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A New Aroma Index to Determine the Aroma Quality of Roasted and Ground Coffee During Storage

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Summary

The staling of ground roasted coffee blend was studied over one year of storage. Solid phase microextraction-gas chromatography-mass spectrometry analysis was carried out to identify and semi-quantify 40 volatile compounds. We focused on volatile compounds related to the ageing of coffee: methanethiol, propanal, 2-methylfuran, 2-butanone, 2,3-butanedione, 2-furfurylthiol and hexanal. Changes in sensory properties of coffee beverage prepared from the ground roasted coffee samples during storage in laminate packages (PET/aluminium foil/PE) under air atmosphere, under nitrogen or in punctured packages were determined. Samples were stored at (23±2) °C and (40±10) % relative humidity. The reference coffee was packed similarly in laminate packages under nitrogen and stored at (–20±1) °C. Applying a statistical assessment of the results, we found the loss of most volatile compounds from all stored coffee samples, including even those stored under freezing conditions. The ratios of 2-methylfuran/2-butanone and methanethiol/hexanal proved useful indicators of staling. Our new proposed aroma index, the ratio of 2-furfurylthiol/hexanal, proved to be the most effective, giving high correlation coefficients (R≥0.80) with aroma freshness for coffee brews prepared from the ground roasted coffee stored under different conditions at room temperature.

Key words: roasted coffee, volatile compounds, modified atmosphere, aroma index, solid phase microextraction

Introduction

The quality of coffee is related to its flavour and aroma developed in chemical reactions during the process of roasting. Many factors affect the resulting quality, such as the origin of the coffee beans and time, temperature and degree of roasting. Reducing sugars and free amino acids are precursors of a great number of aroma compounds. Free amino acids react with reducing sugars in Maillard reactions or Strecker degradations, or the compounds are thermally degraded. The result is the pleasant smell arising from a freshly opened ground coffee package, which is an indicator of quality (*1,2*).

To date more than 850 volatile compounds are known, with a wide variety of functional groups (*3*). Different studies have demonstrated, however, that only a limited number of volatile compounds contributes to the characteristic flavour and aroma of ground coffee or coffee beverage. The identification of potent odourants by aroma extract dilution analysis (*4,5*) and gas chromatography of headspace samples (*6–8*), calculation of their odour activity values (*7,9*) and, finally, the reconstitution of coffee aroma (*10–12*) revealed 25 compounds for coffee beverage and 28 for ground coffee as the most active components of odour constituting the aroma of fresh coffee products. Among these compounds, 2-furfuryl-

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thiol was confirmed as the most outstanding odourant in the flavour of the beverage as well as in that of the powder.

Roasted coffee needs to be protected from outside influences by proper packaging. Coffee undergoes certain physical and chemical changes during storage which can lead to its rapid loss of quality. Changes in sensory properties of roasted coffee during storage are generally attributed to the loss and oxidation of odourants and to the development of oxidation reactions of coffee oil. Loss of freshness during storage, called staling, depends mostly on temperature, moisture and, especially, oxygen (*6*,*13*,*14*).

The ratios of several pairs of compounds present in coffee headspace have been observed to change with storage time, and these compounds could be used as indicators of product quality. The association of coffee staling with chemical changes has led to several simple chemicals being proposed as indicators of staling. Various authors (*15–17*) observed that the ratio of 2-methylfuran and 2-butanone (M/B), and the ratio of methanol and 2-methylfuran are related to freshness, showing good correlation with sensory analysis. Kallio *et al.* (*18*) and Leino *et al.* (*19*) observed that the ratio of several pairs of compounds present in coffee headspace changed during storage. The sum of the contents of several volatile compounds (2-methylpropanal, 3-methylbutanal, 2,3-butanedione, 2-methylfuran), termed the S-index, was found to decrease with the loss of quality of coffee during storage (*13*,*20*). Holscher and Steinhart (*6*) indicated methanethiol as a compound having a strong impact on aroma freshness, showing the largest decrease recognisable only one day after roasting. For this reason, Sanz *et al.* (*21*) followed the evolution of seven ratios of compounds including methanethiol. New aroma indices presented excellent linear correlation coefficients with the aroma freshness of coffee. Czerny and Schieberle (*22*) found that the staling of coffee is the result of degradation of important odourants, mostly 2-furfurylthiol. They proposed that 2-furfurylthiol can be used as an indicator of coffee staling. Unfortunately, all these indices are strictly related to the blend composition, roasting conditions and packaging technique, so that these parameters are unsuitable for absolute measurement of the degree of freshness of unknown roasted coffee blends.

Staling of roasted coffee during storage originates partly from oxygen absorption, while losses in coffee flavour have been shown to correlate with the exposure of roasted and ground coffee to air (*14*). Roasted coffee contains 10–15 % of lipids, 75 % of which are triglycerides, and these contain 40–55 % of linoleic acid (*23*). The product of autooxidation of linoleic acid is hexanal. The oxidation of coffee lipids occurs in two steps, first into hydroperoxides and then into volatile compounds, but to a smaller extent. As a result of oxidation reactions, hexanal increases depending on the exposure time of roasted and ground coffee to the air. Hexanal is not a potent odourant, but volatile secondary oxidation products develop oxidised off-notes in coffee when the positive coffee flavour compounds in roasted coffee start to decompose (*20*,*24*).

The aim of the present work is to study the influence of the access of oxygen to volatile compounds in roasted and ground coffee during storage. We evaluated seven ratios of volatile compounds and related their changes to the changes of sensory properties of coffee beverages.

Materials and Methods

Coffee samples and chemicals

A blend of Arabica and Robusta coffee species was roasted to a medium degree and ground to get a Turkish type of coffee (68–78 % of coffee particles are smaller than 0.25 mm in diameter). Immediately after grinding the coffee was packed in 100-gram laminate bags (PET/aluminium foil/PE) with air atmosphere, modified atmosphere with nitrogen or in punctured packages. At the moment of packaging the volume percentage of oxygen was below 1 % in packages with the modified atmosphere. The samples were stored up to one year at room temperature. The reference coffee was stored in laminate packages with modified atmosphere at –20 °C. Samples were analysed on the day that they were packed and after 14 days, 1, 2, 3, 4, 5, 6, 7 and 12 months of storage, in order to evaluate the loss of aroma freshness.

Pure reference standards of 2-butanone, acetaldehyde, propanal, 2-methylpropanal, 2,3-butanedione, ethylpyrazine and 2-furfurylthiol were purchased from Fluka (Neu-Ulm, Germany) and hexanal was obtained from Sigma (Steinheim, Germany).

Sample preparation

Just before analysis, 4 g of ground coffee were taken out of the bags, put into 20-mL headspace vials with the addition of 2.0 μL of internal standard (isobutylmethylketone) and closed immediately. All samples were prepared in triplicate from three different bags for each packaging technique. Solid phase microextraction (SPME) sample preparation was performed using a MultiPurpose Sampler MPS (Gerstel, Mülheim an der Ruhr, Germany), operated in SPME mode with a temperature-controlled heating module. The headspace vials were conditioned at a prewarming temperature of 70 °C for 20 min.

The next step was SPME, using a fibre with 85-μm Carboxen™/polydimethylsiloxane coating (Supelco, Bellefonte, PA, USA) for 30 min. The extraction temperature was the same as the prewarming temperature. Reproducibility of the method was checked by analysing six times the same sample of reference coffee that had been stored for one month.

Gas chromatography-mass spectrometry analysis

The components were separated by GC analysis on a Hewlett-Packard 6890 Series gas chromatograph (Agilent Technologies, Waldbronn, Germany) equipped with an injector, 0.75 mm i.d. liner (Supelco, Bellefonte, PA, USA) and mass-selective detector Hewlett-Packard 5971A (Palo Alto, CA, USA). The gas chromatograph was fitted with ZB-WAX capillary column, 60 m×0.32 mm×0.5 μm (Phenomenex, USA). Helium 6.0 was used as carrier gas with a flow rate of 1 mL/min at 40 °C. For thermal desorption, the SPME fibre remained in the injector for 5 min. The temperature of the injector was 270 °C. The oven temperature program was 5 min at 40 °C, from 40 to 220 °C at a rate of 4 °C/min, and 5 min at 220 °C. The mass selective detector was operated at 70 eV with electron impact ionisation. The transfer line was set to a temperature of 280 °C. Electron impact mass spectra were acquired in full-scan mode (30–300 *m/z*).

Identification of volatile compounds and quantitative measurements

Coffee volatile compounds were identified by comparing retention times with those of reference standards, and based on the comparison of their mass spectra with library spectra (*25*).

Headspace volatile compounds were semi-quantified by the addition of a known, weighed amount of internal standard (isobutyl methyl ketone) to each sample before analysis.

Aroma indices

Following the literature, several aroma indices were evaluated. Aroma indices are ratios of compounds used to determine possible statistical relationships to sensory results. Following our identified volatile compounds, we utilized several indices during the storage of coffee: 2-methylfuran/2-butanone, 2-methylfuran/2,3-butanedione, 2-methylfuran/propanal, methanethiol/2,3-butanedione, methanethiol/hexanal, methanethiol/2-butanone and our new proposed index: 2-furfurylthiol/hexanal.

Sensory analysis

Sensory analysis, conducted parallel with GC analysis, was carried out by cup tasting. Six trained assessors (4 male and 2 female) tasted the coffee brews to determine sensory discrimination between the ageing ground roasted coffee packed with different packaging techniques and the reference coffee. Coffee brews were prepared under strictly standardised conditions. A volume of 300 mL of boiling water was poured onto 21 g of roasted and ground coffee. The coffee brews were tasted after settling of the powder at 50 to 55 °C. Each sample was prepared immediately before tasting and served in white porcelain coffee cup labelled with a 3-digit code. Each assessor was served only one cup at a time. The order of presentation was randomised among assessors and sessions. Rinse water was provided for the assessors between individual samples. The assessors were asked to evaluate the odour and taste of coffee brews, with the emphasis on stale and rancid perception. Sensory results were obtained by compiling an evaluation sheet with a 0.0–5.0 scale. A score of 5.0 indicated a perception equal to fresh roasted and ground coffee, whereas lower scores corresponded to the loss in freshness quality. The limit of acceptability in terms of stale coffee was set at rating 3.0. Reproducibility of the sensory panel was checked by sensory analysis of three different samples in three repetitions served in random order.

Statistical analysis

All data were treated statistically with SAS (*26*) and GLM procedures (General Linear Models). Correlation coefficients between aroma indices and sensory results for all coffee brews prepared from ground roasted coffee stored at room temperature were evaluated with the CORR procedure. The coefficients of variation for all measurements, taken as the ratio of the standard deviation to the mean value, were lower than 7 % for all analyses.

Results and Discussion

For this reason 40 coffee volatile compounds were identified and semi-quantified by solid phase microextraction-gas chromatography-mass spectrometry analysis. The results show that solid phase microextraction sampling in the headspace of a sample is an effective and reproducible method for quantifying coffee volatile compounds. Of all the volatile compounds identified, only those that gave a better chromatographic response are shown in Table 1.

Among the 40 volatile compounds we identified 9 coffee odourants (methanethiol, acetaldehyde, propanal, 2-methylpropanal, 2-methylbutanal, 3-methylbutanal, 2,3-butanedione, 2-furfurylthiol and guaiacol) that contribute to the characteristic flavour of ground coffee or coffee beverage. According to Czerny *et al.* (*10*), these are some of the potent coffee odourants that give rise to the pleasant aroma of roasted coffee. The flavour of each odourant depends on its concentration. The final flavour of coffee depends on the interactions of odourants, like synergy or inhibition and suppression (*10,27*).

The influence of the presence of oxygen on the quality of roasted ground coffee was studied. Also, the influence of different packaging techniques on changes occurring during storage was evaluated. Different techniques resulted in different quantities of oxygen in contact with the roasted coffee. Ground roasted coffee was packed in an atmosphere modified with nitrogen, in air atmosphere or in punctured packages. The reference coffee was stored in laminate packages with modified atmosphere and stored under freezing conditions at (–20±1) °C.

The loss of most volatile compounds in all stored coffee samples, even those stored in the refrigerator (reference coffee), was found. Only concentrations of hexanal and 2,6-diethylpyrazine increased during storage in air atmosphere and in punctured packages, but their odours are not characteristic of stale coffee. The greatest, statistically significant ($p<0.05$), loss of volatile compounds was observed in coffee packed in punctured packages and the least in reference coffee. Comparison of the data showed that no off-flavour compounds were generated in coffee samples during storage, but the levels of 2-furfurylthiol, 2,3-butanedione and methanethiol, which are potent coffee odorants, were significantly ($p<0.05$) lower in coffee packed under air atmosphere than in coffee packed under modified atmosphere. Ethylacetate, phenol and 1-methylpyrrole were also identified as volatile compounds, having much lower concentrations in coffee packed under air than under modified atmosphere.

Many authors (*4,22*) have shown that products formed by oxidative degradation of unsaturated fatty acids in roasted coffee do not play a significant role in the flavour of roasted coffee. In our coffee samples the

Table 1. Identified volatile compounds in ground roasted coffee

Compound	Retention indices[d]	Compound	Retention indices[d]
Methanethiol[a,b]	688	4-Methylthiazole[a]	1312
Acetaldehyde[c]	712	3-Hydroxy-2-butanone[a]	1320
Propanal[b,c]	808	2,5-Dimethylpyrazine[a]	1357
Furan[a]	813	2,6-Dimethylpyrazine[a]	1362
2-Methylpropanal[c]	827	Ethylpyrazine[c]	1370
Methyl acetate[c]	843	2,3-Dimethylpyrazine[a]	1383
2-Methylfuran[a,b]	888	2-Ethyl-6-methylpyrazine[a]	1420
Ethylacetate[a]	904	2-Ethyl-5-methylpyrazine[a]	1428
2-Butanone[b,c]	918	2,6-Diethylpyrazine[a]	1469
2-Methylbutanal[a]	930	2-Furfurylthiol[b,c]	1472
3-Methylbutanal[a]	934	3-Ethyl-2,5-dimethylpyrazine[a]	1480
2,5-Dimethylfuran[a]	972	1,2-Ethanedioldiacetate[a]	1490
2,3-Butanedione[b,c]	1000	2-Furancarboxaldehyde[a]	1503
Phenol[a]	1096	2-Ethenyl-5-methylpyrazine[a]	1538
Hexanal[b,c]	1103	Furfurylacetate[a]	1566
2-Methylthiophene[a]	1148	5-Methyl-2-furancarboxaldehyde[a]	1620
1-Methylpyrrole[a]	1168	2-Furanmethanol[a]	1689
Pyridine[a]	1213	γ-Butyrolactone[a]	1695
Pyrazine[a]	1243	2-Thiophenecarboxaldehyde[a]	1754
Methylpyrazine[a]	1297	Guaiacol[a]	1911

[a]compound identified by mass spectra pattern
[b]compound chosen for shelf-life evaluation
[c]compound identified by comparing retention time and mass spectra pattern with those of standard compounds
[d]Kovats retention indices on ZB-WAX

only product of lipid oxidation identified was hexanal, whose concentration increased during storage. Based on the knowledge of coffee odourants, it can be assumed that the decrease of aroma quality during storage is correlated with disturbance of the balance of odourants. One reason for this is the loss of odourants by evaporation or by oxidation reactions (*13,14,20,22*). According to other authors (*6,13,22,28*), decreasing concentrations of 2-methylpropanal, 3-methylbutanal, 2,3-butanedione, 2-methylbutanal, methanethiol, and 2-furfurylthiol are responsible for changing the aroma and flavour quality of roasted coffee during storage. Like most of the volatile compounds, 2,3-butanedione also decreased during storage in the following order: reference coffee<modified atmosphere with nitrogen<air atmosphere<punctured packages (Fig. 1).

Sensory analyses were performed simultaneously to establish a relationship with the analytical data. The results of sensory analysis, focused on odour and taste of coffee brews with the emphasis on stale and rancid perception, indicate that the shelf life of ground roasted coffee packed with modified atmosphere was at least 1 year, for coffee packed with air atmosphere 5 months, and for coffee packed in punctured packages 3 months. The limit of acceptability in terms of stale coffee was set at a rating value of 3.0. Panellists detected some rancidity in the brew prepared from coffee packed in punctured packages after 4 months of storage, but clear rancidity was perceived only after 1 year of storage. Some rancidity was also detected in the brew prepared from coffee packed in air atmosphere after 4 months, which persisted until the end of storage. In the brew prepared from coffee packed under nitrogen, some rancidity was found, but only after one year of storage. No statistically significant differences (p>0.05) were found among sensory properties for reference coffee over one year (Fig. 2).

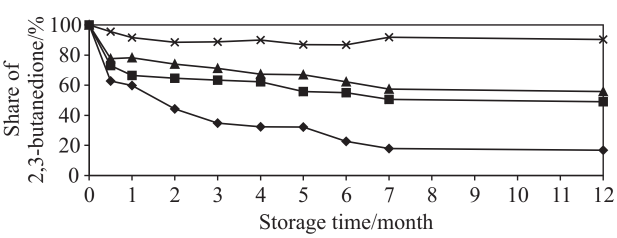

Fig. 1. Changes of proportion of 2,3-butanedione during storage of coffee (♦ punctured packages, ■ air atmosphere, ▲ modified atmosphere, × reference coffee)

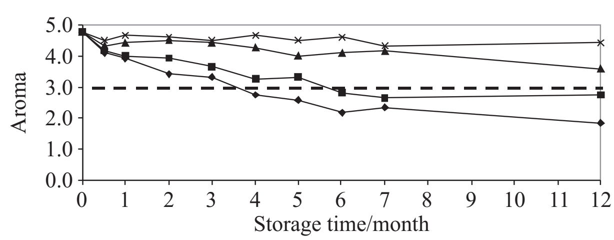

Fig. 2. Evolution of the aroma for coffee brews during storage (– – – limit of sensory acceptability, ♦ punctured packages, ■ air atmosphere, ▲ modified atmosphere, × reference coffee)

Sensory results are therefore in agreement with those from gas chromatography. The samples stored in a refrigerator preserved their aroma for a longer time, with decreased staling rate. We can therefore confirm the recommendation of Kallio *et al.* (*18*) that coffee stored in a freezer be used as an unaltered reference for staling studies.

The quality of roasted and ground coffee relates to changes in the chemical composition of the headspace vapour. Rather than resolving the entire secret of the chemistry of fine coffee aroma, a small number of volatile compounds were used as statistical indicators of quality. Following the literature, several indicators of freshness quality have been evaluated. On the basis of our identified volatile compounds, we evaluated the following: M/B (2-methylfuran/2-butanone), M/BD (2-methylfuran/2,3-butanedione), M/P (2-methylfuran/propanal), MT/BD (methanethiol/2,3-butanedione), MT/HE (methanethiol/hexanal), MT/B (methanethiol/2-butanone) and our new proposed index FT/HE (2-furfurylthiol/hexanal). Correlations were calculated between the aroma indices and the sensory results of odour.

The best known aroma index is the ratio of 2-methylfuran to 2-butanone concentrations, a decrease of which is related to staling (*15,16*). Intact packages exhibited a continuous but slow decrease, with a low correlation coefficient (R<0.70). This is in accordance with the results of Kallio *et al.* (*18*) and Leino *et al.* (*19*) where hermetically sealed samples did not change their aroma index over the storage period. Thus, the M/B ratio is not a useful index for hermetically sealed packages or for coffee without direct air contact. The ratio for punctured packages decreased sharply, compared to other samples, and showed very good correlation coefficients with aroma index (R=0.87) (Fig. 3). The decrease of M/B index takes place due to oxidation and loss of 2-methylfuran through diffusion. Similarly, the ratio M/BD proposed by Kallio *et al.* (*18*) gave a high correlation coefficient (R=0.80), but only with coffee packed in punctured packages.

Good correlation coefficients (R≥0.74) for aroma freshness for all coffee brews prepared from ground roasted coffee stored at room temperature were obtained for the ratio MT/HE proposed by Sanz *et al.* (*21*). Other indices had been calculated, such as M/P (*18*), MT/BD (*21*) and MT/B (*21*), but without any significant specific trend.

The most useful indicator of staling was the FT/HE ratio (Fig. 4). This new aroma index showed very good correlation coefficients with aroma freshness for all coffee brews prepared from ground roasted coffee stored at room temperature: 0.80 for coffee packed with modified atmosphere, 0.85 for coffee packed with air atmosphere and 0.83 for coffee packed in punctured packages.

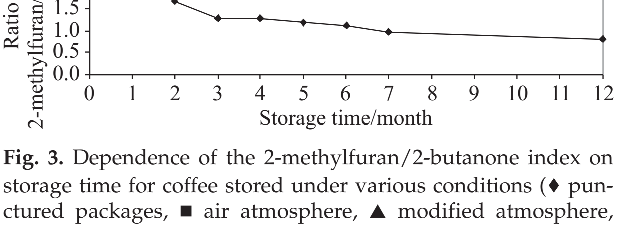

Fig. 3. Dependence of the 2-methylfuran/2-butanone index on storage time for coffee stored under various conditions (♦ punctured packages, ■ air atmosphere, ▲ modified atmosphere, × reference coffee)

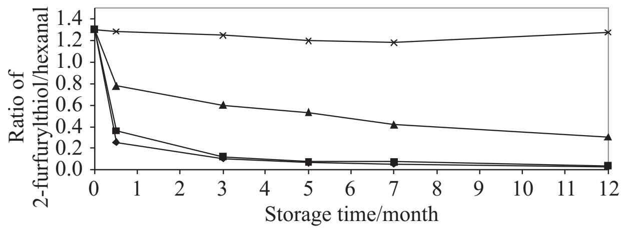

Fig. 4. Evolution of 2-furfurylthiol/hexanal index (FT/HE) during storage of coffee (♦ punctured packages, ■ air atmosphere, ▲ modified atmosphere, × reference coffee)

This index is very reliable, 2-furfurylthiol being considered as the key and most outstanding odourant of coffee (*10*). During storage the concentration of 2-furfurylthiol decreased by evaporation and oxidation. This process is accelerated in the absence of protection from the outside influences by proper packaging. On the other hand, the concentration of hexanal, which is a product of autoxidation of linoleic acid, increased during storage. For this reasons, it is an important indicator of inadequate storage and packaging for roasted coffee.

Conclusions

Although roasted coffee is a product with long shelf life, it is prone to some physical and chemical changes during storage that can lead to its rapid quality loss. Staling of roasted coffee during storage is generally attributed to the loss of volatile compounds and to development of oxidation reaction of coffee lipids. High quality ground coffee requires packaging systems that protect it from oxygen and moisture in order to preserve the initial coffee quality for a longer time.

With appropriate packaging materials and techniques we can largely counteract the factors affecting the quality of roasted coffee during storage, such as oxidation and evaporation of odourants. The results indicate that some packaging techniques can reduce the shelf life of roasted and ground coffee. Coffee packed in a modified atmosphere under nitrogen (gas flushing) has a longer shelf life than that packed under air. Flushing with nitrogen prevents reactions of coffee odourants and lipids with oxygen. Punctured packages admit oxygen and moisture during storage, reducing the quality of the product and decreasing its shelf life. For consumers, an alternative to preserving their roasted coffee in airtight packages is to store it in a refrigerator.

The staling of coffee was found to be the result of evaporation or degradation by oxidation of important coffee odourants, such as 2-methylpropanal, 3-methylbutanal, 2,3-butanedione, 2-methylbutanal, methanethiol and 2-furfurylthiol, rather than of formation of off-flavour compounds.

We observed that the ratio of several pairs of compounds present in coffee headspace increased or decreased with storage time and that these compounds could be used as indicators of product quality. Correlations between aroma indices and the sensory results of odour of coffee brews were calculated. The ratios of 2-methylfuran/2-butanone and methanethiol/hexanal were useful indicators of staling. However, the new aroma index, the ratio of 2-furfurylthiol/hexanal (FT/HE), proved to be more useful, due to high correlation coefficients ($R \geq 0.80$) for the aroma freshness for all coffee brews prepared from ground roasted coffee stored at room temperature.

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