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The Characterization of Cigarette Smoke from Cytrel® Smoking Products[+] and its Comparison to Smoke from Flue-Cured Tobacco

I. Vapor Phase Analysis*

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INTRODUCTION

The composition of cigarettes has changed considerably over the past twenty years. Partly because of economics, the utilization of "blown tobaccos", cut-rolled stems, and sheet reconstituted tobacco has become an accepted practice in many countries. Also, where permitted by law, the use of non-tobacco ingredients such as humectants, flavorings and casings, and gum binders for reconstituted tobacco is common. The latest innovation to be introduced into cigarette manufacture is the development of tobacco supplements. Ideally these supplements should add nothing new to the smoke produced by all-tobacco cigarettes and in fact they should reduce where possible those compounds known or suspected to irritate the respiratory tract or display other biological activity.

The purpose of this article, the first of a series, is to begin to describe the composition of smoke from Cytrel smoking products**. In these articles the three major fractions of cigarette smoke — vapor phase, semi-volatile phase, and particulate phase — will be dealt with individually. The description of these three phases will include the determination of more than 250 compounds and 67 elements in mainstream smoke from tobacco and Cytrel-containing cigarettes and account for more than 90 % of Cytrel mainstream smoke. This first paper describes the determination of more than 50 compounds in that fraction of cigarette smoke which is generally classified as vapor phase plus the determination of hydrogen cyanide, ammonia and certain amines which have been measured in whole smoke.

EXPERIMENTAL

Cigarette Manufacture

Cytrel was incorporated at three different levels (10, 20, and 50 %) into a blend of flue-cured tobaccos typical of that found in commercial U. K. cigarettes. Filter-tipped cigarettes were made from these three Cytrel-tobacco blends as well as the 100 % materials according to the specifications shown in Table 1.

Smoking Procedure

The cigarettes for these tests were conditioned and smoked according to test procedures established by the United States Federal Trade Commission and commonly practised by the U.S. tobacco industry. All cigarettes were conditioned at least 48 hours at 74 ± 2 °F and 60 ± 2 % relative humidity before they were weight-selected and smoked to a 23 mm butt length (filter tipping paper plus 5 mm) according to the internationally recognized standards of one 35 ml puff of 2 seconds' duration, once per minute. A piston-action machine (1) was used for smoking where the analyses required either liquid-filled or solid adsorbent gas-scrubbing traps. An alternate restricted smoking machine, designed by *Keith* and *Newsome* (2, 3), was used for the remainder of the tests. A listing of the analyses done with each smoking machine is shown below. Only one style of smoking machine was used in any specific analysis; 100 % Cytrel, Cytrel-tobacco blends, and 100 % tobacco cigarettes were smoked identically for any given test.

Piston-action smoking machine:	*Keith-Newsome smoking machine*
Hydrogen cyanide	Hydrocarbons
Nitric oxide	Aldehydes and ketones
Ammonia	Furans
Primary amines	Nitriles
Hydrogen sulfide	Carbon monoxide
Sulfur dioxide	Carbon dioxide

Analytical Procedures

Organic Vapor Phase: Compounds determined by this procedure were the volatile hydrocarbons, aldehydes and ketones, furans, and nitriles. The smoke sampling device used in this analysis has been described in detail (3, 4). Six cigarettes were inserted into the smoking machine and a timer was started. The individual cigarettes were then ignited, one per puff, on puffs one

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** Cytrel smoking product Type 361, Lot 227, was the specific variant used in these studies.

[+] Cytrel® is a registered trademark of Celanese Corporation.

Table 1. Physical properties of cigarettes containing blends of tobacco with Cytrel.

	Sample (nominal Cytrel concentration)				
	100 % tobacco	10 % Cytrel	20 % Cytrel	50 % Cytrel	100 % Cytrel
1. Cigarette characteristics:					
Length (mm)	72	72	72	72	72
Weight (g)	0.98	0.98	0.98	0.98	0.98
Circumference (mm)	25.0	24.9	25.0	25.1	25.0
Pressure drop (mm H_2O)	116	118	118	107	90
Puff count	10.2	9.2	8.5	7.0	5.8
Actual % Cytrel in blend	0	10	17	51	100
2. Filter characteristics:					
Composition:					
acetate (mm)	6	6	6	6	6
paper (mm)	10	10	10	10	10
Total length (mm)	16	16	16	16	16
Weight (g)	0.1801	0.1807	0.1790	0.1798	0.1784
Circumference (mm)	25.0	24.9	25.0	25.1	25.0
Pressure drop (mm H_2O)	61	59	62	60	60
Smoke removal efficiency	52.4	55.1	54.2	55.0	57.0
Nicotine removal efficiency	47.3	46.5	47.5	50.0	—*
Tar removal efficiency	48.1	49.3	48.7	49.1	49.0

* Nicotine is absent in this variant.

through six. The smoke was puffed into an evacuated 210 ml sample volume and after a short time delay, the sample volume was re-evacuated to make ready for the next puff. The sample was injected into a gas chromatograph (GC) on puff number eight and was a mixture of smoke from six cigarettes of different butt lengths.

For cigarettes whose puff counts precluded their being smoked to puff number 8, the cigarettes were lit sequentially as before, but the injection was made into the gas chromatograph before the butt length limit was violated and no sample was injected on a "lighting puff". If, for example, only 5 cigarettes could be lit before injection, then the smoke sample was diluted by one puff out of 6, and the measured deliveries were multiplied by 6/5 in order to obtain the true delivery of the "average puff". Separation in the GC was accomplished on an 18 ft. × 1/16 in. stainless steel column plus a one-foot precolumn, each filled with 100/120 mesh Porapak Q. After a six-minute hold, the column was heated from ambient temperature to 240 °C at 3°/minute; the separated components were detected with a flame ionization detector and measured with a Perkin-Elmer PEP-2 GC data system. The "average puff" deliveries obtained in this analysis were then multiplied by the cigarette's puff count to obtain the delivery per cigarette. Organic vapor phase chromatograms typical of Cytrel and tobacco smoke are shown in Figure 1.

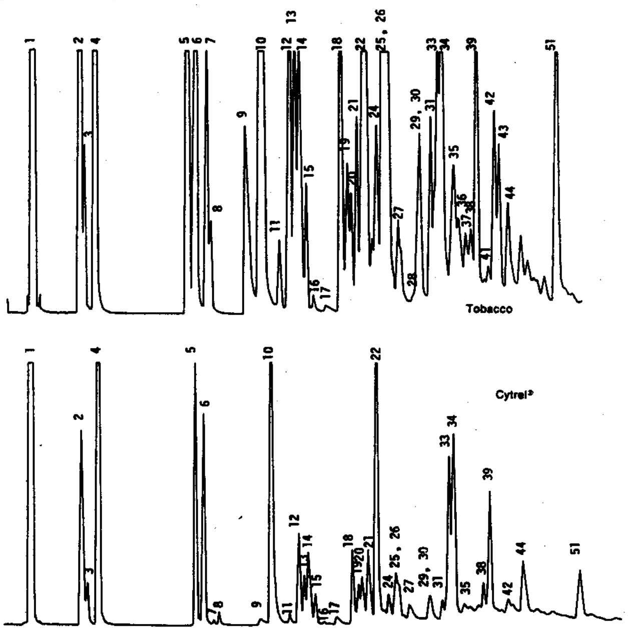
Carbon Monoxide/Carbon Dioxide: Carbon monoxide and carbon dioxide were determined by a gas chromatographic procedure utilizing a novel reactive column (5). This column, consisting of three sections, separates carbon dioxide from air and carbon monoxide in the first zone, oxidizes carbon monoxide to carbon dioxide in the second section, and then further separates both carbon dioxide peaks from air in the third zone. For both convenience and sampling uniformity, the carbon monoxide/carbon dioxide and organic vapor phase determinations were performed simultaneously, employing the same sampling procedure.

Hydrogen Cyanide: Hydrogen cyanide was quantitatively determined by the method of *Vickroy* and *Gaunt* which is based on an ion-selective electrode determination of cyanide ion (6). Hydrogen cyanide was trapped by passing mainstream smoke through an Ascarite column. This column was then solutioned, and cyanide was measured directly with a cyanide ion-selective electrode versus a standard KCl reference electrode.

Nitric Oxide: In *fresh* cigarette smoke, nitric oxide has been shown to be the major oxide of nitrogen present. Nitrogen dioxide is gradually formed by the slow oxidation of nitric oxide (7). Nitric oxide was determined according to the method of *Urbanic* and *Sutt* (8).

Primary Amines and Ammonia: The determination of ammonia and primary amines in cigarette smoke is in large part based on techniques developed by *Mathewson* (9, 10). Mainstream cigarette smoke was drawn through two bubble traps in series, each containing 50 ml of 0.1 N sulfuric acid and 1.28 mg of an internal standard (t-butylamine). This highly acidic scrubbing solution reacts with the alkaline components in smoke to produce non-volatile sulfate salts. After 15 cigarettes were smoked, the contents of the two traps were combined and the pH adjusted to 3.0 with

Figure 1. Typical chromatograms from the organic vapor phase analysis.



1. Methane
2. Ethylene
3. Acetylene
4. Ethane
5. Propene
6. Propane
7. Propadiene + methyl chloride*
8. Propyne
9. Methanol
10. Acetaldehyde
11. 2-Methyl propane
12. C_4-Alkene
13. 1,3-Butadiene
14. Butane + trans-2-Butene
15. cis-2-Butene
16. 1,2-Butadiene
17. Ethanol
18. Acetonitrile
19. Furan
20. Acrolein
21. Propionaldehyde
22. Acetone
24. C_5-Alkene
25. Isoprene + C_5-Mixture
26. C_5-Mixture
27. 1,3-Pentadiene
28. C_5-Alkenyne
29. Propionitrile
30. Isobutyraldehyde
31. 2-Methyl furan
32. Methyl vinyl ketone
33. 2,3-Butanedione
34. 2-Butanone
35. Hexane + C_6-mixture
36. C_6-Mixture
37. Isobutyronitrile
38. Crotonaldehyde
39. Benzene
40. C_6-Cycloalkene
41. n-Butyronitrile
42. Isovaleraldehyde
43. 2,5-Dimethyl furan
44. 2-Pentanone
51. Toluene

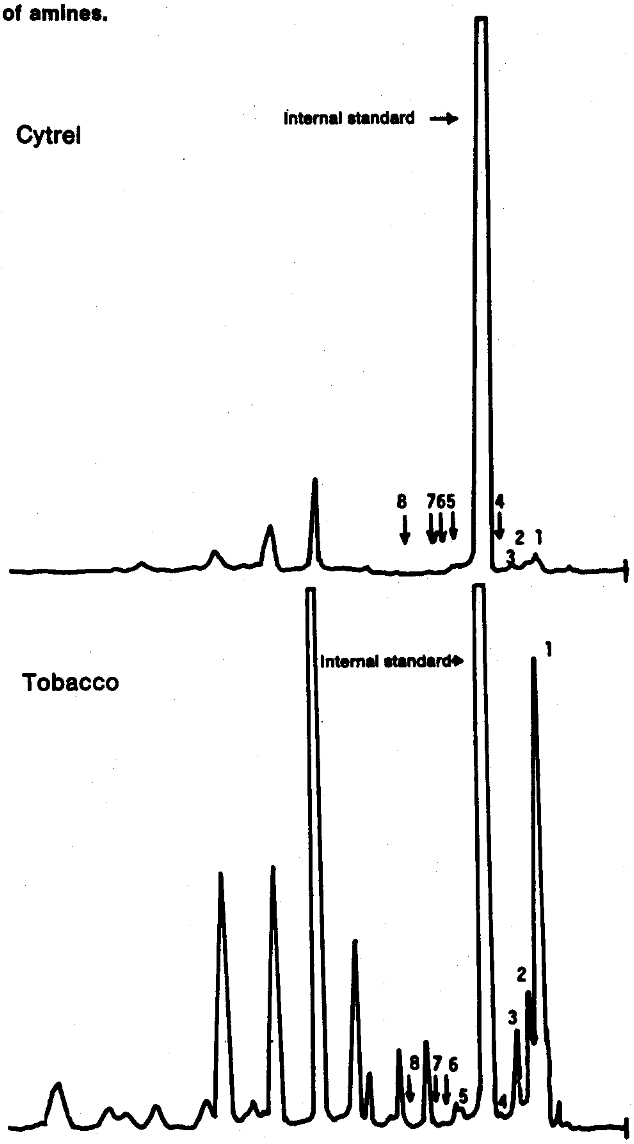
* Tobacco only

sodium hydroxide. After this solution was concentrated under vacuum at ~ 80 °C to 1.0 ml with a rotary evaporator, an aliquot of this concentrate was injected into a specially fabricated gas chromatographic injector packed with either soda lime or barium oxide (11, 12). Soda lime, a mixture of sodium hydroxide and calcium oxide, and barium oxide were both capable of converting ammonium and amine sulfates to their corresponding free bases. Under these highly alkaline conditions the amines moved readily and with little tailing through the injector and into a chromatographic column suited for the separation of free amines and ammonia (13). While the trapping and concentrating procedures were identical for both amines and ammonia, the exact chromatographic conditions and methods of detection were quite different and are treated separately below.

a) Primary Amines

Summary of gas chromatographic conditions:

Figure 2. Typical chromatograms from the determination of amines.



1. Methylamine
2. Dimethylamine
3. Ethylamine
4. Isopropylamine
5. n-Propylamine
6. sec-Butylamine
7. Isobutylamine
8. n-Butylamine

Column:	12-foot stainless steel tubing treated with alcoholic KOH and then packed with 10 % Pennwalt 223 + 4 % KOH on 80/100 mesh Gas-Chrom R.
Injector:	Stainless steel packed with soda lime.
Temperature:	Injector: 150 °C. Column: Initial 70 °C for 3 minutes — program rate 12°/minute — final 170 °C for 32 minutes. Detector: 200 °C.
Carrier gas:	Helium, approximately 22 ml/minute, adjusted for maximum chromatographic resolution.
Detector:	Perkin-Elmer nitrogen-selective detector, Part No. 228-0301.

The column used in this analysis permitted excellent

Table 2. Vapor phase analysis of smoke from cigarettes containing blends of tobacco with Cytrel.

Test	Sample				
	100 % tobacco	10 % Cytrel	20 % Cytrel	50 % Cytrel	100 % Cytrel
1. Hydrocarbons (µg/cigarette):					
Methane	1221	1097	827	493	174
Ethylene	343	292	243	147	59
Acetylene	45	39	34	22	17
Ethane	500	436	348	215	79
Propene	351	297	245	147	52
Propane	281	241	195	118	40
Propadiene	7	6	6	3	2
Methyl chloride*	493	370	298	158	ND**
Propyne					7
2-Methylpropane	35	27	23	14	2
C_4-Alkene	147	122	101	60	18
1,3-Butadiene	77	65	58	35	13
Butane	105	88	70	43	13
trans-2-Butene	49	41	33	19	7
cis-2-Butene	54	45	38	23	8
1,2-Butadiene	7	6	5	3	1
C_5-Alkene	68	56	45	27	8
Isoprene & C_5-mixture*	1093	809	709	382	11
C_5-Mixture					10
C_5-Mixture	16	14	11	7	<1
C_5-Mixture	54	44	35	21	7
C_5-Alkenyne	3	2	2	1	1
C_6-Mixture	67	52	45	26	6
Hexane & C_6-mixture	23	18	15	8	4
C_6-Mixture	12	9	8	4	<1
Benzene	104	88	69	44	13
C_6-Cycloalkene	3	3	2	1	<1
Toluene	163	134	108	65	12
2. Alcohols, aldehydes and ketones (µg/cigarette):					
Methanol	558	440	335	208	3
Ethanol	2	2	2	1	<1
Acetaldehyde	1436	1214	1022	675	243
Acrolein	92	86	73	50	31
Propionaldehyde	175	148	125	83	37
Isobutyraldehyde+	<129	<102	<89	<53	<9
Crotonaldehyde	72	54	52	35	17
Isovaleraldehyde	77	54	51	31	5
Acetone	648	552	450	303	107
Methyl vinyl ketone*	285	255	225	147	64
2,3-Butanedione					
2-Butanone	207	174	147	102	44
2-Pentanone	79	64	57	41	23
3. Furans (µg/cigarette):					
Furan	65	58	45	29	15
2-Methylfuran	87	77	55	28	7
2,5-Dimethylfuran	135	119	82	41	7
4. Nitriles (µg/cigarette):					
Acetonitrile	189	161	134	103	25
Propionitrile+	<90	<71	<62	<37	<6
Isobutyronitrile	30	24	20	12	3
Butyronitrile	6	5	4	3	<1
5. Carbon monoxide (mg/cigarette)	17.6	16.4	15.2	11.5	6.0

Table 2 (contd.)

Test	Sample				
	100 % tobacco	10 % Cytrel	20 % Cytrel	50 % Cytrel	100 % Cytrel
6. Carbon dioxide (mg/cigarette)	67.7	59.3	54.6	45.9	48.2
7. Hydrogen cyanide (µg/cigarette)	280	256	248	185	25
8. Nitric oxide (µg/cigarette)	93	98	90	57	31
9. Ammonia (µg/cigarette)	22	22	20	21	50
10. Primary amines (µg/cigarette):					
Methylamine	5.0	4.5	5.0	3.5	<0.5
Dimethylamine[++]	1.0	<0.5	1.0	1.0	<0.5
Ethylamine	1.5	1.0	1.0	1.0	<0.5
Isopropylamine	<0.5	ND[**]	1.0	<0.5	ND
n-Propylamine	<0.5	<0.5	<0.5	<0.5	ND
sec-Butylamine	ND	ND	ND	ND	ND
Isobutylamine	<0.5	<0.5	<0.5	<0.5	ND
n-Butylamine	<0.5	<0.5	<0.5	ND	ND
11. Hydrogen sulfide (µg/cigarette)	46	46	48	56	55
12. Sulfur dioxide (µg/cigarette)	4.2	4.0	3.9	2.2	1.2

[*] Brackets indicate incomplete chromatographic resolution. Maximum deliveries are calculated and shown opposite the major component.
[**] ND = Not detected.
[+] Incomplete chromatographic resolution; maximum deliveries reported.
[++] A secondary amine. Its delivery has also been included because of its possible role in the formation of N,N-dimethylnitrosamine.

separation of all of the primary amines of interest as well as dimethylamine, a secondary amine important because of its possible role in the formation of dimethylnitrosamine. However, even with excellent chromatographic resolution, the possibility of interfering compounds in smoke is not insignificant and made the use of a nitrogen-selective detector advisable. This detector has a greater sensitivity toward carbon-nitrogen compounds than a conventional flame ionization detector and is 5000 times less sensitive to hydrocarbons (14).
The peak areas for all compounds of interest were measured with a Perkin-Elmer PEP-2 gas chromatographic data system. By using t-butylamine as an internal standard and calibrating with known standards, exact deliveries were readily calculated using the computer. The precision of this analysis is such that the amine data have been expressed in Table 2 to the nearest 0.5 µg/cigarette. Chromatograms typical of the amine analysis for Cytrel and tobacco are shown at equivalent detector sensitivities in Figure 2.

b) Ammonia

Summary of gas chromatographic conditions:

Column: Same as amine separation.
Injector: Stainless steel packed with barium oxide.
Temperature: Injector: 150 °C.
Column: 70 °C, isothermal[*].
Detector: 200 °C.
Carrier gas: Same as amine separation.
Detector: Thermal conductivity detector at 300 ma.

Integration was performed with a Perkin-Elmer PEP-2 data system; standard solutions of ammonium chloride in 0.1 N sulfuric acid were used for calibration.

Hydrogen Sulfide: This technique, similar to the hydrogen cyanide analysis, is based on a potentiometric determination of sulfide with an ion-selective electrode. The procedure was identical to that reported by *Morie* (15).

Sulfur Dioxide: Sulfur dioxide was determined according to the method of *West* et al. (16, 17). While this procedure was originally intended for air pollution analyses, it has also been applied to tobacco smoke (18).

RESULTS AND DISCUSSION

A summary of the results from the vapor phase analysis of smoke from cigarettes made with blends of Cytrel and tobacco is given in Table 2. If cigarettes containing only 100 % tobacco or 100 % Cytrel are considered first, it is evident that all but two compounds

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[*] After several injections the column was programmed rapidly to 170 °C until the heavier, less volatile components cleared the chromatograph.

were delivered in lower amounts by the Cytrel-containing cigarettes. The two exceptional compounds which were not observed in decreased levels are hydrogen sulfide and ammonia. Hydrogen sulfide was delivered at equivalent or slightly increased amounts. Ammonia was delivered in significantly greater amounts in this study but was still within the range observed for many all-tobacco cigarettes (12, 19, 20).

In addition to considering cigarettes made from the 100 % materials, it is equally important to evaluate cigarettes containing blends of Cytrel and tobacco. If each material combusts in a blend in the same manner as it does in 100 % form, then the delivery of any component should be proportional to the blend level. That is:

$$\text{delivery of Component A}_{\text{Blend}} =$$

$$= [\text{cigarette Blend Level}_{\text{tobacco}} \times \text{delivery Component A}_{100\,\%\ \text{tobacco}}] +$$

$$+ [\text{cigarette Blend Level}_{\text{Cytrel}} \times \text{delivery Component A}_{100\,\%\ \text{Cytrel}}] \cdot$$

For example, consider the delivery of carbon monoxide for cigarettes made from 100 % tobacco and Cytrel. Their respective deliveries are 17.6 and 6.0 mg/cigarette, and a 50 % blend of the two should deliver:

$$\text{delivery of CO}_{50/50\,\%\ \text{Blend}} = (0.5)(17.6) + (0.5)(6),$$

$$= 11.8\ \text{mg/cigarette};$$

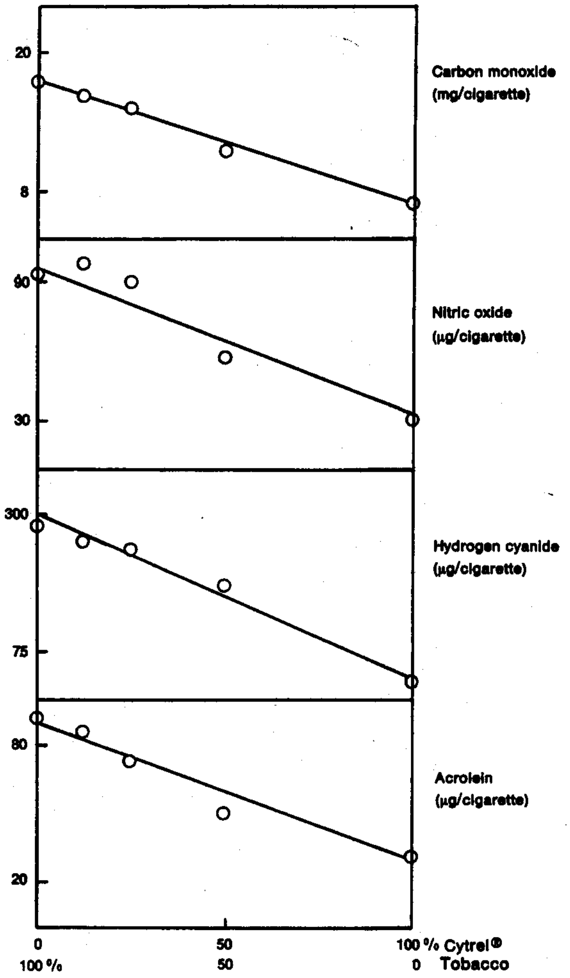
and for an 80/20 tobacco/Cytrel blend:

$$\text{delivery of CO}_{80/20\,\%\ \text{Blend}} = (0.8)(17.6) + (0.2)(6),$$

$$= 15.3\ \text{mg/cigarette}.$$

These predicted deliveries agree well with the measured deliveries of 11.5 and 15.2 mg/cigarette. The easiest technique for determining whether blends are delivering proportionate amounts is to display the data graphically. This has been done for several compounds in Figure 3. In these examples the deliveries are proportional to the blend levels and could be readily predicted if blend levels and deliveries for 100 % materials are known. The delivery of ammonia is the only exception to this predictable behavior. It is essentially constant and equal to that of 100 % tobacco cigarettes for all Cytrel-tobacco blend levels examined.

Figure 3. Comparison of several component deliveries as a function of Cytrel-tobacco blend levels.



SUMMARY

The vapor phase of smoke from cigarettes containing Cytrel has been extensively characterized and compared to that from identical cigarettes made from several blend levels of Cytrel and tobacco as well as those made from 100 % flue-cured tobacco. In only two instances out of sixty did 100 % Cytrel cigarettes deliver equivalent or greater amounts of any compound than did the corresponding tobacco cigarettes. For the remaining compounds reductions were observed in a predictable fashion for all Cytrel-tobacco blend cigarettes examined. In the course of these analyses no compound was observed in smoke from Cytrel cigarettes that was not also present in tobacco smoke.

ZUSAMMENFASSUNG

Die Dampfphase des Rauches von Cytrel enthaltenden Cigaretten wurde eingehend untersucht und mit derjenigen identischer Cigaretten verglichen, welche einerseits aus verschiedenen Mischungen von Cytrel und Tabak und andererseits zu 100 % aus „flue-cured"-Tabak bestanden. Nur bei 2 von 60 Inhaltsstoffen war die Ausbeute in den zu 100 % aus Cytrel bestehenden Cigaretten ebenso groß oder größer als in den entsprechenden Cigaretten aus Tabak. Bei den übrigen Verbindungen wurden Verminderungen in vorhersehbarem Maße für alle Cigaretten mit Mischungen aus Cytrel und Tabak beobachtet. Im Laufe der Untersuchungen wurde im Rauch der Cytrel-Cigaretten keine Verbindung beobachtet, die nicht auch im Tabakrauch vorhanden war.

RESUME

La phase vapeur de la fumée de cigarettes contenant du Cytrel a été analysée de façon détaillée et comparée à la fumée de cigarettes identiques, composées de divers mélanges de Cytrel et de tabac, ainsi que de 100% de tabac «flue-cured». Quelque soit le composé considéré, c'est dans deux cas sur soixante seulement que la quantité produite par une cigarette 100% Cytrel est égale ou supérieure à celle produite par une cigarette de tabac pur. Pour tous les autres composés on a observé des diminutions prévisibles à partir de tous les mélanges tabac-Cytrel étudiés. Dans toutes ces analyses, on n'a retrouvé dans la fumée de cigarettes Cytrel aucun composé qui ne soit également présent dans la fumée de tabac.

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