This result implies that migration occurs by a vapor phase mechanism in which compounds in the filter evaporate and diffuse to the tobacco, where they condense. Because the volatility of a flavor is determined not only by its properties but also by those of the medium it is dissolved in, it should be possible to make limited changes in flavor migration kinetics by altering the filter material or plasticizer blend. Conversely, any factor or filter property which does not influence flavor volatility should not affect the rate of flavor migration. A vapor phase diffusion model cannot predict the equilibrium migration because this quantity is probably determined by the relative affinity of the flavor for the tobacco versus the plasticized filter.

# Delivery of Filter Flavors

The percentages of the total amount of each model compound delivered in the Cambridge pad particulates during smoking are listed in Table 4 versus cigarette age. Values for eugenol are not reported because of interferences in the smoke condensate. For each flavor studied, with the exception of methyl benzoate, delivery was most efficient at week zero and decreased with

# Table 5.Comparison of migration and delivery efficiencyloss at week 1.

Flavor	Migration (percentage of total)	Delivery efficiency loss (%)*
Isoamyl isovalerate	67	50
Methyl benzoate	20	35
Methyl cinnamate	10	61
4-Ethoxyacetophenone	6.1	52
Cinnamyl propionate	4.7	44
Cinnamyl isovalerate	3.8	16

\* Relative to delivery efficiency at week 0.

	Delivery (percentage of total)						
Flavor	week 0	week 1	week 2	week 4	week 6	week 8	
Isoamyl isovalerate	40 ± 5	20	20	21	22	17	
Methyl benzoate	11 ± 2	7.2	10	11	12	13	
Methyl cinnamate	33 ± 2	13	13	12	14	13	
Triacetin*	9.1 ± 0.6	8.5	7.7	6.8	7.0	7.6	
isoamyi benzoate	55 ± 5	47	42	36	34	35	
Isoamyl phenylacetate	51 ± 5	40	36	27	26	26	
4-Ethoxyacetophenone	23 ± 2	. 11	8.7	7.4	8.2	10	
Cinnamyl propionate	27 ± 3	15	12	9.5	9.6	9.9	
Isoamyl cinnamate	29 ± 6	21	20	13	14	21	
Cinnamyl isovalerate	31 ± 5	26	23	16	16	18	

Table 4. Delivery of filter flavors.

\* Present in filter as plasticizer at a concentration of 14.9 mg/20 mm tip.

	Table	6.	Kinetics	of	delivery	efficiency	/ loss
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Flavor	k' (week <sup>-1</sup> )	t <sub>%</sub> (week)	D₀ (%)	D <sub>EQ</sub> (%)
4-Ethoxyacetophenone	1.79 ± 1.6	0.56	14.5	8.5
Cinnamyl propionate	1.13 ± 0.3	0.89	17.3	9.7
Isoamyl benzoate	0.49 ± 0.2	2.0	21.5	33.7
Isoamyl cinnamate	0.53 ± 1.0	1.9	16.2	12.6
Cinnamyl isovalerate	0.49 ± 0.5	2.0	15.5	16.0
Isoamyl phenylacetàte	0.50 ± 0.25	2.0	26.1	24.8

increasing time. Decreases in delivery efficiency were always largest during the first week and usually reached a constant value within experimental variance after about a month.

The observed delivery efficiency decreases may, in part, be attributable to flavor migration to the tobacco column and subsequent losses to sidestream smoke and pyrolysis. However, Table 5 shows that for the six compounds for which migration could be measured, migration does not account for the observed delivery efficiency loss. This is true even in the unlikely case that no flavor is delivered into the smoke from the



tobacco column. For every flavor studied, except isoamyl isovalerate, the delivery efficiency loss after one week was much greater than the observed migration. In some cases, even though 95% of the flavor remained on the filter, about 50% of the delivery efficiency was lost. This is strong evidence that some process in the filter is acting to make the flavor compounds less accessible for elution into the smoke aerosol.

To elucidate the mechanistic aspects of this process, the dependence of delivery efficiency loss was examined as it varied with the properties of the model flavor compounds. First-order rate constants for the kinetics of delivery efficiency loss were calculated by using nonlinear least squares regression analysis to fit an empirical model to the experimental data. The model used for this calculation was:

$$D(t) = D_0 e^{-k't} + D_{EQ}$$

where D(t) is the percentage flavor delivered at time t, Do is the change in percentage delivery between time zero and equilibrium, D<sub>EQ</sub> is the percentage delivery at equilibrium, e is the natural base, and k' is the rate constant for delivery efficiency loss. The validity of this model for estimating rate constants is demonstrated in Figure 2, in which the solid lines represent values of delivery efficiency predicted by the model from a fit to the displayed data points. In order to minimize the effects of flavor migration on the analysis of delivery efficiency loss, only the high boiling flavors with very low vapor pressure and migration were analyzed. Calculated values of k', D<sub>EQ</sub> and D<sub>0</sub> for these compounds are listed in Table 6. Values of  $t_{1/2}$ , the time required for half of the total delivery efficiency loss to occur, have again been reported to better demonstrate the time scale of this phenomenon.

The values calculated for k', the rate constant for delivery efficiency loss, were found to have a 0.96 correlation with the activity coefficients of the flavor compounds according to the following equation:

$$k' = 4.0 - 2.3 \gamma_{25}^{0.01}$$

The most important property which determines a compound's activity coefficient is its solubility. Flavors with an activity coefficient equal to one have high thermodynamic solubility in triacetin, whereas those with large activity coefficients have progressively lower solubilities. This empirical correlation, therefore, indicates that solubility is an important factor controlling the rate at which compounds placed on a filter in plasticizer solution become inaccessible for elution and that highly soluble flavors lose delivery efficiency most rapidly. Although no attempt has been made to rationalize absolute delivery efficiencies, such solubility considerations may be the cause of the low delivery observed for methyl benzoate since this flavor has the highest solubility in triacetin.

These observations support a plausible hypothesis for the delivery efficiency loss phenomenon. It is known

Table 7. Effect of ventilation on flavor delivery at 25 % di
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	Ratio of vented vs. non-vented delivery						
	week 0	week 1	week 2	week 4	week 6	week 8	
Isoamyl isovalerate	0.46		0.45	0.52	0.92	0.97	
Methyl benzoate	0.82	0.86	0.74	0.90	1.08	0.85	
Triacetin	0.89	0.79	0.81	<sup>.</sup> 0.62	0.66	0.60	
Eugenol	0.89	1.06	0.92	0.82	0.88	0.88	
Methyl cinnamate	0.86	0.91	0.85	0.81	1.00	0.86	
Isoamyl benzoate	0.92	0.94	0.81	0.57	0.67	0.58	
Isoamyl phenylacetate	0.90	0.92	0.82	0.57	0.76	0.65	
4-Ethoxyacetophenone	0.91	0.80	0.80	0.68	0.86	0.81	
Cinnamyl propionate	0.82	1.20	0.88	0.71	0.97	0.71	
Isoamyl cinnamate	0.90	0.77	0.71	0.85	1.00	0.49	
Cinnamyl isovalerate	0.89	0.76	0.76	0.66	0.92	0.51	
Average last nine	0.89	0.91	0.82	0.70	0.86	0.68	
	± 0.03	± 0.15	± 0.06	± 0.11	± 0.13	± 0.15	

Table 8. Effect of ventilation on flavor delivery at 50 % dilution.

	Ratio of vented vs. non-vented delivery							
Flavor	week 0	week 1	week 2	week 4	week 6	week 8		
Isoamyl isovalerate	0.57	0.61	0.47	0.50	0.62	0.51		
Methyl benzoate	0.33	0.40	0.35	0.33	0.45	0.40		
Triacetin	0.62	0.53	0.55	0.49	0.46	0.36		
Eugenol	0.65	0.70	0.62	0.58	0.47	0.43		
Methyl cinnamate	0.63	0.58	0.52	0.52	0.50	0.48		
Isoamyl benzoate	0.68	0.64	0.59	0.46	0.42	0.34		
Isoamyi phenylacetate	0.72	0.55	0.60	0.38	0.53	0.38		
4-Ethoxyacetophenone	0.64	0.47	0.48	0.58	0.50	0.44		
Cinnamyl propionate	0.63	0.46	0.62	0.55	0.81	0.47		
Isoamyl cinnamate	0.68	0.46	0.47	0.52	0.58	0.30		
Cinnamyl isovalerate	0.67	0.42	0.44	0.52	0.71	0.31		
Average last nine	0.66	0.53	0.54	0.51	0.55	0.39		
	± 0.03	± 0.09	± 0.07	± 0.06	± 0.13	± 0.07		

that after fabrication, several changes occur in the physical properties of cellulose acetate filters. These changes have been attributed to diffusion of triacetin into the filter fibers and include a lowered equilibrium water content and increased fiber density. These changes are most evident during the first two weeks after filter fabrication, which is about the same time interval as observed for the delivery efficiency loss. Since the flavors were applied to the filters in triacetin solution, it is reasonable to assume that they will also diffuse into the filter fibers. At week zero, the freshly applied triacetin and flavors are near the surface of the fibers and are easily eluted. As the filter ages, the plasticizer and flavors diffuse into the fibers and no longer readily elute into the smoke aerosol. Flavors which are very soluble in the plasticized cellulose acetate readily penetrate the filter fibers and lose delivery efficiency because they must diffuse to the fiber surface to be eluted. However, flavors with lower solubilities diffuse into the fibers more slowly and to a lesser extent and, therefore, exhibit a slower rate of delivery efficiency loss.

# Effect of Filter Ventilation on Flavor Delivery

Tables 7 and 8 list the ratios of each flavor delivered by both ventilated and non-ventilated cigarettes as a function of age for test samples hand-perforated at 25% and 50% ventilation. Expressing flavor delivery as a ratio relative to the delivery of non-ventilated cigarette minimizes the effects of migration and diffusion into

Table 9. Effect of time and ventilation on average flavor delivery.

		R	atio of delivery (ve	nted vs. non-ven	ted)	
Time (weeks)		25 % dilution		50 % dilution		
	flavor	triacetin	wet TPM	flavor	triacetin	wet TPM
0	0.89 ± 0.03	0.89 ± 0.09	0.81*	0.66 ± 0.03	0.62 ± 0.09	0.56**
1	0.91 ± 0.15	0.79 ± 0.18	0.83	0.53 ± 0.09	0.53 ± 0.12	0.55
2	0.82 ± 0.06	0.81 ± 0.09	0.76	0.54 ± 0.07	0.55 ± 0.11	0.54
4	0.70 ± 0.11	0.62 ± 0.03	0.76	0.51 ± 0.06	0.49 ± 0.04	0.53
<b>6</b>	0.86 ± 0.13	0.66 ± 0.06	0.84	0.55 ± 0.13	0.46 ± 0.09	0.55
8	0.68 ± 0.15	0.59 ± 0.09	0.88	0.39 ± 0.07	0.36 ± 0.03	0.57

\* dry TPM ratio equals 0.84.

\*\* dry TPM ratio equals 0.63.

filter fibers and exaggerates the influence of ventilation on the results. If the two volatile compounds tested, isoamyl isovalerate and methyl benzoate, are disregarded, it appears that ventilation has a relatively uniform effect on delivery, and an average delivery ratio can be calculated for a ventilated cigarette as a function of time. These values, as well as delivery ratio values for triacetin and wet TPM, are summarized in Table 9. Average dry TPM delivery ratios are stated because this quantity was a time independent constant.

Although the data have significant variability, it is apparent from Table 9 that ventilated cigarettes deliver filter flavors more efficiently than TPM during the period immediately following their fabrication. However, the relative efficiency of flavor elution decreases with time such that, after two or three weeks, filter flavors are delivered less efficiently than TPM relative to the nonvented cigarette. The flavor delivery ratio should not be affected by flavor diffusion into the filter fibers, but the time dependence of these data strongly indicates that this mechanism is responsible for the observed delivery efficiency loss. Addition of ventilation must, therefore, exaggerate the effect of flavor diffusion into filter fibers on flavor elution relative to a non-vented cigarette. A possible, but untested, explanation is that the lower smoke velocity in the upstream of the ventilated filter elutes flavors more efficiently when they are near the surface of the filter fibers. However, as the flavors migrate into the fibers, diffusion to the fiber surface becomes rate limiting, and this relative advantage is lost.

### SUMMARY

The migration and delivery of filter flavor agents were studied by dissolving 10 model flavor compounds in triacetin at 10 mg/ml each and fabricating cigarettes with flavored filter tips. The concentrations of these compounds in filters, tobacco, and smoke particulates collected on Cambridge filters were determined by capillary column gas chromatography initially and after 1, 2, 4, 6, and 8 weeks. Flavor migration was found to obey firstorder kinetics, with the rate constant being determined by the volatility of the flavor compound dissolved in the plasticized filter. Delivery of flavor compounds decreased with time at a greater rate than could be explained by flavor migration. A high correlation between the solubility characteristics of the flavor and its rate of delivery efficiency loss lead to the proposed explanation that flavor compounds diffuse into the cellulose acetate fibers with time and become less accessible for elution into the smoke aerosol. Model compounds with poor solubilities in plasticized cellulose acetate remain near the surface of the fibers where they are readily eluted into the mainstream smoke and, consequently, exhibit smaller delivery efficiency losses with time. Ventilation was observed to initially increase the ratio of flavor to TPM delivery, but this advantage was lost as the cigarette aged.

# ZUSAMMENFASSUNG

Die Autoren untersuchten das Migrationsverhalten von dem Filter zugesetzten Aromastoffen und deren Übergang in den Rauch, indem sie Versuchszigaretten herstellten, deren Filtermundstück mit zehn Modellsubstanzen, jeweils in Triacetin gelöst (10 mg/ml), behandelt worden war. Im Filter, im Tabak und in dem in Cambridge-Filtern aufgefangenen Rauchkondensat wurde die Konzentration dieser Verbindungen gleich nach der Anfertigung der Zigaretten sowie 1, 2, 4, 6 und 8 Wochen danach unter Einsatz von Kapillarsäulengaschromatographie gemessen. Die Migration gehorcht einer Kinetik 1. Ordnung, wobei die Geschwindigkeitskonstante von der Flüchtigkeit der Aromasubstanz bestimmt wird, die sich in dem mit einem Weichmacher versetzten Filter auflöst. Die Rauchausbeute der Aromaverbindungen nahm mit der Zeit schneller ab, als es durch die Substanzwanderung erklärt werden könnte. Die hohe Korrelation zwischen der Löslichkeit des Aromastoffes und der Verringerung seiner Ausbeute im Rauch legt die Vermutung nahe, daß Aromaverbindungen mit der Zeit in die Celluloseacetatfaser diffundieren und für den Übergang in das Rauchaerosol weniger zugänglich werden. Modellverbindungen mit geringer Löslichkeit in Weichmacher enthaltendem Celluloseacetat bleiben nahe der Faseroberfläche, von wo sie leicht in den Hauptstromrauch eluiert werden und folglich einen geringeren Ausbeuteverlust über die Zeit erfahren. Durch Ventilation erfolgte zunächst eine Erhöhung des Verhältnisses Aromastoffausbeute zu Kondensatausbeute. Dieser Vorteil ging aber mit der Alterung der Zigaretten verloren.

### RÉSUMÉ

L'étude porte sur la migration et le passage en fumée d'aromatisants contenus dans le filtre. A cet effet des cigarettes-tests ont été fabriquées et dotées de filtres aromatisés au moyen de dix substances respectivement dissoutes dans de la triacétine (10 mg/ml). La concentration de ces composés a été mesurée au sein du filtre, du tabac et du condensat recueilli sur des filtres Cambridge et cela, juste après la fabrication de la cigarette, puis après 1, 2, 4, 6 et 8 semaines, en recourant à une chromatographie gazeuse sur colonne capillaire. La migration obéit à une cinétique du premier ordre, la constante de vitesse étant déterminée par la volatilité de l'aromatisant qui se dissout dans un filtre traité au moyen d'un plastifiant. Le rendement en fumée des composés aromatisants diminue davantage avec le temps que ce que la migration permet d'expliquer. L'importante corrélation constatée entre la solubilité de l'aromatisant et la baisse de son rendement en fumée donne à penser que les composés aromatisants se diffusent peu à peu dans les fibres d'acétocellulose, devenant ainsi moins accessibles à une élution dans l'aérosol de fumée. Des composés-tests présentant une plus faible solubilité dans l'acétocellulose du plastifiant, demeurent près de la surface des fibres, d'où ils sont facilement élués dans la fumée du courant principal, ce qui entraîne à la longue une plus faible perte de rendement en fumée. Du fait de la ventilation, on a d'abord constaté une augmentation du rapport rendement en aromatisants / rendement en condensat. Toutefois cet avantage disparait au fur et à mesure du vieillissement de la cigarette.

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