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# STUDIES ON THE CHEMICAL EVALUATION OF TOBACCO QUALITY. III EVALUATION OF TOBACCO QUALITY FROM GAS CHROMATOGRAPHIC ANALYSIS OF PARTICULATE PHASE OF THE CIGARETTE SMOKE

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In the gas chromatogram of the particulate phase from flue-cured tobacco smoke, several pairs of peaks were selected and the logarithms of the ratios in the area of the individual pairs were calculated to see their correlation to the corresponding organoleptic evaluations.

The correlation coefficients obtained were generally higher than those obtained previously merely from the ratios of peak areas. Using the logarithmic values as variables, the linear equations for the evaluation of the individual organoleptic characteristics were computed.

The results were applied to twenty kinds of flue-cured type tobaccos, including tip, leaf and cutter for each, and the calculated values were compared with the corresponding organoleptic evaluations.

The individual estimated values obtained from the present method were significantly correlated to the values of the corresponding organoleptic evaluations.

## INTRODUCTION

In a previous paper (1), we proposed a series of quality equations for the evaluation of flue-cured tobacco from the gas chromatographic values of the particulate phase of cigarette smoke. These equations were considered to be applicable to the evaluation of flue-cured tobacco quality instead of the sensory test, although their applications to low grade samples were not successful.

The object of this paper is to obtain more reliable equations applicable not only to high and middle grade samples but also to low grade ones.

Higher correlation coefficients to the organoleptic evaluation values were generally obtained by the use of the logarithmic values of the ratios of peak areas than the use of mere peak area ratios.

The equations with the highly correlated logarithmic values as independent variables were computed, and their applicability to the evaluation of tobacco qualities was examined.

## EXPERIMENTAL

### *Materials*

The samples used in this experiment consisted of 60 kinds of flue-cured tobacco leaves, among which 40 kinds were domestic tobaccos and the others were foreign ones and the samples were classified to six ten-member groups according to the organoleptic evaluation of aroma ($2 \leq Z > 3$, $3 \leq Z > 4$, $4 \leq Z > 5$, $5 \leq Z > 6$, $6 \leq Z > 7$ and $7 \leq Z$).

Each kind of tobacco sample was manufactured into non-blended cigarettes.

### *Gas chromatography*

A Hitachi Model K-53 gas chromatograph equipped with a flame ionization detector and KP-1 pyrolyzer was used. Chromatographic separations were achieved on a 3 m X 3 mm i.d. stainless steel tube filled with 20% Carbowax 20 M on 60-80 mesh Chromosorb W with helium as the carrier gas at a flow rate of 80 ml/min. Pyrolyzing tube of the pyrolyzer was used as an injection port of the gas chromatograph and was maintained at 280°C. During the analysis, column temperature was elevated linearly from 80°C to 250°C (3°C/min) and then kept at that temperature.

### *Introduction of particularate phase of smoke into the gas chromatograph*

The separation of particulate phase from smoke was accomplished by the use of the glass fiber Cambridge filter. A very small quantity of the particulate matter, stuck in the Cambridge filter, was put in the pipetter which was then placed into the pyrolyzer connected to the gas chromatograph for 20 seconds.

### *Measurement of peak areas on gas chromatogram*

The peak areas on the chromatogram were measured by a Takeda Riken Model TR-2211 digital integrator. The base line corrections were manually carried out twice in an experiment: once just after the area of peak A was printed out, and then just after peak Q.

To see the reproducibility of the ratio of each major peak to nicotine peak (area/area), two types of flue-

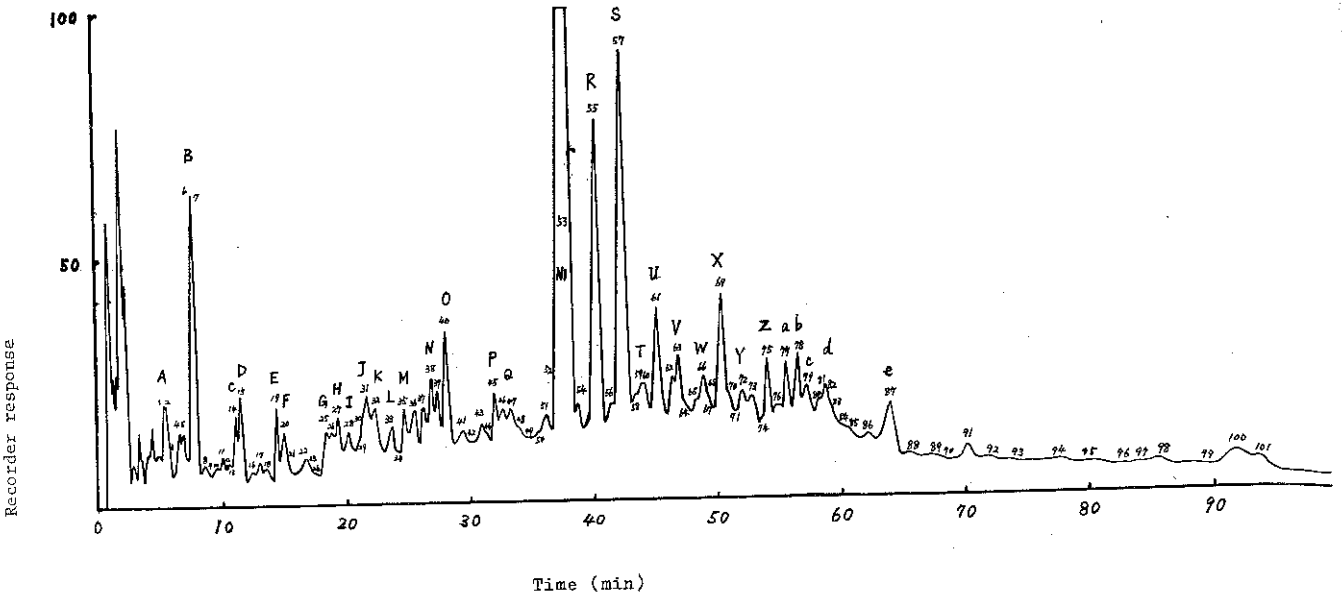


Figure 1. Gas chromatogram of particulate phase of smoke (a middle grade of Leaf from USA).

cured tobacco, one domestic type leaf and one U.S.A. type leaf, were examined by the method described above. The experiments were repeated ten times for each sample, and among the over-twenty peaks examined, 4 peaks (I, J, P, d) were considerably unstable giving the coefficients of variation higher than 20%. These four peaks were excluded in the later experiments.

*Sensory test*

Each sensory test was scored and ranged from 1 to 9, by an expert panel which consists of 10 to 13 trained members engaged in the blending of leaf tobaccos for the cigarette making. All test samples were brought as blind samples. The panel members discussed the results given by each member, and a final evaluation was fixed by mutual agreement. In the cases of aroma and taste, the higher scores of evaluation were marked when the samples were better in such characteristics, while, in the cases of harshness, strength and offensive odor and taste, scores were higher when samples had less such characteristics. In the case of dryness, higher score was marked when samples had better dryness.

## RESULTS AND DISCUSSION

*Gas chromatogram of the particulate phase of the smoke of flue-cured tobacco*

A gas chromatogram of the particulate phase of the smoke of a foreign flue-cured tobacco is shown in Figure 1. As seen in the figure, at least 100 peaks, most of which would contain undoubtedly more than one compound, are observed in the chromatogram. In these peaks, nicotine peak (peak 53) is the largest and the other major peaks are tentatively assigned respectively as dipentene (peak B), furfural and 3-vinyl pyridine (H), 5-methyl furfural (M), phytadiene (R), phenol and o-cresol (S), m- and p-cresol (U), myosmine (X). They seem except for phytadiene to have occurred by pyrolytic degradation of the main tobacco constituents, i.e. alkaloid, sugars, resinous substances, etc.

Further identification of the other peaks has been reported elsewhere (2).

To obtain more relevant correlation coefficients, the ratios of appropriate two peaks which gave high correlations to organoleptic evaluation in the previous paper (1) were taken to the logarithmic values and those values were computed on the correlations to individual organoleptic evaluations. The results are shown in Table 1.

From **Table 1**, it is obvious that the logarithms of the ratios give the higher coefficients than the mere ratios for aroma, taste and strength, whereas there are no appreciable difference among both of them for harshness, offensive odor & taste and dryness. In organoleptic evaluations the panel seems to judge and score the characteristics of smoke not only linearly but

**Table 1. Correlation between the ratio of peak area and organoleptic properties**

| Aroma | | Taste | | Harshness | |
|---|---|---|---|---|---|
| log B/E | 0.74*** | log B/E | 0.73*** | log B/Ni[a] | 0.64*** |
| B/E | 0.69*** | B/E | 0.68*** | B/Ni | 0.54*** |
| log O/M | 0.23 | log O/M | 0.25 | log O/M | 0.54*** |
| O/M | 0.22 | O/M | 0.25 | O/M | 0.52*** |
| log R/U | 0.20 | log R/U | 0.19 | log R/S | 0.48*** |
| R/U | 0.20 | R/U | 0.19 | R/S | 0.49*** |

[a] *Nicotine*
* $|r| \geq 0.26(58, 0.05)$
** $|r| \geq 0.33(58, 0.01)$
*** $|r| \geq 0.42(58, 0.001)$

| Offensive odor & taste | | Strength | | Dryness | |
|---|---|---|---|---|---|
| log B/E | 0.54*** | log E/Ni | 0.75*** | log N/M | 0.72*** |
| B/E | 0.52*** | E/Ni | 0.64*** | N/M | 0.72*** |
| log O/M | 0.53*** | log Ni | −0.68*** | log B/E | 0.62*** |
| O/M | 0.50*** | Ni | −0.67*** | B/E | 0.63*** |
| log R/U | 0.51*** | | | log R/S | 0.41** |
| R/U | 0.49*** | | | R/S | 0.40** |

**Table 2. Linear equations for the evaluation from gas chromatographic values in smoke**

| | Linear equations |
|---|---|
| Aroma | 7.25 x log B/E + 2.25 |
| Taste | 6.69 x log B/E + 2.29 |
| Harshness | 1.50 x log R/S + 2.27 x log B/Ni[b] + 9.34 |
| Offensive odor & taste | 2.87 x log B/E + 3.43 x log R/U + 4.34 |
| Strength | 3.29 x log E/Ni + 11.75 |
| Dryness | 6.10 x log N/M + 6.37 |

[a] *Figures in the parentheses are in the case of antilogarithm.*
[b] *Nicotine.*
*** (0.001)

| Multiple correlation | | Standard error of estimate | |
|---|---|---|---|
| 0.74*** | (0.69***)[a] | 1.16 | (1.26)[a] |
| 0.73*** | (0.68***) | 1.10 | (1.18) |
| 0.70*** | (0.66***) | 0.55 | (0.58) |
| 0.67*** | (0.65***) | 0.86 | (0.88) |
| 0.75*** | (0.64***) | 0.60 | (0.70) |
| 0.72*** | (0.72***) | 0.82 | (0.82) |



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also non-linearly with the qualitative quantities as in the cases of aroma, taste and strength.

The equations with the highly correlated logarithmic values as independent variable were computed. The equations, multiple correlations and standard errors of the estimates are shown in **Table 2.**

As seen in the table, the multiple correlation coefficients of the regression equations for the evaluation by the use of the logarithmic values are higher than those obtained by the use of ratios only for aroma, taste and strength, whereas there are no appreciable difference among both of them for harshness, offensive odor & taste and dryness. The regression equations obtained need not three independent variables as in the case of the previous equations (1) but only one or two independent variables are suffcient. Application of the gas chromatographic evaluation method to some flue-cured tobaccos.

**Table 3. Relation between observed values and estimated values**

| | Aroma | | Taste | |
|---|---|---|---|---|
| | $Z$[a] | $\hat{Z}$[b] | $Z$ | $\hat{Z}$ |
| Japan Tip—3 | 5.5 | 5.9 | 5.5 | 5.6 |
| Japan Tip—3 | 4.0 | 5.0 | 4.0 | 4.8 |
| Japan Tip—3 | 4.0 | 5.2 | 4.0 | 5.0 |
| Japan Tip—3 | 4.0 | 4.1 | 4.0 | 4.0 |
| Japan Leaf—1 | 4.5 | 4.5 | 4.5 | 4.4 |
| Japan Leaf—1 | 4.5 | 3.8 | 4.5 | 3.7 |
| Japan Leaf—3 | 6.0 | 6.2 | 6.0 | 5.9 |
| Japan Leaf—3 | 4.0 | 5.6 | 4.0 | 5.4 |
| Japan Cutter—1 | 4.0 | 2.5 | 4.0 | 2.5 |
| Japan Cutter—1 | 4.0 | 3.6 | 4.0 | 3.5 |
| Japan Cutter—1 | 4.0 | 2.9 | 4.0 | 2.9 |
| Japan Cutter—3 | 4.0 | 4.6 | 4.0 | 4.4 |
| Japan Cutter—3 | 4.0 | 2.8 | 4.0 | 2.8 |
| Japan Cutter—3 | 2.0 | 1.1 | 2.0 | 1.2 |
| Japan Cutter—3 | 2.0 | 1.8 | 2.0 | 1.9 |
| Japan Cutter—3 | 2.5 | 2.4 | 2.5 | 2.4 |
| Japan Cutter—3 | 3.0 | 4.5 | 3.0 | 4.4 |
| Japan Cutter—3 | 3.0 | 4.6 | 3.0 | 4.4 |
| Japan Cutter—3 | 3.0 | 2.6 | 3.0 | 2.6 |
| U.S.A. Cutter—3 | 5.0 | 4.4 | 4.0 | 4.3 |
| Average of $Z-\hat{Z}$ | | 0.77 | | 0.68 |
| Standard deviation of $Z-\hat{Z}$ | | 0.54 | | 0.53 |
| Correlation of $Z$ and $\hat{Z}$ | | 0.74*** | | 0.74*** |

[a] *Observed value.*
[b] *Estimated value.*

| Harshness | | Offensive odor & taste | | Strength | | Dryness | |
|---|---|---|---|---|---|---|---|
| $Z$ | $\hat{Z}$ | $Z$ | $\hat{Z}$ | $Z$ | $\hat{Z}$ | $Z$ | $\hat{Z}$ |
| 3.7 | 4.7 | 5.5 | 4.9 | 3.5 | 4.6 | 6.0 | 6.2 |
| 5.0 | 5.0 | 4.0 | 4.6 | 5.5 | 5.4 | 4.5 | 5.3 |
| 5.0 | 5.2 | 4.0 | 4.6 | 4.0 | 5.7 | 4.0 | 4.9 |
| 4.0 | 4.7 | 4.0 | 4.5 | 5.0 | 5.4 | 4.0 | 5.0 |
| 5.0 | 4.6 | 5.3 | 4.5 | 5.0 | 5.0 | 5.3 | 5.4 |
| 5.0 | 4.7 | 5.0 | 4.5 | 5.0 | 5.5 | 5.5 | 5.3 |
| 4.0 | 5.1 | 6.0 | 5.5 | 4.3 | 4.7 | 6.0 | 6.0 |
| 4.7 | 4.6 | 4.5 | 4.7 | 6.0 | 4.7 | 4.5 | 5.5 |
| 5.5 | 5.2 | 4.0 | 4.5 | 6.0 | 6.4 | 3.5 | 4.5 |
| 5.0 | 5.0 | 5.0 | 4.9 | 5.5 | 5.6 | 5.0 | 5.2 |
| 5.0 | 4.8 | 3.5 | 4.1 | 6.0 | 6.0 | 4.0 | 4.7 |
| 5.0 | 5.1 | 3.5 | 4.8 | 5.0 | 5.4 | 3.5 | 5.5 |
| 5.5 | 4.6 | 4.5 | 3.7 | 6.0 | 5.9 | 4.5 | 5.1 |
| 6.0 | 7.8 | 5.0 | 4.6 | 7.0 | 6.6 | 5.0 | 6.5 |
| 6.0 | 6.5 | 6.0 | 4.4 | 7.0 | 7.3 | 6.0 | 5.5 |
| 6.0 | 6.2 | 4.0 | 5.3 | 7.0 | 6.4 | 4.0 | 5.3 |
| 5.0 | 6.0 | 5.0 | 5.1 | 6.0 | 5.1 | 5.0 | 4.8 |
| 6.0 | 6.9 | 6.0 | 5.2 | 7.0 | 6.3 | 6.5 | 5.6 |
| 6.0 | 6.2 | 4.5 | 4.6 | 7.0 | 6.4 | 4.5 | 4.6 |
| 7.0 | 6.6 | 6.5 | 6.1 | 8.0 | 6.8 | 7.0 | 5.8 |
| 0.52 | | 0.62 | | 0.56 | | 0.72 | |
| 0.47 | | 0.41 | | 0.47 | | 0.54 | |
| 0.72*** | | 0.54* | | 0.78*** | | 0.59** | |

* $|r| \geqq 0.44(18,\ 0.05)$
** $|r| \geqq 0.56(18,\ 0.01)$
*** $|r| \geqq 0.70(18,\ 0.001)$.

The evaluation of each organoleptic property by the present gas chromatographic method was applied to twenty kinds of flue-cured type tobaccos, in which one kind was U.S.A. products and the others were domestic products, including Tip, Leaf and Cutter for each, and the calculated values were compared with the corresponding organoleptic evaluations (**Table 3**).

As shown in the table, the individual estimated values obtained from the present method were significantly correlated to the values of the corresponding organoleptic evaluations.

This indicates that the linear equations obtained are applicable for the evaluation of tobacco quality.

However, the observed differences between the evaluated values and the estimated values seemed not to be reduced by improving this method since the differences in the score among the panel are considered to fluctuate within the range of 1.0, and, furthermore, some delicate differences of organoleptic judgment would not be detected by the gas chromatographic method, as in the case of earthy or green odor.


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## LITERATURE CITED


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