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# Comparison of Changes in Headspace Volatiles of some Coffee Blends during Storage

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Headspace volatiles of commercial blends of Finnish roasted coffee 'Juhlamokka', 'Presidentti' and 'Espresso' were analysed by gas chromatography. The relative proportions of 43 compounds were observed over a 1 year period. The ratios of acetone/butanedione, acetone/propanal, thiophene/propanal, thiophene/butanedione, and butanedione/2-methylfuran were, in most cases, useful indicators of storage age (|r|).

The relative proportions of acetone, methyl acetate, 2- and 3-methylfuran, 2-butanone, and 2,5-dimethylfuran were higher in the headspace of further roasted 'Espresso' blend containing Robusta quality than of the other blends. Only traces of pyridine were found in 'Juhlamokka' and 'Presidentti', but it was 5–10 times more abundant in 'Espresso'. The proportions of methanol, 2-methylpropanal, butanedione, 2- and 3-methylbutanal, 2,3-pentanedione, and 2-furaldehyde were lower in 'Espresso' than in other coffees.

The levels of acetone and 2-methylpropanal were higher in ground coffee than in the respective coffee beans. The proportion of 3-methylbutanal was greater in whole beans than in ground coffee. No marked differences between these two blends could be found.

## INTRODUCTION

The quality of coffee is related to its flavour and aroma, the development of these depending on chemical changes occurring during roasting. Many factors such as the origin and the type of the coffee beans, roasting time and temperature, and the degree and method of roasting, affect the resulting quality.

Reducing sugars and free amino acids are precursors of a great number of volatile aroma compounds. Free amino acids react with reducing sugars in Maillard reactions or Strecker degradations, or the compounds are thermally degraded.

Unroasted Arabica coffee beans contain high concentrations of sucrose, while Robusta is especially rich in reducing sugars glucose and fructose. The total sugar content is higher in Arabica than in Robusta. Over 99% of sucrose, glucose and fructose are degraded during roasting (Tressl *et al.*, 1982).

The level of free amino acids is typically higher in the Robusta than in the Arabica variety. Contents of the 'Strecker-active' amino acids, such as glycine, alanine, valine, isoleucine, phenylalanine, and tyrosine, are especially high in Robusta (Tressl *et al.*, 1982). Alkaline and heterocyclic amino acids are also more abundant in Robusta than in Arabica.

However, data for the levels of free amino acids or sugars of freshly harvested, processed green coffees or stored coffees are not available. It has been stated that the amounts of reducing sugars in Robusta beans are formed during storage from enzymatic hydrolysis (Trugo, 1985).

Pyrazines in roasted coffee are formed, either through pyrolysis or Strecker degradation, depending on the amino acid precursors and roasting conditions (Baltes & Bochmann, 1987). Various compositions of pyrrole and pyridine derivatives are also formed in the presence of sugars and several amino acids (Baltes & Bochmann, 1987; Tressl *et al.*, 1981; Shibamoto, 1977). Pyridines and pyrroles have been observed to increase when lengthening the roasting time (Shibamoto *et al.*, 1981). Tressl *et al.* (1981) found that roasted Arabica contains less alkylated pyrroles and more furfurylpyrroles than Robusta.

Furan compounds are formed especially in the su-

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crose-containing systems (Baltes & Bochmann, 1987). 2-Furaldehyde is reduced during roasting, mostly to 2-furylmethanol, and small amounts of acyl and 5-methylacyl furan derivatives (Silwar & Tressl, 1989). The reactions of 2-furaldehyde and hydrogen sulphide form sulphur-containing furan derivatives (Shibamoto, 1977; Silwar & Tressl, 1989). 2-Furylmethanol increases in the course of roasting, and gives an undesirable flavour to the coffee (Shibamoto *et al.*, 1981).

Acetaldehyde and propanal originate from sugar pyrolysis, while 2-methylpropanal, 2-methylbutanal, and 3-methylbutanal are the Strecker degradation products of valine, isoleucine and leucine (Liardon *et al.*, 1984). The concentrations of 2-methylbutanal, 3-methylbutanal, and 2-methylfuran reach typically higher levels in freshly roasted Robusta than Arabica (Piringer, 1983).

According to Nurok *et al.* (1978) the mixture of Arabica:Robusta 99:1 can be differentiated from pure Arabica coffee by measuring the amount of sulphur-containing compounds present (Nurok *et al.*, 1978).

The aim of this work was to test a quick headspace GC-method to differentiate various coffee blends and to observe their changes during storage.

## MATERIALS AND METHODS

The coffee samples were the commercial blends 'Juhlamokka', 'Presidentti' and 'Espresso' processed by Oy Gustav Paulig Ab (Helsinki, Finland). The raw coffee components in the 'Juhlamokka' blend were Santos, Colombia, Guatemala, Costa Rica and Kenya. The 'Presidentti' blend was composed of the same components together with Mocca, whereas Santos, Colombia, Guatemala, Costa Rica and Robusta were used in the 'Espresso' blend. The Santos, Mocca and Robusta beans were dry-processed whereas wet processing was applied to other beans. The 'Espresso' blend has been developed for the specific coffee makers to produce a dark colour drink with strong aroma. The characteristics were obtained by a special selection of coffee beans, together with a prolonged time for roasting.

Ground roasted 'Juhlamokka' and 'Presidentti' blends were packed in 500 g polyester–Al–polyethylene laminate bags and evacuated. The roasted beans of 'Juhlamokka' and 'Presidentti', as well as ground 'Espresso' coffee, were packed in 250 g bags of polyester–Al–polyethylene laminate and flushed with $CO_2$. All coffee samples were stored at room temperature for 0 to 12 months.

The coffee samples were analysed using the method described earlier (Kallio *et al.*, 1990). Thirty five grams of coffee (ground or beans) were weighed in duplicates in a 500 ml Erlenmeyer bottle closed with a headspace valve (J & W Scientific, Folsom, CA). The samples were kept at 37°C for 19 h, and the headspace volatiles analysed with a gas chromatograph (Varian Aerograph 3700, Walnut Creek, CA, and a Hewlett Packard 3388A integrator, Palo Alto, CA) equipped with an FI-detector.

A DB-1701 fused silica open tubular column (30 m × 0·254 mm i.d., $d_f$ 0·25 $\mu$m, J & W Scientific) was connected to the on-column injector kept at room temperature. The detector temperature was 250°C, and the carrier gas flow rate 27 cm/s at 20°C, the inlet pressure of the He-carrier being 90 kPa.

During injection the He-pressure was lowered to 60 kPa and the oven temperature to +10°C with solid $CO_2$. Part of the front of the column was placed in liquid $N_2$ (Kallio *et al.*, 1990). A 3 ml coffee headspace sample was injected with a 10 ml gas-tight syringe (Hamilton, Reno, NV) at the speed of 0·5 ml/min into a column. The pressure was adjusted to 90 kPa before the cold trap was removed. The GC-program was 10–100°C, 2°C/min.

The compounds from different coffee samples (ground, beans, espresso) were identified by GC–MS. The electron impact mass spectral (EI–MS) analyses (ionization energy 70 eV) were carried out on a VG 7070E spectrometer employed with a VG-11-250 data system and a Dani 3800 HR ch gas chromatograph (VG, Wythenshawe, Manchester, UK). Both the column and program were the same as used in the GC analysis.

## RESULTS AND DISCUSSION

Forty-three volatile compounds were identified according to their mass spectra and Kovats' indices ($I_k$), and the changes of their proportions in the headspace were observed over a 1-year period. The compounds numbered 1–43 are listed in Table 1. Methyl propanoate (15),

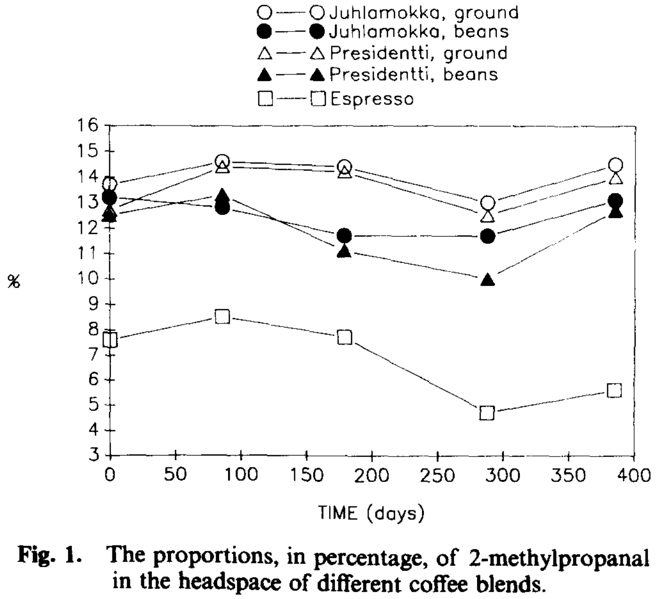

**Fig. 1.** The proportions, in percentage, of 2-methylpropanal in the headspace of different coffee blends.

**Table 1. Volatile Compounds of Coffee Headspace and their Correlation Coefficients ($n = 5$)**

| | Compound | r | | | | |
|---|---|---|---|---|---|---|
| | | 'Juhlamokka' ground beans | | 'Presidentti' ground beans | | 'Espresso' ground |
| 1 | methanol | 0·66 | 0·75 | 0·75 | 0·83$^{a}$ | 0·91$^{b}$ |
| 2 | furan | 0·55 | 0·22 | 0·73 | −0·23 | −0·16 |
| 3 | carbon disulfide | 0·77 | 0·70 | −0·07 | 0·27 | |
| 4 | dimethyl sulfide | −0·83$^{a}$ | −0·30 | −0·36 | −0·34 | |
| 5 | propanal | −0·90$^{b}$ | −0·90$^{b}$ | −0·91$^{b}$ | −0·73 | −0·57 |
| 6 | acetone | 0·69 | −0·51 | 0·29 | −0·62 | 0·76 |
| 7 | methyl acetate | 0·86$^{a}$ | 0·64 | 0·04 | 0·40 | 0·71 |
| 8 | 2-methylpropanal | -0·02 | −0·25 | 0·09 | −0·34 | −0·78 |
| 9 | 2-methylfuran | 0·74 | −0·34 | 0·61 | −0·58 | −0·55 |
| 10 | 3-methylfuran | 0·80 | −0·36 | 0·61 | −0·45 | −0·58 |
| 11 | *n*-butanal | −0·96$^{c}$ | −0·84$^{a}$ | −0·91$^{b}$ | −0·40 | −1·00$^{d}$ |
| 12 | ethyl acetate | 0·11 | 0·72 | 0·29 | −0·67 | −0·73 |
| 13 | 2-butanone | −0·40 | 0·03 | 0·16 | −0·24 | −0·84$^{a}$ |
| 14 | butanedione | −0·82$^{a}$ | −0·93$^{b}$ | −0·95$^{b}$ | −0·92$^{b}$ | −0·87$^{a}$ |
| 15 | methyl propanoate | | | | | −0·48 |
| 16 | thiophene | 0·33 | −0·38 | 0·67 | 0·65 | 0·98$^{c}$ |
| 17 | 3-methylbutanal | 0·19 | −0·02 | −0·03 | −0·09 | −0·82$^{a}$ |
| 18 | 2-methylbutanal | 0·84$^{a}$ | 0·56 | 0·75 | 0·15 | 0·02 |
| 19 | 2,5-dimethylfuran | 0·90$^{b}$ | 0·44 | 0·68 | 0·16 | 0·59 |
| 20 | 3-methyl-2-butanone | | | | | −0·36 |
| 21 | phenol | 0·28 | −0·54 | 0·20 | −0·63 | 0·68 |
| 22 | 3-pentanone | | | | | 0·39 |
| 23 | 2,3-pentanedione | −0·26 | −0·92$^{b}$ | −0·76 | −0·93$^{b}$ | −0·15 |
| 24 | dimethyl disulfide | 0·66 | 0·28 | 0·41 | 0·15 | 0·63 |
| 25 | toluene | −0·26 | −0·70 | 0·30 | −0·22 | |
| 26 | acetic acid | −0·61 | 0·27 | −0·18 | 0·44 | |
| 27 | 1-methylpyrrole | 0·47 | 0·32 | 0·51 | 0·72 | |
| 28 | pyridine | −0·29 | 0·74 | −0·27 | −0·01 | 0·37 |
| 29 | 3-hydroxy-2-butanone | −0·44 | 0·35 | 0·13 | 0·22 | −0·05 |
| 30 | 2,4-dimethyl-3-pentanone | −0·22 | −0·74 | 0·68 | −0·77 | 0·81$^{a}$ |
| 31 | hydroxyphenol | 0·52 | 0·04 | 0·09 | −0·25 | 0·90$^{b}$ |
| 32 | 2-methylpyrazine | −0·61 | −0·10 | −0·28 | 0·54 | 0·19 |
| 33 | dihydro-2-methyl-3(2H)furanone | −0·28 | 0·32 | 0·23 | −0·03 | 0·89$^{b}$ |
| 34 | pyrrole | | | | | 0·50 |
| 35 | 2,5-furandione | −0·66 | −0·76 | −0·77 | −0·39 | |
| 36 | 2-furaldehyde | −0·87$^{a}$ | −0·90$^{b}$ | −0·75 | −0·28 | 0·53 |
| 37 | 2,5-dimethylpyrazine | −0·70 | 0·37 | 0·16 | 0·45 | 0·73 |
| 38 | 2,6-dimethylpyrazine | −0·58 | 0·77 | 0·21 | 0·59 | 0·86$^{a}$ |
| 39 | 2-furylmethanol | −0·56 | 0·64 | −0·18 | 0·36 | 0·43 |
| 40 | 2-butyl acetate | −0·63 | 0·69 | −0·37 | −0·06 | 0·53 |
| 41 | 2-acetylfuran | −0·78 | −0·28 | −0·08 | 0·00 | 0·70 |
| 42 | furfuryl acetate | −0·71 | −0·84$^{a}$ | −0·26 | 0·40 | 0·87$^{a}$ |
| 43 | 5-methyl-2-furaldehyde | −0·72 | 0·33 | −0·04 | 0·53 | 0·80 |

$^{a}$ $p < 0{\cdot}10$, $^{b}$ $p < 0{\cdot}05$, $^{c}$ $p < 0{\cdot}01$, $^{d}$ $p < 0{\cdot}001$.

3-methyl-2-butanone (20), 3-pentanone (22) and pyrrole (34) were always found in 'Espresso', but their contents were often below their detection thresholds in the 'Juhlamokka' and 'Presidentti' coffee samples. The other compounds identified were characteristic of all five coffee samples studied (ground 'Juhlamokka' and 'Presidentti', beans of 'Juhlamokka' and 'Presidentti', 'Espresso').

The proportions of several volatiles in 'Espresso' clearly differed from the other coffee blends. The most distinct examples are presented in Figs. 1 and 2. The results are in good agreement with the observations of Radtke-Granzer and Piringer (1981) and Piringer (1983) who found that Espresso contains large amounts of 2-methylfuran and small amounts of 2-methylpropanal, 3-methylbutanal and butanedione. Characteristic of dark roast coffee are the many heterocyclic compounds, such as 2-methylfuran, which originate

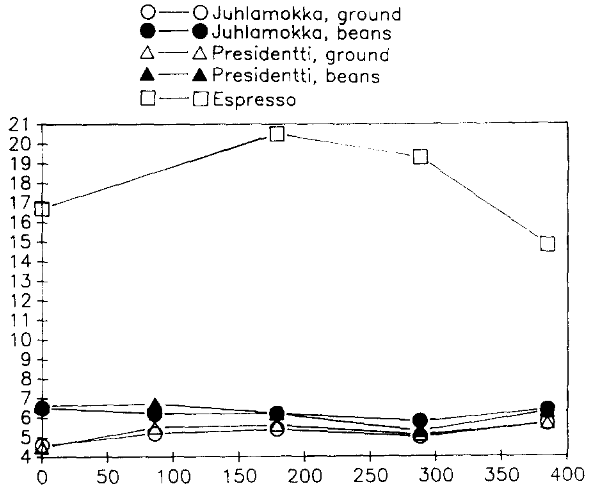

**Fig. 2.** The proportions, in percentage, of 2-methylfuran in the head-space of different coffee blends.

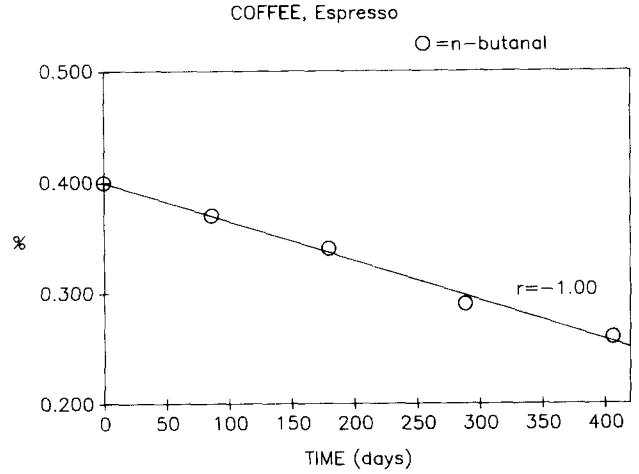

**Fig. 4.** The changes of proportions, in percentage, of $n$-butanal 'Espresso' coffee. $r$-Values are the linear correlation coefficients of the percentile shares versus actual age.

from sugar pyrolysis (Piringer, 1983). 2-Methylfuran was one of the major headspace compounds in 'Espresso', 3-methylfuran being a minor component. The shares of both compounds were clearly higher in 'Espresso' than in the other blends (Fig. 2).

In addition to the two most clear examples, i.e. 2- and 3-methylfuran, the contents of acetone (Fig. 3), methyl acetate, 2-butanone, 2,5-dimethylfuran and pyridine were also higher in 'Espresso' than in the other blends. Only traces of pyridine were found in 'Juhlamokka' and 'Presidentti', but its share in 'Espresso' was 5–10 times higher. On the other hand, methanol, 2,3-pentanedione, and 2-furaldehyde were found to be more abundant in 'Juhlamokka' and 'Presidentti' than in 'Espresso'.

Some differences between the ground coffee and coffee beans were also noticed. For example, Fig. 3 shows clearly that the level of acetone was higher in ground coffee than in the respective coffee beans. An analogous characteristic can be seen when observing the contents of 2-methylpropanal (Fig. 1). In fresh roast, the content of methanol was higher in ground than in whole beans, although its proportion in the latter increased during storage to the same level as in the ground coffee samples. On the other hand, a slight, though clear, difference between the whole and ground beans was the greater proportion of 3-methylbutanal in the whole beans.

In general the volatile compounds of ground 'Juhlamokka' and 'Presidentti' changed analogously during the one year of storage, as did the compounds of the whole beans of the same blends (Figs. 1, 2 and 3).

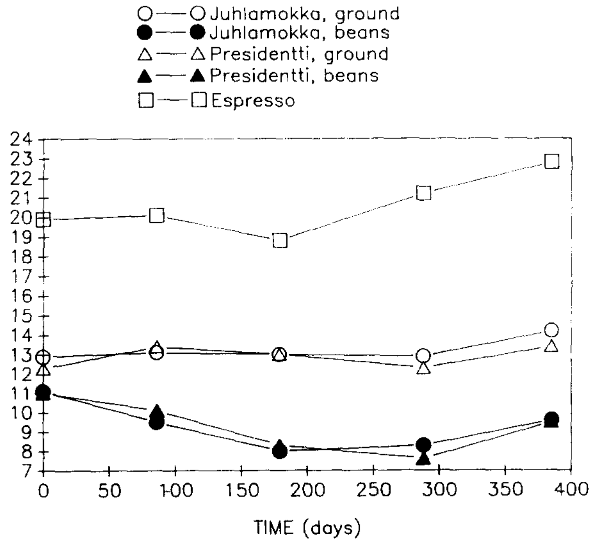

**Fig. 3.** The proportions, in percentage, of acetone in the headspace of different coffee blends.

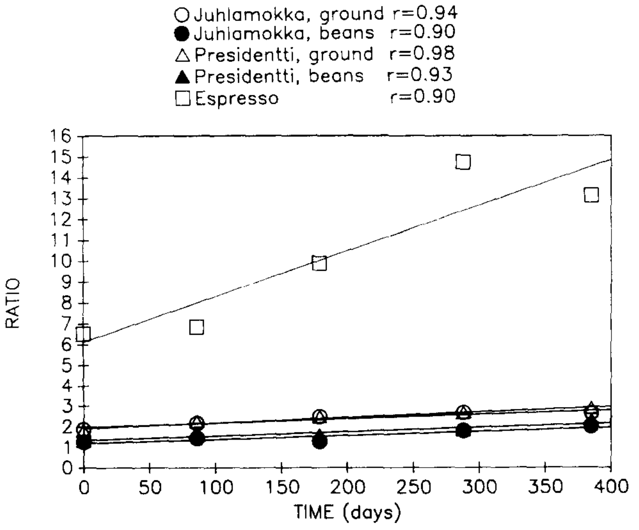

**Fig. 5.** The linear correlations acetone/butanedione versus actual age of different coffee blends.

**Table 2. Definitions of the Age Determination of Coffees ($n = 5$)**

| | 'Juhlamokka' | | | | | |
|---|---|---|---|---|---|---|
| | ground | | | beans | | |
| storage time | | $y = ax + b$ | | | $y = ax + b$ | |
| indicator, y | a | b | r | a | b | r |
| A/P | 0·0107 | 3·5441 | 0·97** | 0·0066 | 3·0871 | 0·96* |
| T/P | 0·0004 | 0·1411 | 0·87$^{0}$ | 0·0005 | 0·1842 | 0·88$^{0}$ |
| T/B | 0·0001 | 0·0763 | 0·70 | 0·0002 | 0·0685 | 0·83$^{0}$ |
| B/M | −0·0014 | 1·3720 | −0·87$^{0}$ | −0·0015 | 1·2805 | −0·97** |
| A/B | 0·0021 | 1·9626 | 0·94* | 0·0019 | 1·1864 | 0·90* |

| | 'Presidentti' | | | | | |
|---|---|---|---|---|---|---|
| | ground | | | beans | | |
| | a | b | r | a | b | r |
| A/P | 0·0116 | 3·2728 | 0·94* | (0·0079 | 4·3367 | 0·40) |
| T/P | 0·0005 | 0·1325 | 0·91* | (0·0008 | 0·2699 | 0·41) |
| T/B | 0·0001 | 0·0765 | 0·94* | 0·0003 | 0·0663 | 0·92* |
| B/M | −0·0016 | 1·3454 | −0·89* | −0·0013 | 1·1367 | −0·91* |
| A/B | 0·0027 | 1·9146 | 0·98** | 0·0021 | 1·3420 | 0·93* |

| | 'Espresso', ground | | |
|---|---|---|---|
| | a | b | r |
| A/P | 0·0114 | 10·162 | 0·74 |
| T/P | 0·0005 | 0·1888 | 0·88* |
| T/B | 0·0007 | 0·1132 | 0·90* |
| B/M | −0·0002 | 0·6174 | −0·79 |
| A/B | 0·0207 | 6·2612 | 0·90* |

A = acetone, P = propanal, T = thiophene, B = butanedione, M = 2-methylfuran
# actual storage time in days
$^{0}p < 0·10$, $^{*}p < 0·05$, $^{**}p < 0·01$

No marked differences between the blends 'Juhlamokka' and 'Presidentti' could be found with the headspace method applied.

Changes in roasted coffee during storage can be measured using selected volatile compounds as indicators, as shown earlier (Kallio *et al.*, 1990). Among those compounds the proportions of which changed during storage, the one best fulfilling the requirements of the linear regression were selected to be the indicators. The correlation coefficients listed in Table 1 were used as the criteria.

Some compounds, e.g. *n*-butanal (Fig. 4), changed in the same way in all coffee samples. Again, some could be used to measure either the changes of ground coffees (2,5-dimethylfuran, 2-methylbutanal) or coffee beans (2,3-pentanedione). Several compounds were useful indicators of the ageing of 'Espresso' coffee. For example, *n*-butanal, 2-butanone, 2,4-dimethyl-3-pentanone, 2-hydroxyphenol, and 5-methyl-2-furaldehyde showed good correlations, the correlation coefficient ($|r|$) varying from 0·8 to 1·00.

Staling of roasted coffee has been widely observed using 'aroma indices'; 2-methylfuran, 2-butanone and methanol are common indicator compounds (Arackal & Lehmann, 1979; Kwasny & Werkhoff, 1979; Vitzthum & Werkhoff, 1979). Several different ratios have been proved to be useful in measuring staling. Vitzthum and Werkhoff (1979) noted that the ratio 2-methylfuran/2-butanone decreased more slowly in beans than in ground coffee samples after opening the bag. In our experiments the samples were hermetically sealed, and the ratio of these compounds did not change during the storage period. The share of 2-methylfuran was, however, higher in beans than in the respective ground coffees. Kwasny and Werkhoff (1979) showed that the ratio 2-methylfuran/2-butanone decreased during storage in dark roasted coffee. The ratio 2-methylfuran/methanol is also commonly used as a measure of age (Vitzthum & Werkhoff, 1979). Neither ratios were good indicators of the staling of coffee in this work.

The ratio acetone (A)/butanedione (B) increased during storage. The content of acetone was higher, and butanedione lower, in 'Espresso' than in other samples. Thus, the ratio of these compounds was greater in 'Espresso' than in other coffees. The linear correlations A/B versus actual age are shown in Fig. 5.

The ratios acetone/propanal, thiophene/propanal, thiophene/butanedione, and butanedione/2-methylfuran were useful indicators of the ageing of coffee as was also observed in our earlier investigations (Kallio *et al.*, 1990). The ratio 2-methylfuran/propanal could not be used as a measure of the storage time of 'Espresso'. The change in the share of 2-methylfuran was not linear. Table 2 shows the linear correlations of the ratios versus actual age ($y = ax + b$, where $y$ is the ratio and $x$ the age), and the parameters $a$ and $b$. 2-methylfuran is formed by pyrolysis, and it has been shown that the ratio butanedione/2-methylfuran is a good indicator of age determinations (Piringer, 1983).

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